A Desulfurization Flotation Approach for the Integrated Management of Sulfide Wastes and Acid Rock Drainage Risks

Jennifer Broadhurst and Susan Harrison
Minerals to Metals Initiative, Department of Chemical Engineering, University of Cape Town, South Africa

ABSTRACT

Wastes from the primary processing of hard-rock sulfide ores and coal have the potential to cause prolonged pollution of the local environment as a result of the long-term generation of acid rock drainage (ARD). Changes in legislation and global thinking have prompted a growing trend towards the development of approaches that remove the ARD pollution risks in perpetuity. In this regard, a two stage process has been developed at the University of Cape Town, which uses froth flotation to both recover valuable material and remove pyritic sulfur from fine mineral waste.

The technical feasibility of this process has been demonstrated at the laboratory scale for porphyry copper and fine coal wastes, with results indicating that it is possible to recover a useable product whilst generating a tailings waste stream with a negligible ARD risk. On the basis of this work, an order-of-magnitude financial model for a fictitious coal waste treatment plant has been developed and applied to demonstrate the economic viability for a selected case study. Furthermore, a life cycle assessment (LCA) study indicated that the pre-disposal removal of sulfide minerals from base metal tailings results in reduced eco-toxicity and human toxicity impacts. However, the LCA revealed that climate change and fossil fuel depletion impacts are increased due to additional energy consumption by the desulfurization process. This LCA study also highlighted the potential to reduce abiotic resource depletion through further processing and recycling of the separated tailings fractions.

The overarching objective of these studies, the key findings of which are presented in this paper, is to improve the environmental sustainability of coal and metal sulfide processing operations by both maximizing resource productivity and minimizing environmental burden.

Keywords: sulfide mine wastes, acid rock drainage, desulfurization flotation, economic feasibility, life cycle assessment

INTRODUCTION

The extraction and processing of coal and sulfide mineral deposits give rise to large volumes of solid and slurry wastes, which are conventionally disposed of in surface impoundments. These mine wastes frequently still contain residual sulfide minerals, particularly pyrite, which can result in the formation of acid rock drainage (ARD) upon exposure to air and moisture (Lottermoser, 2007). Acid rock drainage from sulfide mine waste deposits is typically characterized by low pH, high total dissolved solids and elevated levels of trace elements, and can cause prolonged degradation and pollution of local environments (see for example reports by Bell *et al*., 2001; Lottermoser, 2007; Rosner *et al*., 2001). Solid mine wastes also frequently represent a loss of valuable mineral resources due to incomplete separation during ore processing.

The long-term costs of on-going remedial action, coupled with increasing legislative and public pressure on mines to improve their environmental performance, has prompted a trend towards the development of preventative approaches that are aimed at minimizing or avoiding the generation of ARD (Johnson & Hallberg, 2005). Currently, such approaches focus mainly on avoiding exposure of sulfide minerals to oxygen and moisture, and involve techniques such as backfilling, blending and co-disposal, as well as the use of covers and seals (Johnson & Hallberg, 2005). However, these techniques do not completely remove the risk of ARD formation over the long-term, and in most cases their ability to provide a "walk-away" post-closure situation is yet to be proven. An alternative, and more sustainable, approach is to selectively remove the sulfide minerals from the waste prior to disposal, thereby effectively eliminating the risk of ARD formation in perpetuity (Cilliers, 2006; McCallum & Bruckard, 2009). Previous studies have shown froth flotation to be a technically feasible and cost-effective technique for removing sulfide minerals from a number of base metal and gold process tailings (Benzaazoua *et al*., 2000 & 2008; Benzaazoua & Kongolo, 2003, Bois *et al*., 2004 and Leppinen *et al*., 1997). The conventional desulfurization process produces two mineral waste fractions: a large volume sulfide-lean fraction which can be land disposed without posing an ARD risk, and a smaller volume sulfide-rich fraction which can be used as backfill (Benzaazoua & Kongolo, 2003) or processed further into useful by-products such as sulfuric acid.

