

Re-using bauxite residues: benefits beyond (critical raw) material recovery

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Abstract

Since the world economy has been confronted with an increasing risk of supply shortages of critical raw materials (CRMs), there has been a major interest in identifying alternative secondary sources of CRMs. Bauxite residues from alumina production are available at a multi-million tonnes scale worldwide. So far, attempts have been made to find alternative re-use applications for bauxite residues, for instance in cement / pig iron production. However, bauxite residues also constitute an untapped secondary source of CRMs. Depending on their geological origin and processing protocol, bauxite residues can contain considerable amounts of valuable elements. The obvious primary consideration for CRM recovery from such residues is the economic value of the materials contained. However, there are further benefits from re-use of bauxite residues in general, and from CRM recovery in particular. These go beyond monetary values (e.g. reduced investment / operational costs resulting from savings in disposal). For instance, benefits for the environment and health can be achieved by abatement of tailing storage as well as by reduction of emissions from conventional primary mining. Whereas certain tools (e.g. life-cycle analysis) can be used to quantify the latter, other benefits (in particular sustained social and technological development) are harder to quantify. This review evaluates strategies of bauxite residue re-use / recycling and identifies associated benefits beyond elemental recovery. Furthermore, methodologies to translate risks and benefits into quantifiable data are discussed. Ultimately, such quantitative data are a prerequisite for facilitating decision-making regarding bauxite residue re-use / recycling and a stepping stone towards developing a zero-waste alumina production process.

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INTRODUCTION

Critical raw materials (CRMs) are fundamental to Europe's economy, growth and jobs and they are essential for maintaining and improving our quality of life.¹ This statement by EU's Raw Materials Initiative recognizes factual issues of insecure future raw materials within the EU¹ and is reflected by many major economies (e.g. US National Research Council,² Kawamoto,³ MineralsUK⁴ and Natural Resources Canada⁵). Within the framework of the European Innovation Partnership on Raw Materials, a list of 27 CRMs was derived, including platinum group metals (PGM) and rare earth elements (REE).⁶ REE are divided into light (LREE; La, Ce, Pr, Nd, Sm) and heavy (HREE; Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, including Y) plus Sc.^{7,8} The new 2017 list includes nine additional materials (e.g. V and Sc) compared to the 2014 list, making V economically more interesting in bauxite residue. Sc has been on the list individually since 2017 when HREE, LREE and PGM were assessed individually; however, they remain as groups in the critical list in order to ensure comparability with the previous assessment.⁶ Many industrial sectors crucially rely on CRMs as functional materials in their products. REE are used, for instance, in such diverse applications as phosphors

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(lightning, displays), magnets, catalysts, metallurgical applications and alloys, ceramics and glass, and glass polishing.⁹

The challenges related to safeguarding CRM supply can be addressed in a variety of ways, depending on the particular material. Demand for materials can be reduced through the development of new products and processes substituting CRMs.¹⁰ In some cases, additional supply might be available through new or improved mining of primary ores and production processes (minimizing losses). Furthermore, CRM supply risk can be reduced by exploitation of secondary (i.e. not ore-related) sources (i.e. in the framework of so-called urban mining) at the end of products' lives. Prominent secondary sources include, for example, incineration slags and ashes, demolition wastes or e-wastes.¹¹ One thus-far little tapped CRM secondary source may be bauxite residues.^{12–15} Depending on the origin of the residues, they may contain considerable amounts of CRMs, in particular REE and Sc.^{16–21}

Bauxite residues (also known as 'red mud', 'Bayer process tailings' or 'bauxite process tailings') are generated from alumina production where bauxite is digested in hot NaOH solution via the Bayer process.²² The production of 1 tonne of alumina generates between 1 and 1.5 tonnes of bauxite residue.²³ Currently, the yearly global production of bauxite residues is 150 million tonnes (Mt)²⁴ and the total inventory is estimated to exceed 2.7 billion tonnes¹² to date.

Several strategies (e.g. direct magnetic separation, pyro- and hydrometallurgical processes) have been investigated to recover the main elements present in the residues (Al, Fe and Ti). These investigations were mainly driven by waste reduction considerations.^{23,25–46} Fewer attempts have been made to use the residues as a secondary source of CRMs (see Current bauxite residue reuse options for metal / CRM recovery, below).^{17,20,39,47–55}

Bauxite residue CRM extraction requires the use of chemicals and energy. So far, investment and operational costs of CRM extraction have been simply balanced against the value of the materials being recovered, and all strategies have to date been deemed 'unfeasible' or 'financially unfeasible'. Although certainly justifiable in economic terms, the isolated consideration of benefits from CRM recovery alone is inadequate from a sustainability perspective. The current practice of residue stockpiling in tailings is associated with a number of disadvantages, in particular the inherent risk of dam failure and negative environmental impacts of operation (e.g. dust formation, leaching and soil loss), but also socio-economic concerns (e.g. future financial liabilities associated with tailings). Admittedly, these impacts are harder to translate into quantifiable factors, even though methodologies do exist for doing so [e.g. life-cycle assessment (LCA)]. This review aims at summarizing benefits beyond the mere financial value that is recoverable from bauxite residues. To understand the impacts and benefits of CRM recovery from bauxite residues, a brief overview of bauxite residue generation / deposition, associated impacts and strategies for bauxite re-use is given as well. For technical details, the reader is referred to several extensive reviews on bauxite residue management^{56,57} and element recovery.^{14,20,58,59}

PRIMARY MINING OF ALUMINA AND RESIDUE TREATMENT OPTIONS

Bauxite ore occurrence

Bauxite ore is formed from the intense lateritic weathering of residual clays, which accumulate in topographic lows on continental surfaces.¹³ Bauxite deposits can be classified according to their geological formation into lateritic (89%), karst (10%) and

Tikhvin-type (<1% of world's deposits).^{60,61} Trace elements, including REE, Ga, Ti, Cr and Zr, can be adsorbed onto the surface of residual clays. During lateritic weathering of these clays, the adsorbed elements become concentrated with depth in the resulting bauxite deposits.^{62,63} Bauxite resources are estimated to be 55 to 75 billion tonnes worldwide with the following distribution: 32% in Africa, 23% in Oceania, 21% in South America and the Caribbean, 18% in Asia and 6% elsewhere.^{64,65} The 1970s saw a major expansion of the alumina industry in response to growth in primary Al demand, accompanied by a rapid production growth.⁵⁶ In 2016, the quantity of bauxite consumed was estimated to be 6.8 Mt and more than 90% of this was converted to alumina, with the remainder used in nonmetallurgical products.⁶⁶

