



Using Life Cycle Thinking to Assess the Sustainability Benefits of Complex Valorization Pathways for Bauxite Residue

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Abstract

Bauxite residue, the main waste product of alumina production, is a potentially valuable secondary resource. The MSCA-ETN REDMUD project aims to develop *environmentally friendly* technologies to realize this value, by extracting valuable metals (aluminium, iron, titanium, scandium, rare-earth elements) or utilizing it in construction applications. Simply utilizing a waste product as an input is not, however, sufficient to claim that a process is environmentally friendly; the processes developed must be demonstrably better for the environment, from a life cycle perspective, than business as usual. The earlier in the research and development process that environmental information can be taken into account, the more impact it can have on decision-making. In this study we demonstrate that Life Cycle Thinking approaches can provide actionable environmental information at an early stage in the research process, and that in doing so it can help steer early stage technology development towards overall improved industry environmental performance. Knowledge of the potential environmental benefit from displacing different materials can help identify primary or additional targets, for example the use of metal extraction residues for construction materials. A high-level ‘red flags’ assessment of the relative environmental impact of inputs to valorization processes and the products they displace can be used to identify problematic inputs and processes in the absence of quantitative details. Finally, once preliminary quantitative data are available for a process, streamlined Life Cycle Assessment can be used to calculate the environmental balance of a process, and identify specific hotspots of environmental impact.

Keywords Bauxite residue · Red mud · Valorization · Sustainability · Life Cycle Thinking · Life Cycle Assessment

Introduction

Bauxite residue (BR), or red mud, is a hydrometallurgical sludge generated from the digestion of bauxite in the Bayer process to produce aluminium hydroxide [1], the precursor to alumina, and is arguably one of the world’s most important and abundant industrial by-products [2]. As the aluminium industry grew during the 20th century, volumes of stockpiled BR grew near exponentially [1], and by the 1980s,

in addition to the technical and economic challenges posed by its disposal, the environmental consequences of bauxite residue management became a growing topic of discussion [1, 3]. Stockpiles of BR have continued to grow. By 2010 it was estimated that over 3000 million tonnes of BR existed in stockpiles [4]. With annual production estimated to be around 150 million tonnes, and only 2–3% of this reused [2], this figure is likely to be closer to 4000 million tonnes today.

Much of the progress surrounding bauxite residue management revolved around avoiding its disposal to rivers and the sea, and a move towards dewatering technologies to save space and avoid potential environmental hazards associated with the highly alkaline leachate which pools on top of BR lagoons [5]. However, in addition to this there have been countless attempts to find uses for this residue [2].

The chemical and mineralogical properties of BR are such that a broad range of reuse or valorization options can potentially be exploited. This includes both material extraction and utilization of its bulk properties. From a materials extraction perspective, iron, alumina, titanium oxide,

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rare-earth elements (REEs) and scandium are the most promising targets [6]. As a bulk material, BR can be used as an alternative to bauxite as a source of iron and alumina in the production of ordinary Portland cement [6], and has the potential to be used in much higher proportions in alternative cement formulations [7]. Bauxite residue can also be transformed into a precursor for the production of inorganic polymers [8].

In 2015, a project to research new technologies to extract valuable materials from fresh and stockpiled BR and to utilize it at high volume began. The EU Horizon 2020 funded MSCA-ETN¹ for Zero-Waste Valorization of Bauxite Residue (REDMUD) consists of 15 Ph.D. researchers across nine institutions and industrial partners around the EU. By functioning as a research network, collaboration between MSCA-ETN REDMUD researchers is fostered, allowing potential synergies between individual pathways to be identified. Thus while each of the participants has a primary focus, the ultimate aim of the project is to create a set of combined, multi-stage valorization flow sheets, in which value is maximized and no residual waste remains.

In addition to the technological development of these flow sheets, it is a stated aim of the project that the resulting processes be *environmentally friendly*. Too often, researchers presenting technologies which utilize industrial wastes or by-products use the term environmentally friendly (or related terms such as *eco-friendly*, *low-carbon*, etc.) without providing any quantitative evidence for such a claim. Rather, the claim is based either on the implicit assumption that because waste and waste treatment is ‘bad’, utilization of this waste is inherently ‘good’, or that the conventional material being displaced by the newly generated product has a high environmental impact, therefore, the waste-derived product *must* be better for the environment e.g., [7, 9, 10]. In the same way as claims of the structural and analytical properties of materials must be underpinned by scientific analysis, so must environmental claims.

