Recovery of copper from Chilean mine waste waters

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Abstract

Two copper-containing waste-water streams from a Chilean mine site were identified as potential sources of revenue: Source 1 is raffinate from a copper solvent-extraction circuit; Source 2 originates from a reservoir receiving acid mine drainage as a result of natural snow melt. The streams are currently treated with lime to increase the pH for process and environmental purposes; any dissolved copper is precipitated, which renders it unrecoverable via the mine's conventional sulphide flotation process. These streams contain 80 to 240 mg/L dissolved copper that can be profitably recovered. An options analysis identified four potential technologies that may be applicable: BioteQ (biological or chemical precipitation of copper as the sulfide, which could then be processed through a nearby concentrator or smelter), EMEW (direct electrowinning of copper to produce high-purity saleable copper metal in the form of cylindrical tubes or metal powder), ELSA (a proprietary technology that produces a 95% copper sludge by cementation with iron), and ion exchange (a well-known technology that is widely applied in a variety of waste-water applications, particularly for the removal or recovery of low concentrations of valuable or toxic metals). This paper describes the ion-exchange concept and provides a preliminary process design, financial estimate, and economic feasibility assessment for the application of ion exchange to the recovery of copper from these two waste-water sources. Advantages of ion exchange compared with the other process options are discussed.

Key words: copper, acid mine drainage, mine waste water, ion exchange, economics

Introduction

Los Bronces, an Anglo American-owned mine located 65 km northeast of Santiago, Chile, at an altitude of 3500 m, processes copper sulphide ores. High-grade sulphides are subjected to comminution and then upgraded through a flotation circuit, following which concentrate is trucked to a nearby smelter for further processing. Low-grade run-of-mine material is accumulated in the valleys of the natural topography, where it is dumped-leached using drip irrigation with sulphuric acid. The resulting copper-containing pregnant leach solution (PLS) is treated for copper recovery by solvent extraction (SX). SX produces two streams: a purified concentrated rich electrolyte, from which copper is recovered by electrowinning (EW), and a copper-barren raffinate, a portion of which is limed for neutralisation. Sludge from the neutralisation process reports to tailings and clarified water is re-used for process purposes. The raffinate (~600 m³/h) still contains an average of 240 mg/L Cu prior to lime neutralisation, representing some 1220 t/a Cu.

A second waste source of copper is the mildly acidic water that is pumped from the mine pits, combines with snow melt and clarifier overflow from the raffinate neutralisation plant, and accumulates in a reservoir. This source has an average flowrate of 2000 m³/h (with seasonal fluctuations) and contains 80–180 mg/L Cu (up to 3060 t/a Cu). On eventual closure of the mine, this source will continue to generate acid mine drainage and acidic copper-containing liquors from snow melt off the leach dumps and has the potential to become an environmental liability if not treated.

These two waste-water sources represent a combined loss of some 4200 t/a Cu, with an estimated annual value of USD 21M, assuming a price of USD 5000/t for London Metal Exchange (LME) Grade A cathode. Recovery of the metal value from these wastes not only provides additional revenue to the

mine, but would reduce the volume of neutralisation sludge generated by 7000 dry t/a during the operational phase and limit long-term environmental impact and closure liabilities.

Various technologies were evaluated for application to these two waste waters. The objectives were to recover copper as a saleable product and minimize the amount of copper reporting as metal sludge to the tailings dam. The technologies considered included lime neutralisation, reverse osmosis, conventional SX–EW, sulphide precipitation, ELSA cementation, ion exchange (IX), and the EMEW process. This paper outlines the basis of the IX process, discusses reasons for its preference as the waste-treatment methodology of choice, and provides a preliminary business case for application to the recovery of copper from these two sources. Comparison with the alternative technologies is also provided.

Waste-Water Compositions

Table 1 presents the composition and physical characteristics of the two streams identified for potential copper recovery at Los Bronces.

Table 1 Composition of Los Bronces copper-containing waste streams (data from 2012).