Bauxite residues from alumina production

Aluminium production consists of two key stages: The first is alumina refining (Bayer process), which involves the generation of alumina from bauxite ore; the second stage is aluminium smelting (Hall–Héroult), the process of transforming alumina into aluminium.⁶⁷ Most of the world's alumina is produced using the Bayer process, although minor volumes are obtained from non-bauxite sources such as kaolin, anorthosite, alunite and dawsonite in some refineries (particularly in China and Russia).^{68,69} The Bayer process is often referred to as the 'red side' and affects the properties of the residue produced. It includes the steps of bauxite milling, pre-desilication, digestion, cooling, clarification, washing and disposal of residues. The steps associated with the obtained pregnant liquor are precipitation / crystallization, classification and calcination.⁷⁰

Depending on the requirements of the residue storage facility, further filtration steps and amendments (e.g. seawater, CO₂, SO₂, gypsum, mineral acids) are added prior to disposal.⁵⁶ Various size fractions can be distinguished within bauxite residues (from 100 nm to coarse fraction process sand (>150–200 µm)).^{71,72} The majority (~80%) of the particles in red mud may be less than 100 µm.⁷³ The amount of process sand is typically around 5%, but may vary between <1% to as high as 50%. In several cases the process sand is separated before clarification and is transferred to washing in a separate system.⁷⁴

Bauxite residue composition

Bauxite residues are solid–solution mixtures ranging in initial solids content from 20% to 80% by weight (depending on the disposal method of the refinery) characterized by high pH (up to 13), a high sodium (Na⁺) content and electrical conductivity (EC).^{75,76} Roughly 70% of the solid bauxite residue is in a crystalline phase,⁷⁵ including primary mineral phases which are those that are already present in bauxite (e.g. hematite, diasporite, boehmite and goethite) and secondary mineral phases which are formed during the Bayer process, such as hydrogarnet, cancrinite, perovskite and gibbsite.⁷⁷ The bauxite parent material, climate, age and topography⁷⁸ influence bauxite residue composition. Hematite is the most prevalent of the minerals found in bauxite residue, ranging from 7% to 29%.⁷⁵ Aluminous goethite is generally found in residues which have used bauxite from Jamaica and Australia,⁷⁵ which shows a low substitution of aluminium within the goethite structure.⁷⁹ Further factors influencing bauxite residue composition are NaOH, heat and pressure conditions used during digestion, as well as chemical additives used.⁸⁰ For instance, residues produced from 'low grade', high silicon bauxite differ due to the amount of caustic lime that is added in the Bayer process. Here, the

major minerals present are calcite, perovskite, illite, hematite and magnetite.⁸¹

A typical order of elemental abundance within the bauxite residue would typically be Fe > Al > Si~Ti > Ca > Na,⁷⁵ these elements being present as oxides (Fe₂O₃, Al₂O₃, SiO₂, TiO₂, CaO and Na₂O) (for a review, see¹²). However, comparatively little data are available on the content of CRMs in bauxite residues (Table 1). Some studies have indicated that bauxite residues may be rich in valuable CRMs (Sc, Cr, Ga, REE) and other valuable elements (Ni, V, Zn, Zr).^{12–14} Mohapatra *et al.*¹⁹ reported that the amount of Sc, Ni and Cr increased in the order bauxite ore < process sand < red mud, whereas the amount of Ga was depleted in the order bauxite ore > process sand > red mud. Moreover, it was observed that the concentration of other valuable elements such as Co, Y, Zr, V, Zn and Nb was highest in process sand.¹⁹

Traditional bauxite residue treatment and disposal

Up until the 1970s, marine discharge and lagooning were primarily used to dispose of bauxite residue. So called dry stacking (even though residues are not entirely dry, at ~70% solid) and dry cake disposal have become the industry norm. Dry stacking involves thickening the bauxite residue to 48–55% solids, which subsequently is pumped through a sloped pipeline. Then, de-watering / air drying occurs before the next thin layer is released at the disposal area.⁵⁶ The bauxite residues are emitted as a slurry-type paste of pH > 11, a high fine silt to clay size proportion, and a high sodium content. This leads to a preference for particle dispersion,⁸⁶ and as a result difficulties in handling and storing residue^{87,88} (see below). Dry cake disposal involves thickening and pressure filtration, before using dump trucks to transfer residues to the storage area.⁵⁶ Pressure filtration not only results in a residue with lower water content, but also reduces losses of entrapped NaOH. This makes thickened / filtered residue that is more suitable for safer storage, and for transport and utilization in other industries (e.g. cement, iron) (see next Current bauxite residue reuse options for metal / CRM recovery, below).

The current best practice in the industry is disposal in engineered bauxite residue disposal areas (BRDAs), using dry stacking and 'mud farming' (amphirolling) to aid dewatering for compaction / consolidation of residue at reduced alkalinity and leachate treatment.^{24,89–91}

CURRENT BAUXITE RESIDUE REUSE OPTIONS FOR METAL / CRM RECOVERY

Efforts to reuse bauxite residue have long been proposed, but no more than 2–3% of the 150 Mt of bauxite residue produced annually is used in a productive way.²⁴ Currently, these minor uses include cement and ceramic production [see e.g.^{12,92–95}]. Further, technologies have been proposed for major metal (Al, Fe, Ti) recovery, such as direct magnetic separation, smelting in a blast furnace, and pyrometallurgical and hydrometallurgical recovery.¹⁴ For example, Fe recovery by direct magnetic separation was found to reduce energy costs compared to pyrometallurgical recovery.⁹⁶ The magnetic fraction can be used for Fe production, whereas the nonmagnetic portion can be used in building materials or added back into the Bayer process.⁹⁷ Al has been extracted by both organic or inorganic acids (including acids produced by bacteria).¹⁴ Pyrometallurgical (and hydrometallurgical) routes have been proposed to recover Ti from bauxite residues.²⁵

In particular due to the high costs of disposal (see below), some attempts for complete re-use of bauxite residues (towards

zero-waste objectives) have been undertaken. For instance, Erçağ and Apak²⁸ developed a procedure for the recovery of TiO₂, Al₂O₃ and pig iron from Turkish bauxite residues. Here, bauxite residues were mixed with dolomite and coke, pelletized, sintered and smelted. The slag was leached and solvent extraction applied. Ultimately, silica and Al₂O₃ were recovered from the raffinate, while Ti was stripped, hydrolysed and calcined to produce pigment-grade TiO₂.