In the case of BR, for on-site, or low value measures, such as soil amelioration or revegetation [2, 11], a simple analysis of the environmental impacts associated with BR disposal which are avoided by these practices may suffice for this purpose. The proposed valorization processes emerging from the REDMUD project, however, are complex, often comprising multiple stages, utilizing a variety of additional input materials and high-energy processes or producing multiple products. A more holistic approach is therefore required.

The production of useful materials from BR potentially displaces the need for these materials to be produced elsewhere in the economy. Avoiding the need to produce these materials also avoids the environmental burdens associated

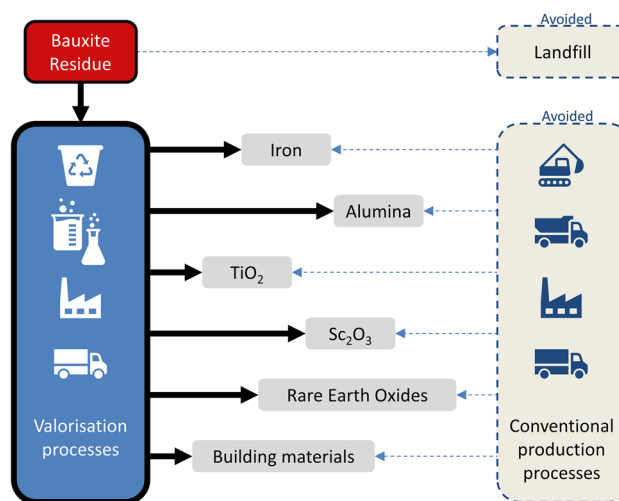


Fig. 1 Simplified environmental balance from the valorization of bauxite residue (Color figure online)

with the extraction of raw materials, transport, processing, etc. required to produce them. The net environmental impact of a given BR valorization system, its *environmental balance*, is therefore determined by two factors: the environmental impact incurred through the processes carried out, and the environmental benefit associated with the production of the conventional products and processes which are avoided as a result (Fig. 1).

Multi-stage valorization, where value is extracted from BR in multiple ways, has the potential to beneficially influence both sides of this balance. *Sequential* production of multiple resources adds to both the incurred environmental impacts and the avoided environmental impacts. However, both the incurred and avoided environmental impact of the second process in the sequence may be positively affected, through increased process efficiency (lower impact incurred) or enhanced recovery (higher product displacement) as a result of the ‘pretreatment’ provided by the first process. Alternatively, *combined* production of two or more useful materials in a single step can allow whole valorization processes, and their associated impact to be avoided.

Life Cycle Thinking (LCT) is an approach which can be used to critically assess the environmental balance of products and processes. LCT is a decision-making paradigm in which the potential upstream and downstream consequences of an action are taken into account alongside the direct consequences. A key tenet of LCT is the acknowledgement that while a given process may appear to be beneficial for the environment in isolation, there may be consequences elsewhere in the life cycle that may attenuate, cancel out, or even override these benefits. For example, from a technical standpoint, the use of a novel reagent may allow BR to be used as a source of a given metal, thus avoiding the need for

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both the extraction of the primary resource and the disposal of the BR. From a life cycle perspective, however, the environmental impact incurred through the production of the novel reagent, and the disposal of any waste streams which result from the novel extraction process may outweigh the environmental benefits associated with the avoidance of the original disposal and primary extraction processes.

Life Cycle Thinking can be used to qualitatively identify possible sources of environmental impact in a process. When enough information about a process becomes available, these can be further investigated using Life Cycle Assessment (LCA). LCA provides the scientific underpinning to LCT by allowing the life cycle environmental impacts of given interventions to be quantitatively estimated.

LCA is an internationally recognized assessment methodology for investigating the environmental impacts of complex product systems along multiple environmental axes [12]. LCA is used to assess the whole life cycle of a product, encompassing every stage from extraction of raw materials, through each of the processing and transportation stages required to transform these raw materials into products (known as a cradle-to-gate assessment), and commonly, the use of these products and their final disposal (a cradle-to-grave assessment). Using LCA, environmental impact can be assessed according to a broad range of impact categories (e.g., climate change, acidifying emissions, and toxicity measures). In addition to the total environmental impact, sources of impact within the life cycle and their relative magnitude can be identified.