Species	Source 1: SX raffinate Concentration (mg/L)	Source 2: Mine water reservoir Concentration (mg/L)
Al	1610	64.9
Ba	—	0.15
Ca	440	590
Cd	—	0.05
Cl	75	—
Co	8	0.55
Cu	240	80–180
Fe	260	65
K	10	34
Mg	1170	270
Mn	320	23
Na	30	69
Ni	4	0.4
P	30	<0.02
Zn	110	7.0
SO_4^{2-}	22 200	3380
NO_3^-	—	20.3
pH	1.6	3.4
TSS	40	110
Eh (mV)	600	—
Flowrate (m³/h)	580 ± 200	1940 ± 210
Temperature (ºC)	12–20 (avg. 17)	12–20 (avg. 17)

Application of Ion Exchange to the Recovery of Copper

IX is widely used in waste-water treatment, both for recovery of valuable metals and for removal of hazardous species before discharge. A resin with iminodiacetic acid functionality (Fig. 1) was selected for this application. These resins reject Ca and Mg strongly and are selective for copper over base-metal cations other than Fe(III): Fe(III) > Cu >> Ni > Zn ~ Co ~ Cd > Fe(II) > Mn > Ca > Mg > Na. Selectivity for copper arises from the chelating mechanism by which loading occurs. Copper loads at ~pH 3, at which pH most Fe(III) has been removed from solution by precipitation. Fe(III) loading is kinetically much slower than that of Cu(II) so discrimination against iron is possible when the residual amounts in solution are small. These resins are capable of removing Cu to <0.05 mg/L from both low ionic strength streams and strong electrolytes (Jurrius et al. 2014). Elution is carried out using 100 g/L H_2SO_4 and the resin is returned to the loading cycle in the H^+ form. The product stream is a high-purity electrolyte containing ~50 g/L Cu that is suitable for subsequent processing for metal recovery by electrowinning or crystallisation.
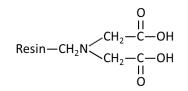
$$
\text{Resin}-\text{CH}_2\text{N}
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Figure 1 Iminodiacetic-acid resin functionality in the hydrogen form.

Process Description for Ion-Exchange Treatment of Los Bronces Waste Waters

A conventional fixed-bed ion-exchange (FBIX) configuration is proposed, using three columns in a lead–lag–elution configuration, as shown in Fig. 2. The operating conditions are given in Table 2.
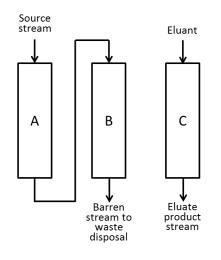

Figure 2 Lead–lag–elution fixed-bed ion-exchange configuration.

Feed solution is fed to Column A (lead column) and this outlet is fed to Column B (lag column). Lead Column A will reach capacity first when the resin becomes completely loaded with copper; lag Column B scavenges any residual copper that exits from Column A. Column A is then be taken off-line for elution. The feed is then directed to Column B, which now becomes the lead column, and eluted Column C becomes the lag column. The columns continue to rotate in this manner: there are always two columns on loading and one on elution.

Table 2 Process design specifications for copper removal from Los Bronces mining waste waters by cation exchange using an iminodiacetic-acid resin.

Parameter	Source 1	Source 2	Comment
Cu in feed (mg/L)	240	130	Can handle wide variability in feed [Cu]
Cu in barren (mg/L)	<10	<10	Can achieve <1 mg/L Cu if required
Cu in eluate product (g/L)	20–50	20–50	Eluate [Cu] can be tailored to suit selected downstream product-recovery operation
Cu recovery (%)	>95	>95	Can be readily increased to >99% as required with small impact on capital cost
Eluant	100 g/L H_2SO_4		
Feed flowrate (m³/h)	600	2000	
Product flowrate (m³/h)	2–10	7–17	Depending on [Cu] in product stream
Plant footprint (m²)	50–60	170–180	Estimated for 2.5 m diameter columns

Source 1 requires pH-adjustment from pH 1.6 to pH 3, which will also remove ~260 mg/L Fe present in the stream. Source 2 (pH 3.4) can be treated as received. Source 1 originates from an SX circuit so it may be contaminated by small amounts of organic phase, which is well known as a resin foulant. Installation of an upfront organic-removal system may be required, which will have the added benefit of returning recovered organic to the SX circuit and thereby lowering operating costs of that unit operation. A clarified feed solution (total suspended solids (TSS) <10 mg/L) is required for FBIX:

both sources can contain up to 100 mg/L TSS, so upfront clarification of the streams is required, in addition to making provision for regular removal of solids from the top of the resin bed by backwashing after elution.