CRM recovery strategies can be categorized either as direct leaching, leaching following pyrometallurgical / mechanical operations (reductive smelting, roasting) and combinations thereof. In direct hydrometallurgical leaching mostly mineral acids (H₂SO₄, HCl or HNO₃) have been used to recover CRMs from bauxite residues (Table 2). Leaching following pyrometallurgical treatment may result in more efficient / more selective extraction, yet at the expense of higher energy investments. For instance, Fe removal by smelting reduction was shown to be beneficial for selective leaching of REE from the resulting slag phase.^{12,48,98,99} Borra *et al.*⁴⁹ investigated a combined sulfation–roasting–leaching process to selectively leach REE while leaving Fe undissolved in the residue. They reported about 60% Sc and 80% other REE extracted while the dissolution of other major elements was minor (<1% Fe, Ti; <20% Al). Similar procedures were developed for selective extraction of Ni and Co from Fe-rich lateritic ores,^{90–102} and also applied for selective extraction of Nd from Nd–Fe–B magnets.¹⁰³

After extraction, dissolved metals are mostly purified / concentrated using either solvent extraction and / or ion exchange. Further proposed techniques, such as carbon adsorption, precipitation, ionic liquids or ultra / nanofiltration as alternatives found minor use only (Table 3). Solvent extraction (liquid/liquid extraction) is based on the partitioning of the dissolved metal into a nonmiscible organic phase (extract), followed by regeneration (stripping) of the solvent phase.¹⁰⁶ In particular dissolved impurities such as Fe pose a challenge to solvent extraction, and are usually removed by precipitation before proceeding.¹⁰⁷ A limited number of studies are available regarding extraction / concentration of CRM from bauxite residue (reviewed in Liu and Naidu¹⁴). Nonaqueous solvents (i.e. ionic liquids, salts in the liquid state) can be an alternative to conventional water / organic solvent systems. In ion exchange, a charged solid phase applied as a packed column or in direct contact is used to electrostatically attract the metal of interest.¹⁰⁸ Anion exchange resin (Amberlite®IRA-400) was investigated for V removal from synthetic bauxite residue leachate solution.¹⁰⁴ It should be noted that the concentration of CRM from BRDA leachates does not require extraction as a precursor step. Next to commercial ion exchange resins,⁵¹ some further (hybrid) materials have been applied [e.g. functionalized chitosan-silica particles with ethyleneglycol tetraacetic acid (EGTA) and diethylenetriamine pentaacetic acid (DTPA) groups].⁵³ Nanofiltration is a pressure-driven process which makes use of both fixed charges in and on a membrane material as well as steric effects (i.e. size of ions or molecules).^{109,110} Acid-resistant nanofiltration may prove to be a suitable approach in the future,¹¹¹ but to date has not been applied to bauxite residue leachates.

Further refining is required to get the dissolved metals in refined solution to end users. Various techniques have been used for the recovery of elemental metal or its suitable compound from the concentrate: metal compounds by crystallization or ionic precipitation, metals/metal compounds by reduction with gas, metals by electrochemical reduction and metals by electrolytic reduction.¹¹²

Table 1. Concentration and maximally recoverable financial value of CRM and elements of high economic importance in bauxite residues of different origin

Elements	Market price ^a [US \$/t]	Price basic	Australia		China		Greece		Hungary		India		Russia		Turkey	
			Content ^b [mg kg ⁻¹]	Financial value [US \$/t]	Content ^c [mg kg ⁻¹]	Financial value [US \$/t]	Content ^d [mg kg ⁻¹]	Financial value [US \$/t]	Content ^e [mg kg ⁻¹]	Financial value [US \$/t]	Content ^f [mg kg ⁻¹]	Financial value [US \$/t]	Content ^g [mg kg ⁻¹]	Financial value [US \$/t]	Content ^h [mg kg ⁻¹]	Financial value [US \$/t]
Ce	2000	Oxide	n.a.	1.7	842	0.8	368	430	0.9	191	0.4	n.a.	n.a.	n.a.	n.a.	
Co	26 444	Metal	n.a.	n.a.	n.a.	n.a.	n.a.	59	1.6	24	0.6	n.a.	n.a.	n.a.	n.a.	
Cr	11 000	Metal	497	5.5	848	9.3	n.a.	646	7.1	740	8.1	n.a.	n.a.	n.a.	n.a.	
Dy	184 500	Oxide	n.a.	8.9	48	3.1	17	21	3.9	4	0.7	n.a.	n.a.	n.a.	n.a.	
Er	5500	Mischmetal	n.a.	0.1	28	0.1	14	12	0.1	1	0.0	n.a.	n.a.	n.a.	n.a.	
Eu	66 000	Oxide	n.a.	7.3	110	0.3	5	6	0.4	2	0.1	n.a.	n.a.	n.a.	n.a.	
Ga	400 000	Metal	89	35.6	570	228.0	n.a.	27	10.6	91	36.4	n.a.	n.a.	n.a.	10.0	
Gd	5500	Mischmetal	n.a.	0.3	56	0.1	22	0.1	0.1	7	0.0	n.a.	n.a.	n.a.	n.a.	
Ho	5500	Mischmetal	n.a.	0.1	25	0.1	4	4	0.0	n.a.	0.0	n.a.	n.a.	n.a.	n.a.	
In	240 000	Metal	n.a.	n.a.	n.a.	n.a.	n.a.	1	0.1	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	
La	2000	Oxide	n.a.	0.8	416	0.2	114	166	0.3	112	0.2	n.a.	n.a.	n.a.	n.a.	
Lu	5500	Mischmetal	n.a.	0.1	14	0.0	2	2	0.0	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	
Nd	39 500	Oxide	n.a.	13.5	341	3.9	99	151	6.0	48	1.9	n.a.	n.a.	n.a.	n.a.	
Ni	9298	Metal	31	0.3	169	1.6	n.a.	307	2.8	53	0.5	n.a.	n.a.	n.a.	n.a.	
Pr	5500	Mischmetal	n.a.	0.5	95	0.1	28	39	0.2	18	0.1	n.a.	n.a.	n.a.	n.a.	
Sc	4 600 000	Oxide	54	248.4	158	726.8	121	80	368.0	58	266.8	n.a.	n.a.	n.a.	n.a.	
Sm	5500	Mischmetal	n.a.	0.3	64	0.1	21	28	0.1	9	0.1	n.a.	n.a.	n.a.	n.a.	
Tb	417 500	Oxide	n.a.	76.8	184	1.7	4	4	1.5	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	
Tm	5500	Mischmetal	n.a.	0.1	14	0.0	2	2	0.0	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	
V	6889	Oxide	730	5.0	4220	29.1	n.a.	337	2.3	517	3.6	n.a.	n.a.	n.a.	112	
Y	35 000	Metal	68	2.4	266	9.3	76	100	3.5	13	0.5	n.a.	n.a.	n.a.	n.a.	
Yb	5500	Mischmetal	n.a.	0.1	28	0.1	14	11	0.1	2	0.0	n.a.	n.a.	n.a.	n.a.	
Zr	918	Metal	1469	297.2	10 566	1116.6	911	n.a.	410.4	2169	320.3	n.a.	n.a.	n.a.	137	
SUM over all elements																10.8
Reference																
Alumina production in 2016 [TMT] ⁱ			Wang <i>et al.</i> ⁵⁵	20 900	Qu and Lian ²¹	60 900	Borra <i>et al.</i> ¹⁷	821	Ujaczki <i>et al.</i> ²⁰	n.a.	Mohapatra <i>et al.</i> ¹⁹	6030	Petrakova <i>et al.</i> ⁸²	2680	Abdulvaliyev <i>et al.</i> ¹⁶	n.a.