In this paper, we use LCT and LCA to assess the potential environmental sustainability aspects associated with the kinds of multi-stage, multi-product, zero-waste valorization flow sheets which are being developed by the MSCA-ETN REDMUD project. In doing so, we hope to demonstrate the value of these techniques to evaluate and steer early stage technology development towards overall improved industry environmental performance.

Routes to Valorization of Bauxite Residue in the MSCA-ETN REDMUD Project

In the following sections some of the major valorization routes being developed by the MSCA-ETN REDMUD project are briefly described.

Bulk Metals: Iron and Aluminium

Iron oxide, in the form of hematite and goethite and magnetite, is the largest single constituent of BR, with concentrations commonly exceeding 40 wt% [13]. As a result, a commonly proposed valorization route for BR is the extraction of this iron via various methods. The technological feasibility

of extracting iron from BR is well established [14–16], and iron recovery is carried out on an industrial scale at some plants in China, however, a widespread economically viable, industrial scale process for iron recovery from BR, particularly European BR, remains elusive.

The options for iron recovery investigated in the REDMUD project include roasting and smelting processes. Roasting (at temperatures ranging from 500 °C to over 1000 °C [17, 18]) is used to transform the iron in BR into magnetic forms (metallic iron, maghemite and magnetite [17]). The magnetic fraction, separated using magnetic separation, can potentially be used as a feed for direct reduction to ‘sponge iron’ [14], which in turn can be used as a replacement for iron scrap in the production of steel via the electric arc furnace method. Smelting processes take place at a much higher temperature (~1500–1600 °C) and reduce the iron oxides in BR directly to metallic iron, resulting in the production of pig iron [15, 19, 20].

Another line of investigation in the REDMUD project is altering the process conditions for iron extraction to facilitate the downstream extraction of other valuable constituents of BR. This includes altering the process conditions of carbothermic reductive smelting to generate recoverable calcium aluminates for aluminium extraction [21], or to create conditioned slags for titanium and scandium recovery [22]. Additionally, both smelting and microwave roasting of BR are being investigated as a high-temperature process to prepare precursors for inorganic polymers [23, 24].

Aluminium minerals, including both incompletely digested minerals from the original bauxite and *desilication products* (complex sodium aluminium silicates produced via side reactions in the Bayer process), are present in BR at levels between 2 and 35 wt% [25]. The primary focus of the REDMUD project with respect to aluminium recovery is the generation of leachable sodium aluminates through the reaction of the remaining aluminium minerals with sodium carbonate (Na_2CO_3) [21]. Soda sintering (sintering in the presence of Na_2CO_3) of bauxite ore is used in some regions (primarily Russia and China) as a primary alumina extraction method where the silica content of the bauxite is too high for the Bayer process to be used, and is a mature industrial process [6]. The application of this process to BR is being investigated by the REDMUD project.

Minor Metals: Titanium, Scandium and REEs

Prior research has shown that the valuable minor elements of BR, including titanium, scandium and REEs, can be dissolved using various mineral acids (HCl , HNO_3 , H_2SO_4) [6]. However, acid leaching of BR also leads to the dissolution of major elements (iron, aluminium). Acid leaching of BR is further complicated by its relatively high silica content, which leads to the formation of silica gels at low acid

concentrations [26], impairing material handling and solid liquid separation. As a result, acid leaching of BR has typically been proposed with a low solid-to-liquid ratio and high acid concentration (e.g., [27]). The use of large volumes of highly concentrated acids presents a potential obstacle to industrial uptake of direct acid leaching of BR [26].

Within the REDMUD project a number of approaches to minor metal extraction dissolution are being investigated. These include pretreatment of BR to remove iron and aluminium prior to leaching [22], or the use of selective precipitation to remove these bulk elements after leaching [28]. Approaches to reduce silica gel formation, including dry digestion with mineral acids and the addition of hydrogen peroxide are also being investigated [26, 29]. The use of ionic liquids, molten salts which can act as highly selective solvents [30], is another important line of inquiry. Direct leaching of BR using ionic liquids is being investigated as a route for the extraction of valuable metals [31].

Following on from leaching, the dissolved metals must be selectively separated and recovered, most commonly in the form of oxides. In the REDMUD project, this is being undertaken using selective precipitation and solvent extraction [28, 32], column separation [33], and supported ionic liquid phases (SILPs) [34, 35].

Bulk Properties

A potential high volume application for either BR or the subsequent solid residues derived from the metal extraction techniques described above is their use in construction products. There are two main avenues of research into this area in the REDMUD project—high BR content cements, and inorganic polymers.