The eluate product is a concentrated $CuSO_4$ solution, containing 40–50 g/L Cu in acidic media. It may contain small quantities (<5 mg/L) of Co, Ni, and Zn, as well as trace amounts of Ca and Mg. The electrolyte will, however, be of quality suitable to be fed to an electrowinning circuit for the production of LME Grade A cathode. The flowrates of the product streams are less than 20 m^3/h (and may be as low as 2 m^3/h) because of the high upgrading that can be achieved by IX. This makes the downstream capital requirements very small. This stream can be treated in several ways, and the composition (copper concentration and acidity) of the eluate can be readily adjusted to suit the requirements of the downstream operation. Options for product recovery include the following:

(i) Electrowinning: The preferred option at sites where adequate tankhouse capacity exists is to send the eluate directly to an existing conventional electrowinning process for copper recovery as cathode.

(ii) Crystallisation: Where an existing crystalliser exists with spare capacity, then crystallisation of a $CuSO_4 \cdot 5H_2O$ product represents a cost-effective option to treat the product stream.

(iii) EMEW: The eluate can be fed to a small compact EMEW circuit from which high-grade copper cathode can be produced. The size of unit required will be considerably smaller than for direct EMEW processing of the source streams. Furthermore, much higher recovery and current efficiency will be achieved because of the significantly higher copper concentration and elimination of Fe(III) from the electrolyte.

(iv) Sulphide precipitation: Copper can be precipitated as a saleable sulphide. This method of product recovery is least desirable because it requires both precipitation and smelting steps, and a relatively low-grade copper product is obtained.

Source 1 also offers the possibility of recovering cobalt and nickel using a two-step elution process, should their value warrant this: cobalt and nickel are less strongly bound on the resin so can be eluted first using dilute H_2SO_4 (~50 g/L), followed by copper elution using strong H_2SO_4 (80–120 g/L). As required, cobalt and nickel can be further separated and concentrated by IX, although, for this application, it would be preferable to precipitate a mixed hydroxide product for toll refining.

With respect to other streams generated in the IX process, the initial and final portions of the eluate stream will contain low concentrations of copper, but will still be high in acid so can be recycled to the leach circuit where the acid can be used and there is further opportunity to recover any contained copper: only the high-grade portion of the eluate proceeds forward as the product liquor. The barren stream exiting the IX loading cycle will have similar composition to the feed, except that copper concentrations will be reduced to 0.1–10 mg/L and most iron will have been removed in the upfront pH adjustment step for Source 1. Copper is be replaced by the hydrogen ion equivalent. The barren stream is suitable for discharge following liming to remove trace residual base metals.

Business Case for the Recovery of Copper from Mine Waste Waters by Ion Exchange

Table 3 lists the financial estimates for this process, based on the use of Lewatit TP 207 resin (Lanxess) and the operating conditions given in Table 2.

A typical standard IX column has a volume of 12 m^3 and diameter of 2.5 m. Using a lead–lag FBIX configuration and assuming a loading flowrate 10 BV/h, Source 1 will require five columns (2 × lead–lag plus one for elution), while Source 2 will require 16 columns (the elution cycle is very short compared with that of the loading cycle, so an addition elution column is not necessary). The columns will be piped in a manifold arrangement, such that any column can be in any position, allowing for optimum usage of the resin. If space constraints dictate, the column diameter can be increased up to 5 m, enabling fewer columns and a lower footprint to be achieved. This will have the advantage of lowering the capital cost. For these applications, increasing the column diameter from 2.5 m to 3.0 m reduces the capital cost by approximately 30% for both sources. There is no particular advantage or disadvantage of using larger diameter columns: the maximum size is usually limited by transport of the constructed columns to the site. The diameter:height ratio is typically designed at 1:2. The total resin volumes required for Sources 1 and 2 are 60 m^3 and 190 m^3, respectively.