^a Prices 2016 USGS.⁸³ For Er, Gd, Ho, Lu, Pr, Sm, Tm and Yb, no market price is available; mischmetal price was used, probably underestimating actual prices.

^b The metal concentrations in all aqueous samples were determined by ICP-AES.

^c Chemical analysis was performed after total digestion according to US EPA SW 846 Method 3050B method (nitric acid (HNO₃) and hydrogen peroxide (H₂O₂)) by ICP-MS.

^d Chemical analysis was performed after complete dissolution by alkali fusion and acid digestion in a 1:1 (v/v) HCl solution by ICP-MS.

^e Chemical analysis was performed after aqua regia assisted microwave digestion with ICP-MS.

^f Chemical analysis was performed after acid digestion (HNO₃ and HF (1:1)) based on Balram *et al.*⁸⁴ method by ICP-MS.

^g Information about the metal analysis was not available.

^h Chemical analysis was performed by XRF.

ⁱ Data in thousand metric dry tons (TMT). 2016 USGS.⁸³ Producing 1 tonne of alumina generates 1–2 tonnes of bauxite residue.⁸⁵

Table 2. Summary of CRM extraction techniques for bauxite residues

Bauxite residue source	Target metals	Technique	Recovery efficiency [%]	Fe co-extracted	Scale of the study	Reference
Greece	REE	Extraction using HCl	68% Sc, 33–70% REE	11% Fe	Lab-scale in 0.5 L reactor	Ochsenkühn-Petropoulou et al. ⁵¹
Greece	REE	Extraction using HCl	63% Sc, 70–85% REE	80% Fe	Lab-scale in 0.5 L reactor	Borra et al. ¹⁷
Greece	REE	Extraction using HNO ₃	80% Sc, 29–96% REE	3% Fe	Lab-scale in 0.5 L reactor	Ochsenkühn-Petropoulou et al. ⁵¹
Greece	REE	Extraction using H ₂ SO ₄	21–77% REE		Lab-scale in 0.5 L reactor	Ochsenkühn-Petropoulou et al. ⁵¹
Greece	REE	Extraction using HbetTf2N	45% Sc, 60–80% REE	3% Fe	Lab-scale ~ 0.5 L reactor	Davris et al. ⁵⁰
Hungary	REE, Co, Cr, Ga	Extraction using HCl	64% the total CRM	67% Fe	Lab-scale in 0.05 mL tubes	Ujaczki et al. ²⁰
India	La	Extraction using H ₂ SO ₄	100%		Lab-scale ~ 0.1 mL vessel	Abhilash et al. ⁴⁷
India	Ce	Extraction using H ₂ SO ₄	100%		Lab-scale ~ 0.1 mL vessel	Abhilash et al. ⁴⁷
Turkey	Ga, V	High-temperature leaching using a high-modulus solution	56% Ga, 66% V ₂ O ₅		Lab-scale ~ 0.1 mL vessel	Abdulvaliyev et al. ¹⁶
China	REE, Ga, V	Bioleaching by filamentous fungi (<i>A. niger</i>)	47% Ga, 40% Sc, 25–55% REE		Lab-scale in 12 L reactor	Qu et al. ³⁹
China	Cr, Ni, Zn, Zr	Bioleaching by filamentous fungi (<i>A. niger</i>)	25% Cr, 50% Ni, 80% Zn, 11% Zr		Lab-scale in 12 L reactor	Qu et al. ³⁹
China	REE	Bioleaching by filamentous fungi (<i>P. tricolor</i>)	73% Sc, 28–80% REE		Lab-scale in 12 L reactor	Qu et al. ³⁹

Table 3. Examples of concentration/purification of CRM from bauxite residue

Bauxite residue source	Target CRM	Technique	Efficiency	Fe co-extracted	Reference
Australia	Sc	Combined LLE with D2EHPA/TBP in Shellsol D70	99% Sc transfer from leachate to solvent	1% Fe	Wang et al. ⁵⁵
Hungary	REE	Combined LLE with D2EHPA in kerosene	62% Sc transfer from leachate to concentrate	13% Fe	Ujaczki et al. ⁵⁴
Hungary	V	Anion exchange by Amberlite®IRA-400	76% V eluted from resin		Gomes et al. ¹⁰⁴
Greece	Sc	Ion exchange with functionalized hybrid materials	100% Sc transfer from leachate to ion exchange		Roosen et al. ⁵³
Greece	Sc	Solvent extraction using ionic liquids	>90% Sc transfer from leachate to solvent		Hoogerstraete et al. ¹⁰⁵