Ideally, the pre-disposal treatment of sulfide mine tailings should be used in an integrated approach that not only removes risks of ARD pollution, but also provides opportunities for value recovery and re-allocation of unavoidable wastes as feedstock for other uses (Benzaazoua *et al*., 2008; Hesketh *et al*., 2010a). In this regard, studies at the University of Cape Town have demonstrated that desulfurization flotation of a porphyry-type copper sulfide tailings can be conducted in stages through manipulation of the reagent regime, producing an additional metal-rich stream that could be recycled to the conventional metal recovery plant (Hesketh *et al*., 2010a). The concept of using froth flotation to both recover value and remove ARD risks from mine waste has subsequently been extended to fine coal wastes in a two-stage process that recovers coal by means of oily collectors in stage 1, followed by desulfurization flotation to remove pyritic sulfur in stage 2 (Amaral Filho *et al*., 2011; Kazadi Mbamba *et al*., 2012). Selected case studies have also been conducted to explore the economic viability (Jera, 2013) and broader environmental implications (Broadhurst *et al*., 2014) of the proposed desulfurization flotation approaches.

This paper presents the key findings of these studies, with a view to demonstrating the techno-economic and environmental feasibility of using froth flotation to both recover valuable material and remove ARD risks from fine metal sulfide and coal processing wastes.

METHODOLOGY

Laboratory-scale flotation tests

All flotation tests were carried out in a 3-liter modified Leeds batch flotation cell at the natural pH of the slurry in tap water (pH 6-8), an impeller speed of 1200 rpm and an aeration rate of 5-6 L/min. Detailed experimental procedures are outlined by Hesketh *et al*. (2010a) for the porphyry copper case study and by Kazadi Mbamba *et al*. (2012) for the coal case studies. Acid generating potential was determined using standard acid-base accounting (ABA) and net acid generation (NAG) static tests (Stewart *et al*., 2006), as well as the batch biokinetic test developed at the University of Cape Town (Hesketh *et al*., 2010b)

Porphyry copper case study

In this case study, an oxidized low-grade porphyry copper ore, milled to 70 % passing 150 μm, was used as a proxy for a typical porphyry-type copper sulfide tailings. The ore had a sulfur and copper grade of 3.84 % and 0.16 %, respectively. Pyrite (7.03 %) and chalcopyrite (0.12 %) were the major sulfide minerals. Flotation was carried out in stages (maximum 4), using methyl isobutyl carbinol (MIBC) frother (30 g/ton) and a diothiophosphate collector from CYTEC mining chemicals, at a dosage rate of 9 kg/ton for each stage. The retention time for stage 1 was 8 minutes, and thereafter 5 minutes for each stage. Solids were analyzed for iron, copper and total sulfur.

Coal waste case studies

Two-stage laboratory-scale flotation tests were carried out on the following coal waste samples: fine thickener underflow slurry waste from an operating colliery in Middleburg, South Africa-filtered and milled to 75 % passing 150 μm; fine flotation tailings waste from an operating colliery in Criciúma, Brazil; coal discard, generated through the destoning of low-grade coal by means of an experimental X-ray sorter at a South African coal-fired power plant, and subsequently crushed and milled to 75 % passing 106 μm.

Dodecane (1.86 kg/t) or oleic acid (2.89 kg/t) were used as coal collectors in stage 1, together with isobutyl carbinol (0.11-0.28 kg/t) as a frother. Desulfurization flotation of the coal flotation tailings was subsequently carried out using potassium amyl xanthate (PAX) as a sulfide collector (2.33 kg/t), dextrin as a coal depressant (0.93 kg/t) and methyl isobutyl carbinol (MIBC) as a frother (0.11 kg/t). All flotation tests were carried out at a solids content of 6-7 %, and collection periods of 5 minute and 20 minutes for the coal and desulfurization flotation stages, respectively. Samples were analyzed for ash and total sulfur content.

Economic assessment: coal case study

A framework was developed for providing an order-of-magnitude cost estimate (± 30 % to ± 50 % accuracy) for a fictitious two-stage coal flotation desulfurization plant to treat nominal coal fines from a dump of an abandoned mine in the Witbank/Middleburg coal field (Jera, 2013). The plant flow sheet was based on the laboratory-scale test results obtained for the South African fine coal waste, as described in the previous section, and included milling, flotation, filtration, thickening and pumping as key operations. A plant throughput of 100 t/h with a capacity of 720,000 tons per annum was assumed, at a plant life and availability of 15 years and 82 % respectively. Capital costs were based on major equipment costs in combination with Lang costing factors. Operating costs were based on "rule of thumb" industry estimates and flow sheet data. Revenue was generated from the sale of the coal product at a price of 349 South African rands (R) per ton. The net present value (NPV) and internal rate of return (IRR) were calculated at a discount rate of 14 %, using Microsoft Excel.