Three types of high BR cement clinkers have been developed within the REDMUD project: aluminoferritic, ferrobeltitic [7] and calcium sulfo-ferro aluminate [36] clinkers. Engineered slags from the carbothermic smelting of BR for iron extraction are also being investigated as a potential clinker for calcium aluminate cement.

Inorganic polymers are amorphous binders formed from the reaction of reactive aluminosilicate precursors with an alkali activating solution [37]. The low reactivity of BR means that thermal and chemical modifications are required to transform it into such a reactive precursor [8]. Within the REDMUD project, a number of processing conditions and techniques are being investigated to generate a reactive inorganic polymer precursor. This includes high-temperature processing of BR, in traditional [8] and microwave [23] kilns. As for the cements above, the use of iron depleted engineered slags as an inorganic polymer precursor is also being investigated as a potential high volume valorization route.

Assessment Methodology

Assessment Approach

The assessment in this study consists of three parts: calculation of the theoretical maximum potential life cycle environmental benefit from the valorization of BR, analysis of initial flow sheets from the REDMUD project for potentially important sources of environmental impact (red flags), and a streamlined LCA case study investigating an integrated valorization route to extract iron and produce construction materials.

Potential Environmental Savings

Displacement of conventional materials with materials recovered from BR valorization avoids the impact associated with their production, and the impact associated with the landfilling of BR. To calculate the potential magnitude of this environmental saving, the environmental impact associated with the landfilling of BR and the conventional production of five products recoverable from Greek BR (iron, aluminium, TiO_2 , Al_2O_3 and rare-earth oxides) were calculated. In addition, the impacts associated with the conventional production of two building products which can be displaced by products derived from bauxite residue were calculated: conventional cement clinker (potentially displaced by BR derived clinkers) and ordinary Portland cement (potentially displaced by BR derived inorganic polymer binders). These seven displaced products are hereafter referred to as the *avoided products*, and are listed beside their corresponding valorization product in Table 1.

The environmental impact associated with landfilling of BR, and the production of each avoided product was calculated using cradle-to-gate Life Cycle Inventory (LCI) data² taken from the ecoinvent 3 database [38] where available, and literature sources [39] where they were not. Life Cycle Impact Assessment (LCIA) results for climate change, acidification, freshwater eutrophication and freshwater ecotoxicity (using the characterization methods recommended by the ILCD³ [45]) are reported here.

Data for neodymium oxide production are used as a proxy for rare-earth oxides. Data from Nuss and Eckelmann [39] for the energy requirements of scandium extraction were

² Life cycle inventory data is compiled industrial process data collected in linked secondary databases.

³ Climate change: IPCC 100 year baseline model [40]; Acidification: Accumulated Exceedance [41, 42]; Freshwater eutrophication: EUTREND model (as implemented in ReCiPe [43]); Freshwater ecotoxicity: USEtox model [44].

Table 1 Theoretical maximum avoided burden for products from 1 metric tonne of Greek bauxite residue, according to composition reported by (Borra et al. [27])

Valorization product	Avoided product/process	Amount avoided (upper limit) (kg)	Climate change (kg CO ₂ eq)	Acidification molc H+ eq	Freshwater eutrophication (kg P eq)	Freshwater ecotoxicity (CTUe)
(All)	Landfill of BR	1000	8.1	0.08	0.34	67,774
Rare-earth oxides	Rare-earth oxides	0.23	6.3	0.04	0.00	103
TiO ₂	TiO ₂	57	325	2.69	0.14	5610
Al ₂ O ₃	Al ₂ O ₃	236	327	3.08	0.13	15,911
Iron	Iron	312	553	2.92	0.20	1900
Inorganic polymer binders	Cement	1000	903	2.10	0.07	1217
High BR content clinkers	Clinker	1000	976	2.24	0.07	549
Scandium oxide	Scandium oxide	0.186	1127	7.35	0.44	3539

used to model the potential environmental impact of conventional scandium oxide production.

The environmental impact per kg of avoided product produced was subsequently used in the ‘red flags’ analysis described in “[Semi-quantitative ‘Red Flags’ Analysis](#)”. The impact per kg was also multiplied by the amount of each product theoretically recoverable from 1 tonne of BR, based on the composition recorded by Borra et al. [27] (Table 1), to calculate the potential ‘size-of-the-prize’ associated with the various possible ways of treating 1 tonne of BR. The amounts reported represent a theoretical upper limit and do not represent the technically feasible extraction of these products.