Table 3 *Preliminary financial assessment of ion-exchange options for copper removal.*

Parameter	Source 1	Source 2	Comment
Cu in product (g/L)	40	40	Can be tailored to suit downstream operation
Cu recovery (%)	95	95	Can be increased to > 99.5% as required
Cu production (t/a)	930	2330	Depends on recovery targeted
Revenue (MUSD/a)*	5.19	9.4	Assumes that product recovery is by EW, producing LME Grade A copper quality
Operating cost (USD/t Cu)**	1,730	1,230	
Operating profit before deductions (USD/t Cu)	2,970	3,460	Dependent on Cu price
Capital cost (MUSD)***	5.3	13.0	Includes first-fill resin; can be reduced by using larger diameter columns
Payback period (month)	19	22	Depends on [Cu] in feed and flowrate

* Based on an LME copper price of USD 5000/t and a conservative 94% LME value paid for a cathode product.
** Based on labour, reagent, utility, and maintenance costs.
*** Excludes upstream organic- and TSS-removal steps that may be required and pumping and pipeline costs.

Capital costs are estimated from typical 2016 global equipment and materials costs, and based on construction of columns and tanks in rubber-lined mild steel with polypropylene piping. The plants can also be constructed in stainless steel, but capital costs will be higher. A basic programmable logic controller for automatic process control is included. This preliminary costing employs estimates of the costs of civils, plant housing, utilities, engineering design, and project management based on accepted percentages of main mechanical equipment (columns, tanks, pumps) costs for similar engineering projects. Capital costs exclude pipeline and pumping costs (to transport the feed from the raffinate pond or reservoir to the plant) and any provision for removal of entrained organic or TSS, should these prove to be necessary. Calculations are based on an estimate of 95% plant availability, which is readily attained for a plant of this nature. The downstream product-recovery process is excluded, as the selection of this unit operation will be site-specific.

Operating expenses are based on Chilean reagent, utilities, and labour costs (April 2016). One supervisor (USD/a 100,000) and six operators on shift rotation (USD/a 55,000 ea.) are required to operate the plant. Maintenance is estimated at 3% of capital equipment cost. The reagent costs and consumptions are given in Table 4. The lime requirement includes both pre-neutralisation of the feed ahead of the IX circuit (Source 1) and neutralisation of the barren stream for disposal. (Note: this lime requirement is currently required to neutralise iron and acid for direct disposal, so does not represent an additional cost to the existing circuit.) Resin replacement, based on a lifetime of five years, comprises 25–30% of the total operating costs.

Table 4 *Estimated reagent consumption and costs.*

Reagent	Cost (USD/t)	Consumption (t/t Cu)	
		Source 1	Source 2
Lime (85% CaO)	150	3.3	0.1
Sulphuric acid (98%)	80	1.6	1.6

In terms of sustainability, although both of these streams can be recycled for process purposes during the operational phase of the mine, Source 2 in particular (and likely run-off from other areas, including the leach dumps) will become a waste stream post-closure that will require treatment. Installing a copper-recovery process during the operational phase can generate ongoing revenue; post-closure, it would be fully paid off and can potentially continue to generate revenue while addressing ongoing environmental issues. A process that can provide flexibility in terms of flowrate throughput and wide variations in copper concentration is required.

Summary of the Applicability of Ion Exchange to Copper-Containing Mine Waste Waters

IX is a simple concept and the process is easy to operate and readily automated. It is possible to achieve very low Cu in the barren solution (< 0.1 mg/L), enabling this stream to meet legal discharge limits. The process is capable of handling fairly wide variations of copper concentration, flowrate, and temperature, provided the design criteria are appropriately specified. A relatively inexpensive iminodiacetic-acid resin can be used for this application and a product stream containing up to 50 g/L Cu is produced. The technology is very well known for copper removal, with more than 1000 applications to industrial and process wastes globally, so limited piloting is necessary, which allows for rapid implementation. Use of an FBIX configuration requires a small plant footprint.