Life cycle assessment: base metal case study

Two scenarios for the treatment of a base metal sulfide tailings stream were developed to evaluate the environmental consequences of incorporating a desulfurization flotation unit into a conventional tailings management plant, which entails dewatering in a thickener and disposal of the thickener underflow to a tailings impoundment (base case scenario). Input-output data for the two tailings management options were derived from a combination of literature information, in-house knowledge and mass balance calculations. An empirical base metals case study by Benzaazoua & Kongolo (2003) formed the basis of this study (Table 1), producing two output streams: a non-acid generating sulfide-lean tailings comprising 72% of the feed; and a sulfide-rich concentrate which could be recycled to the primary extraction plant for additional metal and/or acid recovery.

Background data for electricity production were derived from the Eco-invent database (version 2.2), whilst data for the production of xanthate were obtained from a recent in-house study of the local production of liquid ethyl xanthate at Senmin® International in Sasolburg, South Africa (Kunene *et al*., 2013). Life cycle assessment modelling for each of the scenarios was conducted at a reference flow rate of 100 tons of dry tailings per day, using Simapro Software (version 7.3.3) and USEtox (eco-toxicity and human toxicity impacts) and ReCiPe (climate change, fossil fuel depletion, terrestrial acidification, urban land occupation and natural land transformation) impact assessment methodologies. The detailed methodology and case study assumptions are outlined in Broadhurst *et al*. (2014).

Table 1 Desulfurization flotation LCA case study data[1] (after Benzaazoua & Kongolo, 2003)

Component	Stream compositions (mass %)			Recovery to concentrate (% of feed)
	Feed tailings	Desulfurized tailings	Sulfide-rich concentrate	
Pyrite	17.4	2.18	56.53	91[2]
Sphalerite	0.19	0.02	0.62	90
Chalcopyrite	0.10	0.07	0.19	61
Calcite	2.70	3.38	0.96	10[3]
Other (gangue)	79.61	94.36	41.67	15[2]
Sulfide sulfur	9.38	1.17	30.49	91
Total solids	100	100	100	28

[1]Desulfurisation carried out at pH 6-11, using KAX (potassium propyl xanthate) as a collector and Sasfroth Sc 39 as a frother (Benzaazoua & Kongolo, 2003); [2]derived from mass balance calculations; [3]assumed on the basis of literature information (Broadhurst *et al*., 2007).

RESULTS AND DISCUSSION

Laboratory-scale flotation tests: porphyry copper case study

Flotation tests on the porphyry copper sulfide sample indicated that, whilst the recovery of both pyrite and chalcopyrite increased on increasing the dosage of collector from 9 g/t to 27 g/t, the effect was more significant in the case of pyrite. This differential effect of collector dosage created the opportunity to selectively recover copper in a multi-stage process using 9 kg/t collector in stage one and an additional 27 kg/t collector in subsequent stages. This approach resulted in three tailings fractions: a low volume value-rich fraction containing 58 % of the copper; a low volume sulfide-rich fraction containing > 85 % of the sulfur; and a large volume sulfide-lean fraction containing ~ 90 % of the total mass and < 6% of the total sulfur (Figure 2 and Table 2).

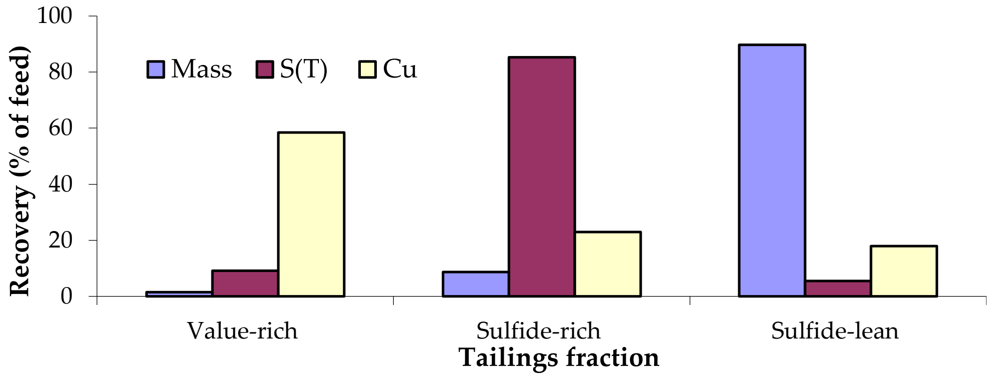

Figure 2 Recovery to tailings fractions in the multi-stage desulfurization of porphyry copper

Table 2 Flotation test results for the multi-stage desulfurization of porphyry copper