Semi-quantitative ‘Red Flags’ Analysis

Many of the flow sheets developed by the MSCA-ETN RED-MUD are yet to be optimized at lab scale, and none have been demonstrated at industrial scale. Therefore, while the inputs required, and outputs generated from these valorization processes are known, the exact amounts are unknown. Although this means that a precise environmental balance cannot be calculated, semi-quantitative LCT approaches can be used to identify possible items of concern, or ‘red flags’, within these flow sheets. The screening process used to identify possible red flags in BR valorization is described below.

Flow sheets and descriptions of individual BR valorization unit processes were provided by researchers in the MSCA-ETN REDMUD project [46]. These were combined into a very large single, branching flow sheet (Online Appendix A), the layout of which is shown in Fig. 2. Each of the inputs to, and sources of direct emission from this combined flow sheet were identified and compiled into a list. Each item in this list was assigned to the production of one or more of the possible outputs of bauxite residue valorization, and its corresponding *avoided product* identified in the analysis above (“[Potential Environmental Savings](#)”) based on the branches of the flow sheet they were used by (for the

fully assigned list, see Online Appendix C). For example, electricity is used in all branches, and was therefore assigned to all avoided products, however, boric oxide was only used in branches producing iron (which is assumed to displace iron) and inorganic polymers (which are assumed to displace cement), and was therefore assigned to the ‘iron’ and ‘cement’ analyses. LCI data from the ecoinvent 3 database and literature sources [47, 48] were then used to assess the environmental impact of each item in this list, with LCIA results for climate change, acidification, freshwater eutrophication and freshwater ecotoxicity calculated.

For each pair of inputs and avoided products, the ratio between the environmental impact of one unit of each input, and the production of 1 kg of avoided product was calculated, for each of the four impact categories identified above, according to Eq. 1 below. This gives an ‘impact break-even point’ for each input, i.e., the use of any more than this amount of this input in a process will automatically cause the environmental balance of the process to be negative (a net environmental impact) in that category without having to take into account the impact associated with any other inputs.

$$BEP_{XY} = \frac{EI_Y}{EI_X}, \quad (1)$$

where BEP_{XY} is the break-even point for input X with respect to avoided product Y , and EI_X and EI_Y are the cradle-to-gate environmental impact of one unit of each (in a given impact category), calculated using LCI data.

An alternative way to think of this break-even-point is as the absolute maximum ‘budget’ for use of this input in a process which produces the corresponding output. The lower the break-even point, the more likely the use of this input is to significantly contribute to the environmental impact associated with a process.

For each product listed in Table 1, approximate required amounts of each input to produce 1 kg were compared