DIRECT AND INDIRECT BENEFITS OF CRM RECOVERY FROM BAUXITE RESIDUES

There are a number of benefits that can arise from using bauxite residues as a secondary source of CRM. To some of them a monetary value can be easily assigned. For instance, the financial value of recovered materials is directly reflected in their market prices, and reduced financial costs through savings in disposal can be quantified easily as well. Thus, CRM recovery is in particular financially interesting under the frame of zero-waste overall reuse of bauxite residues (see above), or even on itself depending on the market price of recovered CRM.^{113,114}

The reduction of long-term liabilities can be quantified in monetary terms as well. In addition to direct monetary benefits, improvements in environmental protection and health can be

expected due to emission reduction on the disposal sites, but as well savings in emissions from conventional primary mining of CRM. Sustained social and technological development are certainly harder to quantify; nevertheless, they should be considered and appropriately communicated to decision makers. Fertile soil provides more services than just food and feed provision (e.g. recreational value and protection of biodiversity), thus quantifying the benefit conferred by tailing volume reduction certainly touches upon the economic, environmental and social fields and is a prerequisite for sustainable development.

Financial value of recovered metals and CRM

When comparing bauxite residues of different origin, the maximally recoverable financial value (quantity × estimated market

price of pure element) may be used. It should be noted that this maximally recoverable financial value does not correspond to a benefit that can actually be achieved (because it is based on total content and pure, refined metals). Still, it allows weighted consideration of minor amounts of CRMs, if they are of high economic value. A comparison (Table 1) shows that most bauxite residues are in the same range of overall CRM contents (i.e. sum of concentrations of all CRMs). Some higher concentrations of valuable elements were reported in Chinese bauxite residues, even though Chinese, Greek and Hungarian bauxite residues stem from the same bauxite ore deposits (karst type). The difference in maximally recoverable financial value is mainly conferred by high value metals (i.e. Ga, Sc and Tb). Therefore, many research efforts have been focusing for instance on Sc,¹¹⁵ because it holds between 65%²¹ and 93%¹⁷ of the maximally recoverable financial value (Table 1). One should note that prioritizing extraction of CRMs according to their maximally recoverable financial value is limited by the fact that only mischmetal (REE) or pure elemental / oxide prices are available for most elements.⁸³ Further, the price is often extrapolated from minute amounts bought at commercial suppliers (and not on commodity markets; see, e.g., Sc¹¹⁶). Production of pig Fe from bauxite residue (BR) alone cannot provide financial feasibility, thus a combination of base elements and CRM extraction is mandatory to obtain a financially viable process.¹¹⁵

Reduced disposal costs

There are relatively few published data on the cost of disposal of bauxite residue but it is generally estimated to be 2% of the alumina price, with an estimated disposal cost around US\$9 per ton.^{22,117,118} The evaluation of current available data does not allow an extended elaboration on this matter; however, a detailed assessment of site-specific costs and benefits is required to calculate true disposal costs and evaluate the feasibility of re-use technologies.

Alumina refineries tend to be located close to bauxite mines and / or ports to facilitate the cost-efficient transport of raw materials and final products.⁵⁶ For the same reason, residue disposal areas are usually close to alumina refineries as well. Topography, availability of land and rainfall are three of the key determinants that limit the choices for tailings.⁵⁶ As a consequence, land available for tailings may be limited in some locations, and the ever-growing demand for BRDS, ultimately threatens the longevity of established alumina refineries. As soon as the capacity of currently used land is reached, the refineries either have to find new disposal areas meaning additional investment costs (acquisition of land, construction of the disposal site) and potentially higher operational costs (transport).

Reduction of long-term liabilities

Tailing failures causing release of bauxite residues are rare and unintended events. There have been numerous scientific studies assessing the key risks and impacts associated with the largest single release of bauxite residue to the environment: the Ajka bauxite residue spill in western Hungary in October 2010. This accident was the largest documented release of bauxite residue, where more than 800 000 m³ of highly alkaline (pH = 13) residue slurry flooded the environment.^{119,120} Along with the resultant human tragedy, when the flood killed ten people and injured more than hundred people were, and irreversibly damaged many homes, further immediate impacts after the spill were associated with the highly caustic nature of the bauxite residue.^{87,121} In addition, the

fine particle size generated fugitive dust upon desiccation following the spill. Further longer-term environmental impacts were related to the saline nature of the spilled material and the release of oxyanion-forming metals and metalloids (e.g. Al, As, Cr, Mo and V) into the soil–water environment.^{87,122} Other accidents involving bauxite residue spills were those in Dahegou Village (Luoyang, Henan province, China, 2016; unknown amount spilled) and Miraf (Minas Gerais, Brazil, 2007; 2 million m³).¹²³

It is mandatory to analyse the possibility and impacts of tailing failures by quantitative risk assessments, which compare the relevant frequency/extent scenarios with a risk acceptance area. To operate tailings, such risk assessments (together with environmental impact assessments) must be deemed acceptable and comply with local laws to get permissions by the respective agencies.¹²⁴ In general, the global risk can be at least roughly estimated as the product of frequency of incidents and their impact. Looking at the practise of tailing storage of other mining operations, Azam and Li¹²⁵ found a staggering average global failure rate of 2.2 dams per year over the last hundred years. This means that over this time span about 1.2% of all dams failed. Main failure reasons identified were ‘unusual rain’ events and ‘poor management’, which are not exclusive to certain mining sectors. Thus, even though the absolute number of bauxite tailing incidents is low (see above), the actual frequency may be somewhat similar. However, one should note that applying proper engineering standards and maintenance to the disposal dams (as done in most Al operations) can reduce the frequency of such failures significantly. If similar standards typical of water retention dams would be applied, then the failure rate could potentially be reduced by two orders of magnitude¹²⁶ (at the expense of making disposal more expensive).