Stream	Content (mass %)		NAPP	ARD classification[1]
	Total sulfur	Copper	(kg/t H_2SO_4)	
Feed	3.8	0.16	97.4	Acid forming
Value-rich fraction	2.1	5.7	n/d	n/d
Sulfide-rich fraction	33.7	0.39	n/d	n/d
Sulfide-lean fraction	0.21	0.03	-19.2	Acid neutralizing

[1]In accordance with standard static and biokinetic test results (Hesketh *et al.*, 2010b)
Where: NAPP is net acid producing potential determined by the standard Acid Base Accounting (ABA) method, and n/d is not determined

Laboratory scale ARD prediction tests, using both acid base accounting (ABA) and net acid generating (NAG) protocols, indicated that the sulfide-lean tailings fraction, with a sulfur content of 0.2 %, can be classified as non-acid forming (Hesketh *et al.*, 2010a). Subsequent biokinetic tests confirmed that this sample was net-acid neutralizing, even under conditions of microbial activity (Hesketh *et al.*, 2010b).

Laboratory-scale flotation tests: coal waste case studies

Experimental results for the two-stage flotation process are presented in Table 3 and Figure 3.

Table 3 Flotation test results for the coal waste samples

	A: Tests usingdecane as a collector in stage 1							
	South African fine coal slurry[1]				**Brazilian coal tailings**			
	Feed	**Coal product**	**Sulfide-rich**	**Sulfide-lean**	**Feed**	**Coal product**	**Sulfide-rich**	**Sulfide-lean**
Yield (mass %)	100	20	13	67	100	35	43	22
Ash (mass %)	34.4	13.5	28.9	40.8	56.4	24.7	70.4	87.2
Sulfur (mass %)	1.1	0.5	2.7	0.4	5.1	1.6	1.8	0.5
NAPP (kg/t H_2SO_4)	3	n/d	65	-45	-7	n/d	27	-23
ARD classification[2]	PAF	n/d	AF	NAF	PAF	n/d	AF	NAF
	B: Tests using oleic acid as a collector in stage 1							
	South African coal discards				**Brazilian coal tailing**			
	Feed	**Coal product**	**Sulfide-rich**	**Sulfide-lean**	**Feed**	**Coal product**	**Sulfide-rich**	**Sulfide-lean**
Yield (mass %)	100	41	23	36	100	56	24	46
Ash (mass %)	56.4	32.1	53.8	82.4	56.4	42.2	85.3	88.8
Sulfur (mass %)	5.1	3.0	18.7	0.2	5.1	1.6	1.4	0.6
NAPP (kg/t H_2SO_4)	110	n/d	501	-70	-7	n/d	15	-67
ARD classification[1]	AF	n/d	AF	NAF	PAF	n/d	PAF	NAF

[1]After Kazadi Mbamba *et al.* (2012) [2]In accordance with standard static and biokinetic test results
Where: NAPP is net acid producing potential, PAF is potentially acid forming, AF is acid forming, UC is uncertain, NAF is non-acid forming, n/d is not determined.