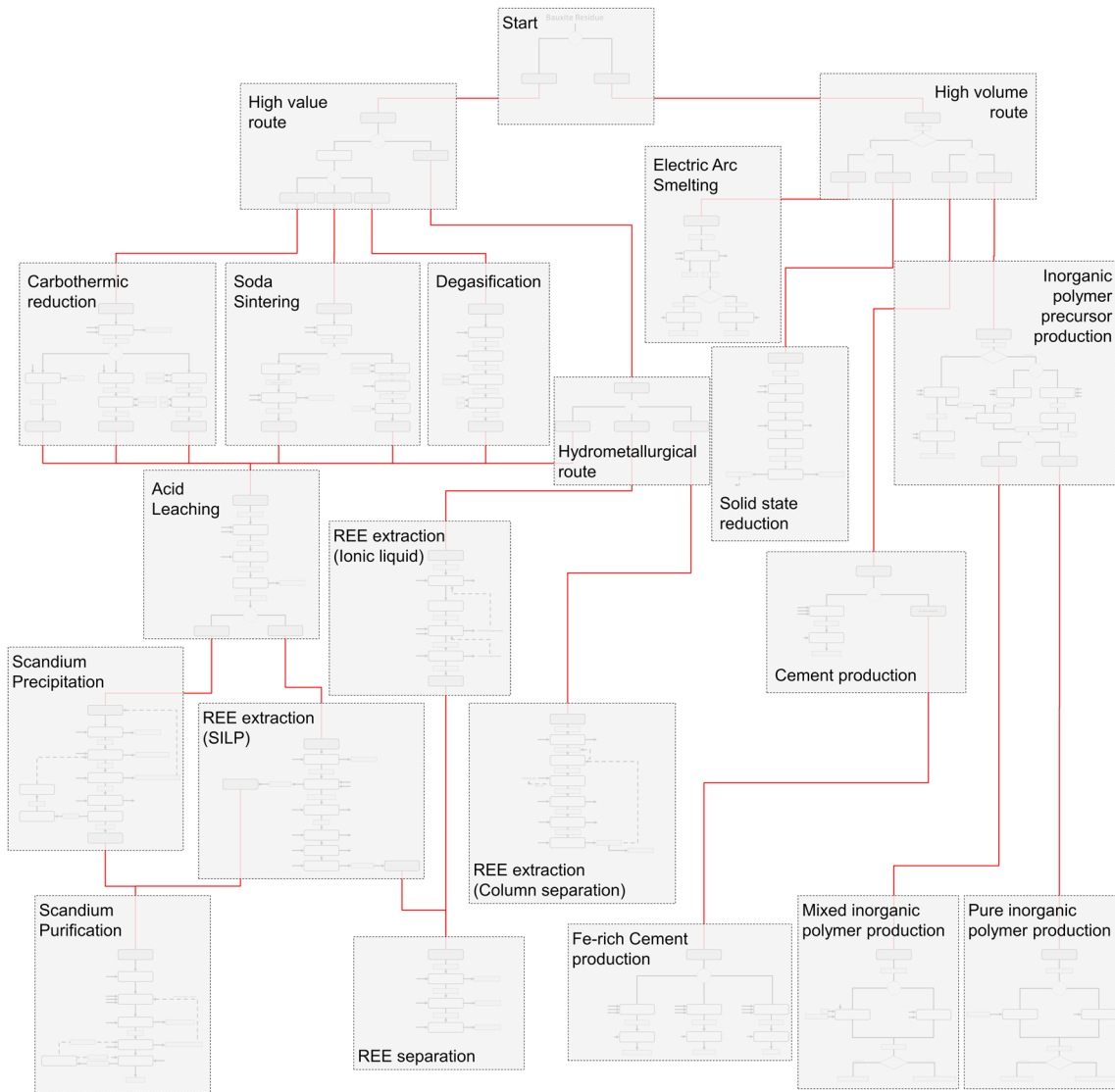


Fig. 2 Layout of combined overall flow sheet

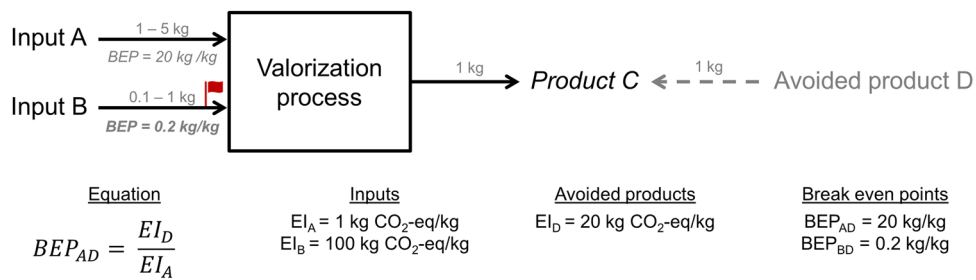


Fig. 3 Theoretical demonstration of the identification of a red flag of climate change impact (Input B) using Eq. 1. Amounts above the arrows show the approximate amount of each input assumed to

be required to produce 1 kg of Product C. BEP break-even point, EI environmental impact (Color figure online)