Other factors that need to be considered with respect to the implementation of this technology include the potential needs to clarify source feeds to ensure TSS <20 mg/L (preferably <10 mg/L) and remove organic contamination from Source 1 to avoid poisoning of resin (<10 mg/L entrained organic). Copper ions in the waste water are exchanged for another ion that reports to the discharge (H^+ in this case). The product stream requires subsequent treatment by electrowinning, crystallisation, or precipitation/cementation to yield a saleable copper product. The preferred option will be dictated by the availability of existing tankhouse or crystallisation capacity already available on a site to limit capital expenditure requirements.

The project economics and payback period are strongly dependent on the prevailing copper price, but, even under the current conditions of commodity price pressure, the project has advantages in terms of sustainability and reducing longer-term mine closure liabilities.

Alternative Copper-Recovery Technologies

Several competing technologies for copper recovery from these wastes were also considered. Their advantages and limitations with respect to these two applications are outlined below.

Lime neutralisation: Lime (CaO) and limestone ($CaCO_3$) are commonly and widely used for treatment of acidic wastes. This approach is easy to engineer, simple to operate, can precipitate toxic metals, and is cost-effective for treating high volumes of waste waters. Metal species are precipitated as a hydroxide metal sludge, which is toxic if heavy metals are involved, requires storage, and presents long-term liabilities post-decommissioning. Stringent water quality requirements are not met. It is uneconomic to recover metal values from the sludge and copper losses would be high.

Reverse osmosis: This purification technique is used mainly in desalination applications. Because the water is processed through a semi-permeable membrane, high concentrations of dissolved metals and sulphates in waste waters cannot be handled. The brine waste produced will require separate treatment. Although strict water-quality criteria for discharge can be met for many applications that contain low concentrations of metal contaminants, this process is costly (both in terms of capital and operating costs), energy-intensive, and uneconomical for selectively recovering dissolved metals, especially when taking into account that further downstream processing would be required.

Solvent extraction–electrowinning: Although SX–EW is widely employed for primary recovery of copper (Schlesinger et al. 2011), it is generally considered to only be cost effective for feed concentrations above about 1 g/L Cu (Sole 2008). The SX process is equilibrium-driven, so has low extraction efficiency at low metal concentrations and low pH: raffinate concentrations can seldom be reduced below 100 mg/L Cu. The large organic inventory and electrical requirements of ~2 MWh/t Cu for electrowinning make this process capital- and energy-intensive.

Sulphide precipitation – BioteQ process: BioteQ's ChemSulphide® process involves the chemically assisted precipitation of copper as the sulphide (BioteQ 2016). The waste stream is fed into a precipitation reactor where the metal species reacts with NaSH. The precipitation slurry overflows to a clarifier, which provides the primary liquid–solid separation. A portion of the underflow is returned as seed material to the precipitation reactor; the remaining underflow is filtered and a copper sulphide cake is discharged. The process exchanges sodium (from NaSH) for copper, thereby increasing the sodium content of the discharge liquor. A biological option is also available, in which an upfront biological reactor generates H_2S gas from the metabolic reduction of elemental sulphur by anaerobic bacteria, which is used instead of NaSH as the precipitation agent.

The BioSulphide® system is more complex and capital-intensive than the ChemSulphide system and produces a product that is lower in copper content. Given the extreme monthly variations in ambient conditions at Los Bronces, the chemical option was considered to be more reliable and easier to control. The process is flexible with regard to flowrate and concentration fluctuations and gives selective and high recovery of copper over other base metals present. A disadvantage of the process is that the sulphide cake contains ~30% moisture, which will incur costs either for drying or for transportation of water: Chagres smelter is, however, relatively close to the Los Bronces site.

Several commercial case studies are available for zinc, cadmium, and nickel precipitation. For copper, in particular, the chemistry of the process has been proven for treatment of an acid mine drainage stream of very similar composition and flowrate at Dexing, China, which has been operating successfully since 2009 and has won a Chinese environmental award (BioteQ 2010).

Cementation – ELSAMetal process: ELSAMetal is a proprietary technology that produces a copper sludge by cementation with iron (ELSAmetal, 2016). The waste stream enters a reactor at its base, the sacrificial metal is fluidised, and the reactor is subjected to ultrasound to increase the effective surface area. The cementation product exiting at the top of the reactor contains ~95% Cu and is in the form of a powder. The process chemistry is well known and was commercially practiced for purification of copper streams by cementation with scrap iron for many decades before the advent of SX; however, it is designed for high concentrations of copper and the extent of reaction is expected to be limited for the low concentrations of this application. Downstream handling of residual iron in the cementation cake is required. Specific details of the technological innovations of this particular process are not available and there are no similar commercial operations for evaluation.