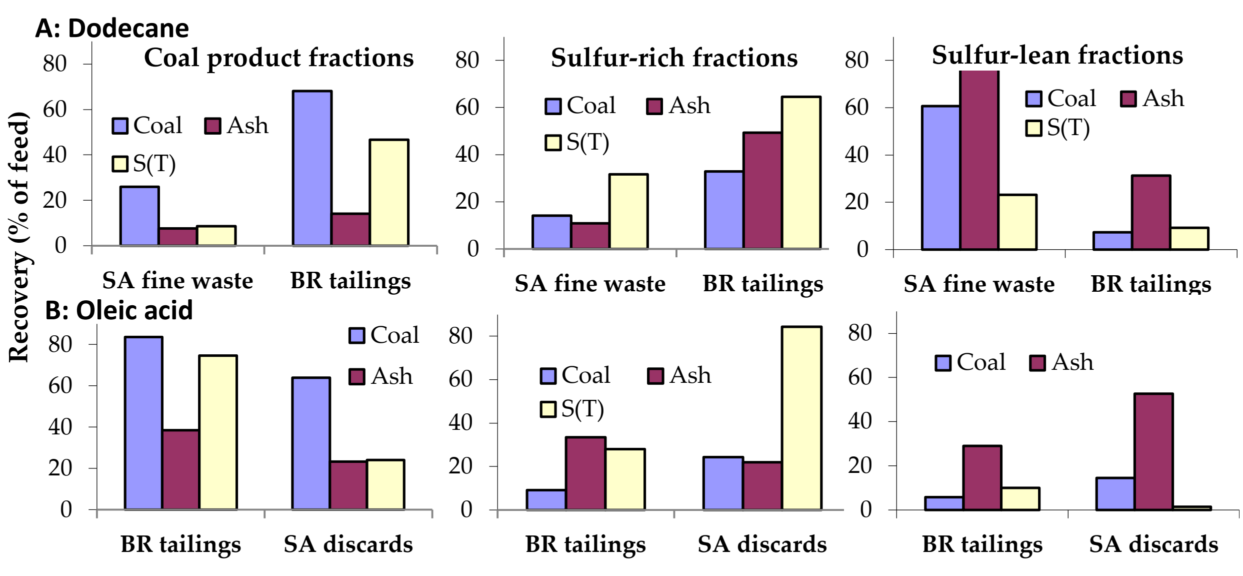

Figure 3 Deportment of key coal components to the flotation output streams, using dodecane (A) and oleic acid (B) as coal collectors (stage 1), and PAX as a sulfide collector (stage 2)

The results indicate that the relative deportment of key components (coal, ash and sulfur) to the flotation output streams varies quite significantly for different coal wastes. This variability can probably be attributed largely to differences in the mineralogical, petrographic and surface characteristics of the coal wastes (Kazadi Mbamba *et al*., 2012). The type of collector also appears to have a significant effect on coal yields in stage 1, with oleic acid resulting in higher recoveries of coal from the Brazilian coal tailings than dodecane. A similar trend has been observed for other coal wastes, and has been attributed to the stronger interaction between the hydrophobic component of the coal surface and the carbon-oxygen double bond of the oleic acid molecule (Kazadi Mbamba *et al*., 2013). Performance variability notwithstanding, in all case studies the two-stage flotation process resulted in a coal product with reduced ash and sulfur contents, and a final tailings fraction with reduced sulfide content and negligible ARD risk potentials.

Economic assessment: coal case study

The results of the order-of-magnitude economic assessment, for the specific coal waste case study described in the methodology section, are summarized in Table 4.

Table 4 Economic assessment results: fine coal waste case study

Item	Unit	Amount	Comment
Capital costs	R 000 000	198	Installed capital costs based on a combination of supplier estimates for major equipment and Lang factors
Operating costs	R 000 000 per annum	161	Based on a combination of flow sheet data and "rule-of-thumb" estimates from the industry and a contingency of 20%
Revenue	R 000 000 per annum	201	Based on a sales price of ZAR349/t
Project NPV	R 000 000	50	Calculated at a discount rate of 14% and based on earnings
Internal rate of return	%	19	before interest, taxes, depreciation and amortization

Although the financial model is based on one case study only, the results in Table 4 are indicative of the potential economic viability of the two-stage flotation process for the recovery of coal and removal of sulfur from coal wastes. Reagent costs were relatively high, contributing 54 % to total operating costs (Jera, 2013). The net present value (NPV) was found to be particularly sensitive to coal sales price, coal yield and reagents costs (Jera, 2013).

Life cycle assessment: base metal case study

The life cycle inventory (LCI) results for the conventional and desulfurization flotation treatment scenarios are compared in Figures 4A and B.

The desulfurization flotation process scenario consumes more carbon than the conventional tailings treatment scenario (base case), and results in higher emissions of carbon dioxide and sulfur dioxide, due largely to the higher consumption of fossil-fuel based electricity in the flotation circuit (Broadhurst *et al*., 2014). However it also results in a decrease in aqueous emissions, whilst simultaneously enhancing the potential to recover water and metal values from the tailings.

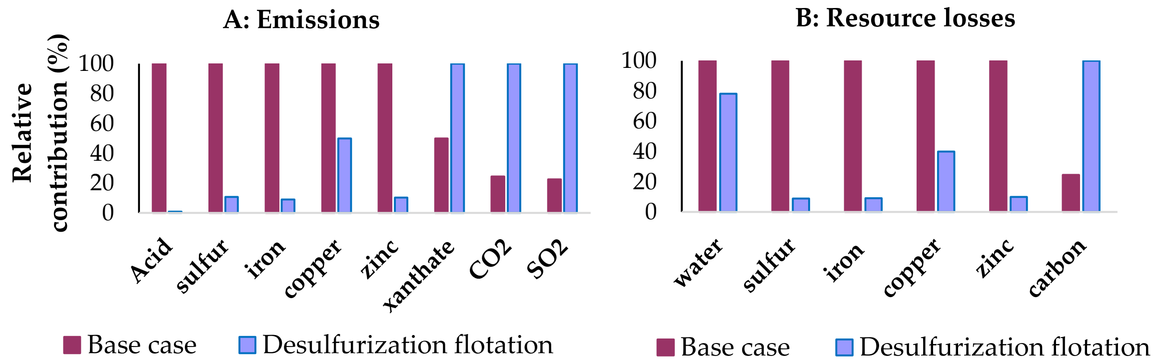

Figure 4 Relative environmental emissions (A) and resource losses (B) for the two tailings treatment scenarios (Broadhurst *et al*., 2014)