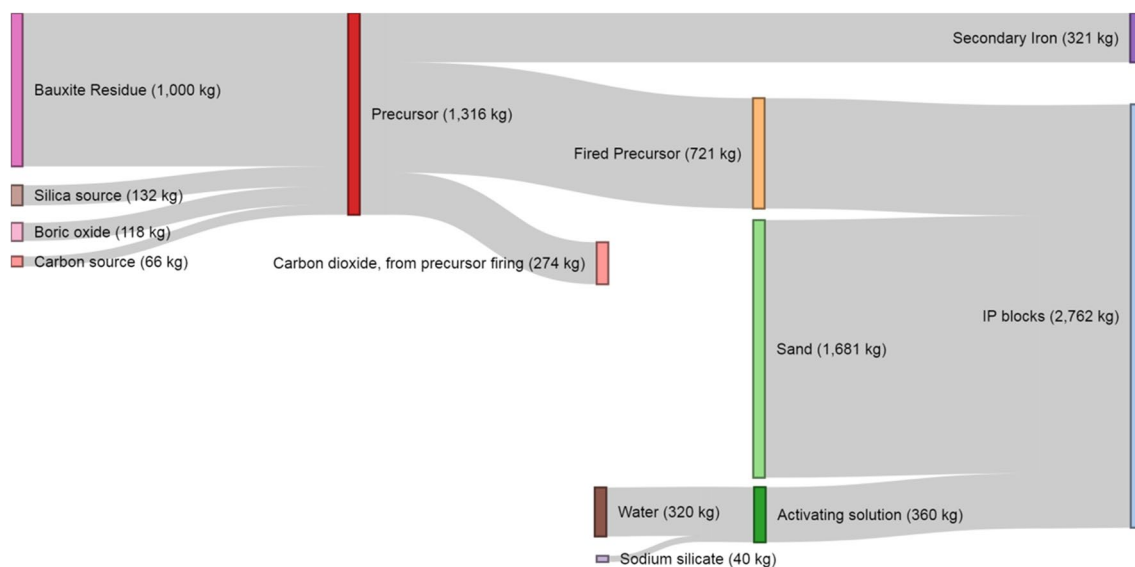


Fig. 4 Mass flow diagram for theoretical combined iron recovery and inorganic polymer production (Color figure online)

to the calculated break-even points. Where there was the potential to approach or exceed this value, these inputs were identified as potential red flags. A simple theoretical demonstration of the identification of a red flag for climate change impact is shown in Fig. 3.

Streamlined LCA Case Study

A theoretical quantified flowsheet for combined iron extraction and inorganic polymer production, based on research carried out within the REDMUD project [24], was developed to assess the environmental implications of one particular multi-stage valorization route in more detail. This represents a future, best case scenario in which lab scale experiments have been successfully scaled up to full industrial production. A full description of the life cycle inventory for the streamlined LCA study is provided in Online Appendix B; however, the system is briefly described below.

Dry BR (76 wt%) is mixed with a silica source (silica sand—10 wt%), a carbon source (graphite—5 wt%) and boric oxide (9 wt%) and the resulting mixture smelted in a rotary kiln at 1200 °C. The iron oxide in the BR is carbothermally reduced to metallic iron, consuming the added carbon, and releasing carbon dioxide. The resulting slag is separated from the iron and milled to provide a reactive precursor powder. This powder is mixed with sand (30% precursor, 70% sand) and the resulting mix is combined with a sodium silicate-activating solution, at a liquid to solid ratio of 0.15:1. The mixture is pressed using a hydraulic press and cured at 60 °C to produce solid blocks with a high compressive strength, suitable for use in paving applications.

From one tonne of BR, 321 kg of iron and 2762 kg of inorganic polymer paving blocks are created. Figure 4 shows a mass flow diagram describing the material flows through the system for one tonne of BR input. The iron produced is assumed to displace the production of pig iron, and the paving blocks are assumed to displace the production of concrete pavers. The concrete pavers are assumed to be composed of a non-structural concrete mix, consisting of 15 wt% ordinary Portland cement, 80 wt% aggregates and 5 wt% water [49]. The aggregates are assumed to be 1/3 sand, 2/3 gravel.

An LCA model was created using the software package Icopt [50] and analysed using Brightway2 [51]. LCIA results for the four impact categories used in the previous analyses were calculated.

Results and Discussion

Potential Environmental Savings

The theoretical maximum avoided burdens from the valorization of BR are presented in Table 1. This consists of the avoided impacts of landfill of BR, which accrue to each valorization route, and the avoided impacts from the production of each specific material. The sum of these impacts represents an absolute upper limit on the potential environmental benefits from a valorization route, and as such the maximum amount of impact which can be caused by any valorization process while maintaining a net environmental benefit. For valorization processes which target multiple products, these limits can be combined. As such, augmenting valorization

processes to yield products with a high-conventional impact may lead to an increased net environmental benefit even when it leads to a greater impact from valorization. For example, the combined limit for the climate change impact of iron recovery from 1 tonne of BR is 561 kg CO₂-eq. For the combined recovery of iron and scandium, however, this limit increases to 1688 kg CO₂-eq. There is therefore environmental ‘leeway’ gained from the extraction of scandium that allows augmented processes, such as the process suggested by Alkan et al. [26] for the conditioning of iron slag to facilitate scandium and titanium recovery, to have a higher environmental impact than the original process while maintaining, or even increasing, the net environmental benefit gained.