Regarding the financial consequences of bauxite tailing failures, there are even fewer reliable data. Generally, the primary economic consequences of tailing failures for companies are business interruption (in particular when authorities close down operations following accidents) and environmental damage (requiring remediation efforts), whereas socioeconomic damages include losses to agriculture, fishing, tourism and nature conservation,¹²⁷ and would fall under liabilities – if legislation is enforced at all. Admittedly, it is difficult to assign an economic value (or price) to the consequences of tailing failures, basically because there is no ‘average’ failure: the materials contained can be of very different environmental concern (e.g. highly problematic cyanide and / or heavy metal-rich tailings from sulphidic ore processing versus comparatively metal-depleted bauxite residues). The released volumes can vary considerably (e.g. 32 million m³ released from Germano mine, Brazil, 2015 versus 0.8 million m³ at Ajka, Hungary, 2010 WISE).¹²³ Furthermore, the targets and costs for remediation measures may vary considerably from country to country, whereby one cannot give an average cost for tailing failures. Nevertheless, it can be assumed that for a major failure, these are easily at the multi-million dollar scale. After the Ajka dam failure, the Hungarian government spent a total of 88 billion Forints (over US\$280 million) to rebuild over 300 houses and to clean up and recultivate the environment. According to a court decision, the operating company was responsible for the accident, and thus should pay compensation to the affected families.¹²⁸ The global risk of tailing failures can be at least roughly estimated as the product of frequency and impact. For serious tailing failures, the global risk certainly reaches millions of US dollars each year. In some cases, the probability of a major failure may grow over time and the hazard may remain long after production has stopped,¹²⁶ which

poses a particular problem: the 'polluter pays' principle as stipulated in many countries may be difficult to enforce if operating companies go bankrupt over the course of time. In the case of take-overs and mergers of these companies, this legacy will represent a major financial risk to be considered.

Environment and health improvements due to emission reduction

A little-considered fact that argues for CRM recovery in the frame of a zero-waste attempt is the mitigation of long-term impacts to the environment and human / animal health, associated both with the current practise of tailing storage, as well as the current practises of primary CRM production.

For tailing storage, key negative impacts are conferred by leaching of metals and metalloids, naturally occurring radioactivity, alkalinity and high sodium content of the residues, and their fine particle size (dust formation, transport). Numerous researchers have reported that bauxite residue may contain problematic concentrations of harmful metals and metalloids, including As, Cr, Ni, Pb, Mo and V.^{75,87,122,129} Upon contact with water, in particular the mobility of oxyanion forming trace elements (e.g. Al, As, Cr, Mo and V) in bauxite residues is expected to be high due to electrostatic repulsion with negatively charged solid surfaces and competitive sorption with salts (EC 1.4–28.4 mS m⁻¹, up to pH 13).^{75,87,129–131}

Most bauxite will contain low levels of radioactive elements (²³⁸U and ²³²Th and their decay products): for instance, Hungary ~ 0.27 Bq/g ²³⁸U, 0.26 Bq/g ²³²Th; Greece ~ 0.15 Bq/g ²³⁸U, 0.47 Bq/g ²³²Th; Australia ~ 0.40 Bq/g ²³⁸U 1.13 Bq/g ²³²Th; Jamaica ~ 0.22 Bq/g ²³⁸U, 0.04 Bq/g ²³²Th; and China ~ 0.46 Bq/g ²³⁸U 0.37 Bq/g ²³²Th.^{22,132,133} Although some of the ²³⁸U dissolves in the Bayer process and is associated with the coarser bauxite residue fraction, ²³²Th is not – it is most often associated with the fine fraction.²² When brought into a safe, stabile form / phase, processing of bauxite residues for CRM recovery may in fact result in an overall lower environmental impact by naturally occurring radionuclides.

Risk assessments following the immediate aftermath of the Ajka bauxite residue spill focussed on the impact of fugitive dusts on ecological and public health.^{87,134} According to Gelencser *et al.*,¹³⁵ bauxite residue showed a particle size distribution that fell predominantly within the particulate matter (PM)₁₀ class, centered around 3–8 µm. A smaller portion was even found in finer aerosol classes (PM_{2.5} and PM₁: with diameters less than 2.5 and 1 µm, respectively). The authors concluded that bauxite residues showed similar distributions to urban dusts and, therefore, no risks above that typically encountered with urban dusts would be anticipated.¹³⁵ However, air pollution is the single largest environmental health risk in Europe: concentrations of PM exceeded the EU limit and target values in large parts of the continent in 2014.¹³⁶

Mining and smelting of primary metal ores has a number of considerable impacts on the environment, human and animal health (next to social impacts; see the next section). All steps from blasting and excavation of the ores, to crushing, milling, beneficiation, roasting / oxidation, cyanidation, electrowinning, smelting and ultimately refining to pure metals are energy- and / or chemical-intensive, causing a number of emissions to air, soil and water. For example, the total greenhouse gas emission emitted by metal production facilities in 2016 was 86.7 Mt CO₂e.¹³⁷ There is a large number of reviews available on these impacts (for instance^{138–141}). Regarding REE, to date, mainly bastnasite, monazite, xenotime and ion-adsorbed clays have been mined. Following mining and beneficiation, hydrometallurgical routes consisting

of cracking, leaching/neutralization/precipitation, and ultimately separation and purification by solvent extraction and / or ion exchange are used to obtain REE.¹⁴² In particular, the cracking process is energy- and chemical-intensive, and generates a range of gaseous and aqueous emissions (e.g. SO_x, HF, CO₂).¹⁴³ Next to these, radionuclides are particularly problematic due to their high contents in some original ores (e.g. Monazite contains up to 12 wt% Th and 14 wt% U).¹⁴³ Potentially, but not necessarily, some of these emissions can be mitigated by tapping alternative secondary sources, such as bauxite residues.