The results of the subsequent life cycle impact assessment (LCIA) modelling (Figure 5) indicate that the desulfurization flotation scenario results in higher climate change, fossil fuel depletion and terrestrial acidification impacts than the conventional treatment scenario, which can once again be attributed largely to the higher consumption of fossil-fuel based electricity in the flotation circuit (Broadhurst *et al*., 2014). Desulfurization flotation also results in a considerable reduction (more than 80 %) in both eco-toxicity and human toxicity impacts, due largely to a decrease in the amount of zinc that deports to the tailings and is thus available for subsequent release into the environment (Broadhurst *et al*., 2014). Incorporation of a desulfurization flotation unit into the conventional (base case) tailings treatment circuit also results in lower urban land occupation and natural land transformation impacts than the base case scenario, due to the smaller volumes of land-disposed tailings.

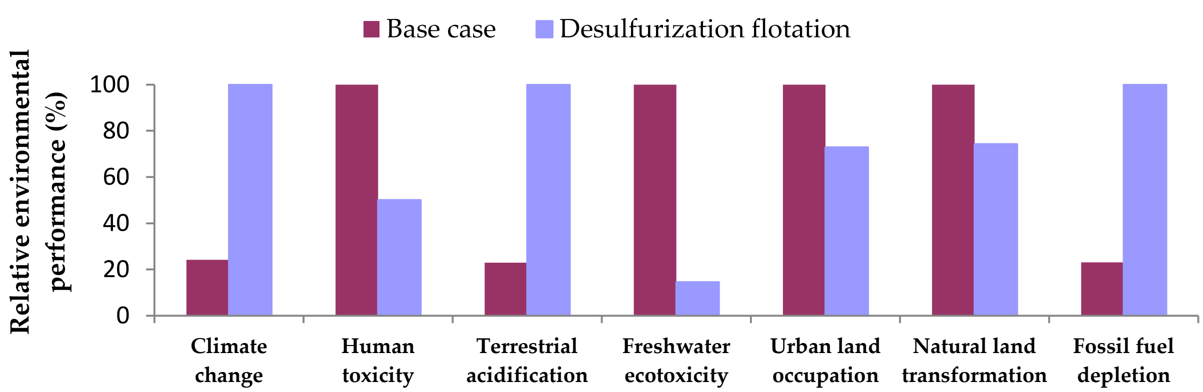

Figure 5 Relative environmental performance for the two tailings treatment scenarios

The study also highlighted the deficiencies of current LCIA tools, in terms of their ability to adequately assess the environmental impacts associated with solid mineral wastes and their management. This pertains, in particular, to aqueous acidification, salinization and trace metal impacts (Broadhurst *et al*., 2014).

CONCLUSION

The technical feasibility of using froth flotation in a two-stage process to both recover valuable material and remove sulfur from mine waste has been demonstrated at a laboratory-scale for the case of porphyry-copper and a range of different coal processing wastes. All case studies resulted in a sulfide-lean tailings stream, which is non-acid generating, thus effectively eliminating long-term ARD risks. Furthermore, a preliminary economic assessment showed the potential viability of the desulfurization flotation treatment process when combined with value recovery. A holistic environmental study using life cycle assessment tools highlighted the environmental benefits of the desulfurization flotation tailings management approach in terms of reduced aqueous emissions and waste burden as well as enhanced resource recovery. However, the flotation process also consumes additional electricity, which results in higher climate change, fossil fuel depletion and acidification impacts in cases where the production of electricity is coal-based.

Studies are in progress to identify and evaluate downstream management options for the sulfide-rich and sulfide-lean phases, with a view to further reducing waste burden and maximizing utilization of mineral resources. Further research and development is also currently underway to optimize the two-stage process in terms of key performance variables, particularly electricity, reagent consumption and waste characteristics, and to extend the techno-economic-environmental studies to a range of sulfidic mine wastes. These studies will form the basis for future publications.

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