EMEW: EMEW® (Electrometals Technologies 2016a, 2016b) is an electrowinning process using a novel electrode configuration that enables efficient copper recovery from dilute solutions. The cathode comprises a stainless steel cylinder (approximately 25 mm in diameter), in the centre of which is an anode rod. Dimensionally stable anodes, made of titanium with precious-metal oxide coatings, are used. The overpotential for oxygen evolution on these materials is reduced by some 15% (Sandoval et al. 2010) when compared with conventional lead anodes, thereby lowering energy consumption. The cells are supplied in 30-cell modules. Electrolyte enters the cylinder tangentially at its base and flows in a spiral manner between the cathode and anode, exiting at the top of the cylinder. The very high flowrate of electrolyte through the cylindrical cell improves mass transfer, enabling electrodeposition to be carried out at high current density and from dilute solutions (1–20 g/L Cu). The technology claims to reduce residual copper in solution to <0.1 g/L. Copper can be deposited in powder form (for electrolytes containing <1 g/L Cu) or as cylindrical cathode tubes, weighing about 25 kg, which are periodically removed from the cells, compressed, packaged into bundles, and sold (Roux et al. 2007).

Although EMEW has been implemented at a wide range of sites for various applications, the low concentrations of the two source feeds for this application (~200 mg/L Cu) are of concern. It is likely that the current efficiencies will be low, especially because the iron content of the streams is similar to that of the copper (65–260 mg/L Fe). The technology is claimed to be suitable for feeds containing a minimum of 100 mg/L Cu, so morphology of the cathode deposit is likely to be powdery and non-adherent to the cathode blank, creating more difficult product handling. One potential advantage of EMEW technology is that it can be operated under outsourced conditions, with the operator taking a portion of revenues. This would provide a low-risk option, allowing the mine to focus on its core function of primary metal production, while still gaining some financial benefit from toll treatment of the waste streams.

Although EMEW is not particularly attractive for direct treatment of these waste waters, it does, however, represent a viable option for treatment of the IX product stream. The flowrate will be two to three orders of magnitude lower, allowing significant reduction in capital and operating costs, as well as plant footprint. Los Bronces currently has no conventional electrowinning or crystallisation capacity available (which would be the preferred product-recovery processes to treat the IX eluate), so inclusion of a small, modular EMEW circuit would provide a cost-effective means of product recovery when used in conjunction with IX.

Recommendations

The various technology options were evaluated based on technical capability, complexity, sustainability, and economic considerations. The BioTeQ ChemSulphide and combined IX–EMEW options were selected for further evaluation. Although the capital cost of the latter is some 40% higher for processing of Source 1 and 80% higher for Source 2, its annual operating costs are 50–55% lower. It should be noted, however, that some 80% of the capital cost of the combined IX–EMEW process is attributed to the EMEW component: if existing conventional tankhouse capacity is available on site, then the capital requirement for the IX component alone (Table 3) becomes extremely attractive.

The ChemSulphide process generates a filter cake containing 44–53% Cu and ~30% moisture, which requires further processing through a smelter; the IX–EMEW process generates a saleable copper cathode of LME Grade A quality. A concentrate containing 50% Cu will generally earn about 65–75% of LME copper value, while Grade A cathode earns 100% of LME value. Piloting of these options will enable more accurate operating performance and operating costs to be assessed, along with resilience of the process to changes in copper concentration, flowrate, and ambient temperature.

Conclusions

Ion exchange is demonstrated to be a straightforward, reliable, and relatively low-capital technology for the recovery of copper from mildly acidic mining waste streams, and can be profitably operated to create additional revenue for mine sites. It readily handles fluctuations in feed concentrations and flowrates, and can be optimised to consistently produce a low-volume, high-purity, high-concentration product stream from which LME Grade A copper cathode or other value-added copper chemicals can be produced.

Acknowledgements

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