Sustained social and technological development

Miners' and local communities' occupational health and safety (OHS) provisions as well as environmental protection often increase financial costs, making raw material production considerably more expensive. In some cases, strict environmental protection requirements even may prohibit production [e.g. closure of Mountain Pass (CA, USA) REE mining due to radionuclides].¹⁴⁴ Owing to less stringent and nonenforced regulations, certain countries or regions where raw materials stem from unregulated primary production and refining may gain a competitive advantage. It is believed that approximately 15% of the LREE and 50% of the HREE produced in China in 2012 came from such unregulated production.¹⁴⁵ For other metals, the situation is even more dramatic: Approximately 90% of Brazil's Au production and 100% of Peru's Sb production is attributable to unregulated artisanal mining,¹⁴⁵ as is about 90% of the Democratic Republic of Congo's mineral resource production (Au, diamonds, Cu, Co, coltan).¹⁴⁶ About 100 million people depend on artisanal mining compared to about 7 million people worldwide in industrial mining.¹⁴⁷ Next to severe OHS and environmental concerns, artisanal small-scale mining also seriously hampers social development of the communities for instance by the absence of working contracts, neglecting of woman rights and the use of child labour. Sustainable (or 'fair') CRM recycling from secondary sources such as tailings would thus, next to an economic benefit, also allow for better social development. Moreover, formalization of artisanal and small-scale mining could reduce environmental contamination, avoid inappropriate disposal of tailings, promote formal employment, eliminate child labour, avoid mineral loss and increase the value of extracted minerals. For example, in 2010 the Peruvian government issued the terms and conditions for mining formalization, including various permitting and accreditation requirements, as well as water and environmental approval criteria.¹⁴⁸ In addition, sustained technological development can be a striking argument for CRM recovery from secondary sources. Although CRMs are crucial for many high technology appliances, supply shortages (and associated increased prices) are less problematic for finished products that contain small quantities of CRMs and have a high market price. Other end products, however, employ high amounts of CRMs (e.g. NdFeB magnets, Sc/Al alloys) and have on a weight basis – compared to mobile phones – a relatively low market price. For such materials, it can be assumed that due to the lack of a secure CRM supply, at least to some extent novel products are not further developed and / or brought to the market, which poses a risk to technological development of society as a whole.

Reduced loss of fertile soil and habitable land

About 2000 million ha of soil, equivalent to 15% of the Earth's land area, have been degraded already owing to human activities.¹⁴⁹

The main types of soil degradation are water (56%) and wind (28%) erosion, chemical (12%) and physical (4%) degradation. The causes of soil degradation include overgrazing (35%), deforestation (30%), agricultural activities (27%), overexploitation of vegetation (7%) and industrial activities (1%).^{149,150} It is estimated that soil loss from land areas occurs 10 to 40 times faster than the rate of soil renewal.¹⁵¹ In the European Union alone, the mean soil loss rate has been estimated at 2.46 t ha yr⁻¹, resulting in a total soil loss of 970 Mt annually.¹⁵² Whereas, on a global scale, soil degradation from surface bauxite deposition appears negligible, it may nevertheless still have detrimental effects, mostly on a local or regional scale. First, areas larger and distant from the actual deposits may be affected through deposition of fine dust particles, in particular in dry stacking. Secondly, functional soils are not equally available, and industrialized and / or densely populated areas may suffer a higher pressure from soil losses than less developed areas. Therefore, soil is a valuable – yet often unvalued – resource that needs protection, in particular in industrialized and more densely populated areas.

The concept of opportunity cost (also known as alternative cost) can be used to assign a monetary value to such soil loss. It is based on the difference in value (i.e. the cost) between the best possible situation (or 'choice') and the actual situation. For instance, Dijkgraaf and Vollebergh¹⁵³ argued that space occupied by a landfill should be valued at the price of the most valuable opportunity forgone. For densely populated Flanders region (Belgium), this would be residential building space at a price of € 155 per m². Although opportunity costs appear to be a straightforward concept to quantify costs of tailing deposition, it suffers a major shortcoming: soil is not only the provider of food, timber or living space. There are a variety of indispensable functions that crucially depend on a functional soil, such as the maintenance of biological diversity, protection against natural hazards, hydrological balance functions, functions as a pollutant filter, and as long-term carbon sink.¹⁵⁰ These are not considered when considering residential building space as the most valuable opportunity forgone. Although the protection from natural hazards or hydrological functions may still be assigned a monetary value (i.e. frequency × cost, see Reduction of long-term liabilities above), it is hard to quantify the value (and thus the opportunity costs) of 'maintaining biodiversity' or 'acting as carbon sink'. There is an approach called The Economics of Ecosystem and Biodiversity (TEEB) that is a global initiative focused on 'making nature's values visible'. TEEB could be used by countries to recognize the benefits provided by ecosystems and biodiversity, demonstrate their values in economic terms and capture these values in decision-making.¹⁵⁴

IMPACTS OF CRM RECOVERY FROM BAUXITE RESIDUE

Extraction of CRMs from bauxite residues comes at a price: environmental emissions during extraction, processing and refining. Thus, next to technical and economic considerations, the first question when it comes to CRM recovery from secondary sources should be: Are there fewer emissions associated with a unit mass of CRM recovered from a secondary source in contrast to primary mining? In other words, is recovery of CRMs environmentally feasible? Only if the emission savings outbalance emissions related to primary mining CRM recovery can be sustainable from an environmental and health impact stance. The quantitative accounting of materials and energy used, and related emissions being caused can be investigated in the frame of an integrated life-cycle assessments

(LCAs).¹⁵⁵ Altogether when considering the industrial implementation of extracting CRMs from bauxite residue, it is important to consider risks in addition to emissions that could be mitigated by the application, which can be assessed by environmental impact assessments (EIAs).

Many strategies for metal / CRM recovery from bauxite residues rely on pyrometallurgical processing in combination with hydrometallurgical methods (leaching, solvent extraction and precipitation).^{28,30,34,35,37,38,45} Understandably, large amounts of energy and chemicals are used during the many process steps. For instance, complex processing of bauxite residue to produce Sc has been reviewed in Borra *et al.*¹⁵⁶ and references therein. In that processing protocol, Bauxite residue is treated by a combination of electric arc furnace smelting, slag acid leaching and solvent extraction. To produce 300 g of Sc from 3 t of bauxite residue (at 100 ppm Sc), staggering amounts of chemicals (including amongst others 3 t limestone, 0.5 t coke, 10–20 kg electrodes, 3.6 m³ H₂SO₄, 64.8 m³ H₂O, 33 m³ solvent extraction agent) and energy (3600 kWh) are used. In addition the production and transportation of the chemicals consume energy and materials. From the mere numbers, however, intuitively it becomes questionable if such investments are justified from an environmental point of view or need to be compared with the currently environmentally best available sourcing path. In contrast to pyrometallurgical strategies, direct leaching (Table 2) in hydrometallurgy may be less energy- and chemical-demanding, yet may yield lower extraction yields.

It should be noted that some bauxite residues may emit ionizing radiation due to the presence of naturally occurring radioactive materials (²³⁸U, ²³²Th and members of their decay chains; Gräfe *et al.*⁷⁵ and references herein), which may be co-extracted and / or concentrated during purification, posing a challenge to CRM recovery from a technical point of view (i.e. purity to be achieved), and raising concern from an environmental / occupational health and safety perspective. Ideally, extraction should aim at selective extraction of target CRM, leaving naturally occurring radioactive materials behind (at low concentration). Comparing concentrations of bauxite-associated radionuclides (usually few mg per kg) to, for example, Monazite currently used for REE mining (many wt% of U/Th), suggests that using bauxite may be beneficial from this perspective.

FUTURE DEVELOPMENTS TOWARDS LOWERING THE IMPACTS OF CRM EXTRACTION / PURIFICATION

For extraction, bioleaching may prove to be a cost-effective means of lowering the environmental impact. For instance, *Penicillium tricolor* and *Aspergillus niger* were used in a one-step and a continuous bioleaching process,³⁹ and *A. niger* showed almost the same metal-leaching efficiency as commercial citric acid at half its cost. Bioleaching is particularly interesting where sufficient organic wastes are available on site, allowing a cost-effective production of organic acids.

For concentration of metals, one may also make use of biological materials instead of solvent extraction / ion exchange. Biosorption using bacteria, fungi, algae or even agricultural waste, exploits both covalent / sorptive processes between functional groups at the bio-based surface and metal ions of interest (reviewed in, e.g.^{157,158}). Ionic liquids, organic molten salts with low or negligible vapour pressure, have recently emerged for metal / CRM

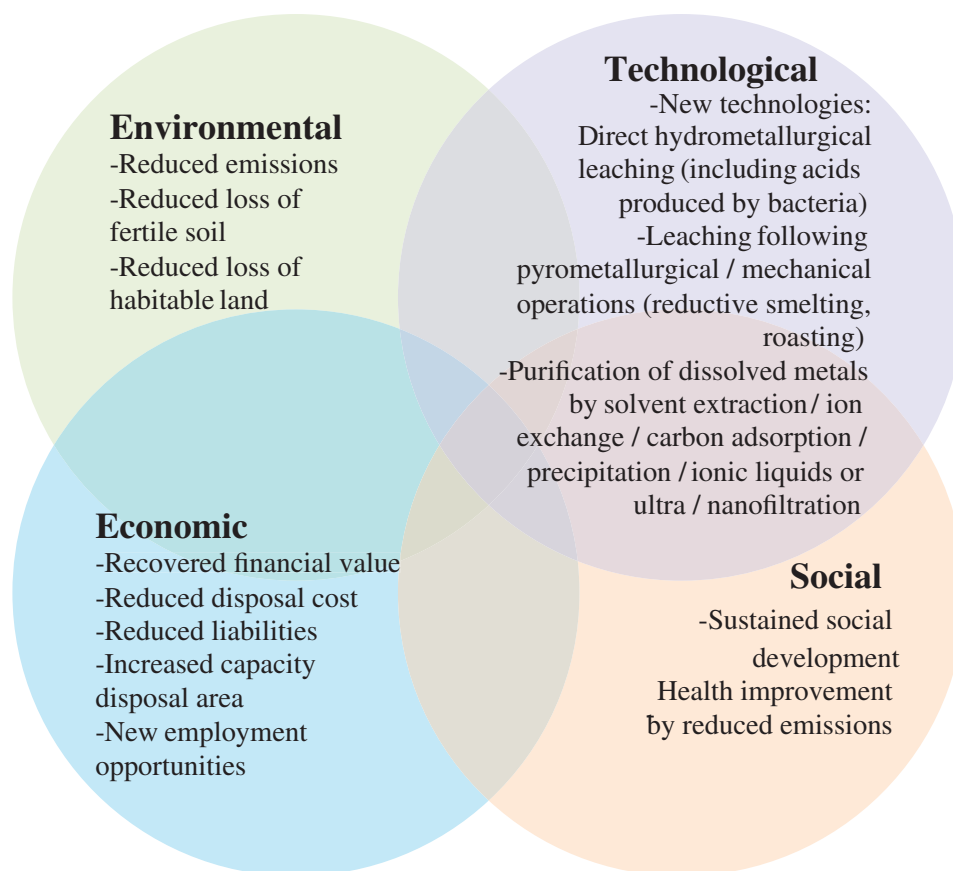


Figure 1. Benefits of CRM recovery from secondary sources such as bauxite residues.

extraction.^{50,159} Ionic liquids may thus offer a benign alternative due to their claimed lower environmental impact in comparison to conventionally used solvents, amongst others due to a potential lower toxicity.¹⁶⁰

One approach for the future of bauxite residue management may be its use in soil remediation and improvement to degraded soils.^{120,161–170} Bauxite residues have been used, for instance, to increase the phosphorus retention of sandy soil^{165,168} and to increase the low pH of acidic sandy soil.^{167–169} The combined presence of ferric, aluminium and tectosilicate-like compounds in bauxite residue makes it capable of immobilizing toxic metals from polluted soils.^{171–174} CRM extraction from bauxite residue will result in acidic solid residues (refractory mineral fractions with acid remains).²⁰ Mixing acidic residues from CRM extraction with original bauxite residues (in an appropriate ratio) for soil improvement may be the ‘golden way’ of unifying remediation (mitigating risks and decreasing costs) with resource recovery (e.g. mitigating supply risk, generating benefits and ensuring social development).

CONCLUSIONS

Recovery of CRMs from bauxite residue is not only attractive from the point of financial by recovered elements alone. It is also motivated by a number of further economic, social, environmental and technological benefits (Fig. 1). Private operators and investors often only take into account immediate financial costs and benefits in their decisions, yet long-term benefits such as reduced liabilities in case of accidents and / or more stringent environmental legislation should be anticipated. Admittedly, it is often

challenging to assign an economic value to environmental, health and other nonfinancial impacts. However, there are valuation techniques (e.g. willingness to pay/accept) to account for these and their value is definitively not zero. Society has a strong interest in accounting for these external costs and benefits. Both legislators and companies truly committed to sustainability are responsible to respect society's interests and make sustainable CRM recovery a reality. There are numerous approaches that have been shown to (technically) efficiently recover CRMs (Tables 2 and 3). However, the various approaches have an associated environmental impact and involve certain risks. In other words, a detailed process layout including energy and material flow analysis as well as risk assessment (through, e.g., EIA, LCA and CBA) is needed in order to decide for the ‘best technology’. Accordingly, the technologies should be compared with current practices of primary mining to prove their superior sustainability. Only if overall there are fewer emissions and risks associated per unit of CRM recovered, should secondary sources such as bauxite be tapped.

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