The effect is more pronounced in some impact categories than others. For example, in the freshwater ecotoxicity category, the main benefit from BR valorization comes from the avoidance of landfill (Table 1). This means that recovering additional products is likely to have a smaller effect on the magnitude of the avoided impact.

One notable point for the REDMUD project is that in the climate change impact category, building materials have a high impact relative to the recovery of other materials per tonne of BR. A prevalent idea for achieving the stated aim of *zero-waste* BR valorization is the use of final residues resulting from high-value valorization processes in the production of building products. This research suggests that there may be a sizeable climate change benefit associated with this practice.

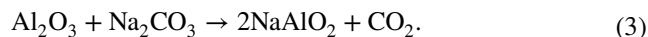
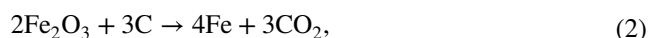
Semi-quantitative ‘Red Flags’ Analysis

The environmental break-even points for the inputs to the iron, alumina, rare-earth oxide and cement flow sheets are shown in Fig. 5. The values in this figure are the minimum break-even point across the four impact categories assessed, such that below this value, the environmental balance is net benefit in all four impact categories. The full results of this assessment, broken down by impact category, can be found in Online Appendix C. These break-even values can be combined with even very basic knowledge of the amount of each of these inputs that may be required (e.g., order of magnitude) to produce 1 kg of the desired outputs to identify possible red flag inputs. For example, alumina reacts with Na₂CO₃ in a 1:1 molar ratio, and their molar masses are similar (102 vs. 106 g mol⁻¹), therefore, in the absence of any other information, we know that at least 1 kg of Na₂CO₃ will be required to produce 1 kg of alumina. The Na₂CO₃/alumina break-even points are 0.6 kg for climate change, 1.0 kg for acidification, 0.45 kg for freshwater ecotoxicity and 2.1 kg for freshwater eutrophication, suggesting that the use of Na₂CO₃ in alumina extraction is a significant red flag in at least the first three of these categories. If the aluminium

hydroxide is recovered from the sodium aluminate via a carbonation process, rather than a precipitation process as used in the Bayer process, a significant amount of the Na₂CO₃ can be recovered. However, the production of industrial carbon dioxide carries a climate change impact of around 0.75 kg CO₂-eq per kg, which somewhat lessens the benefit of Na₂CO₃ recovery.

Several processes in the combined flowsheet (Online Appendix A) for the valorization of BR were identified as red flags on the basis of their break-even points and the approximate amount required. In some cases, for example leaching and precipitation reagents where the optimal concentration, regeneration efficiency and extraction efficiency are currently unknown, these amounts are limited to a representative order of magnitude. These *red flags* broadly fall into four main categories: direct emissions from chemical reactions, chemical synthesis, thermal and mechanical energy use; and are summarized in Table 2.

Direct emissions from chemical reactions include the release of carbon dioxide from the oxidation of carbon in the carbothermic reduction of iron oxide to metallic iron (Eq. 2) and the decomposition of sodium carbonate in the soda sintering process (Eq. 3).



These emissions are an intrinsic part of the chemical processes of iron and aluminium extraction and cannot realistically be avoided or abated. Alternative reducing agents for iron reduction exist, for example hydrogen [52] or biomass pyrolysis biochar [53], but currently are typically expensive or impractical. For the extraction of aluminium via soda sintering, the stoichiometry of the chemical reaction sets an absolute minimum value for CO₂ emissions of 0.431 kg of CO₂ for every kg of Al₂O₃ in BR. In Europe, Na₂CO₃ is typically produced via the Solvay process, which requires the calcination of limestone using high carbon fuels such as coke, coal and lignite [54], compounding the environmental issues associated with this reagent.

Carbon capture and storage (CCS) may represent a future option for managing emissions of CO₂ from chemical processes. Commercial scale applications of CCS are currently limited to the power sector [55], in which fuel combustion rather than process chemistry is the primary source of CO₂ emissions. However, CCS is an active field of research within the cement industry, in which chemical processes account for around 2/3 of CO₂ emissions [56]. CCS technologies are being trialed at pilot scale at multiple cement plants [56]. The environmental effects of the (often significant) increased energy demand associated with the use of CCS would need to be considered alongside the benefits of CO₂ capture were this option to be implemented.

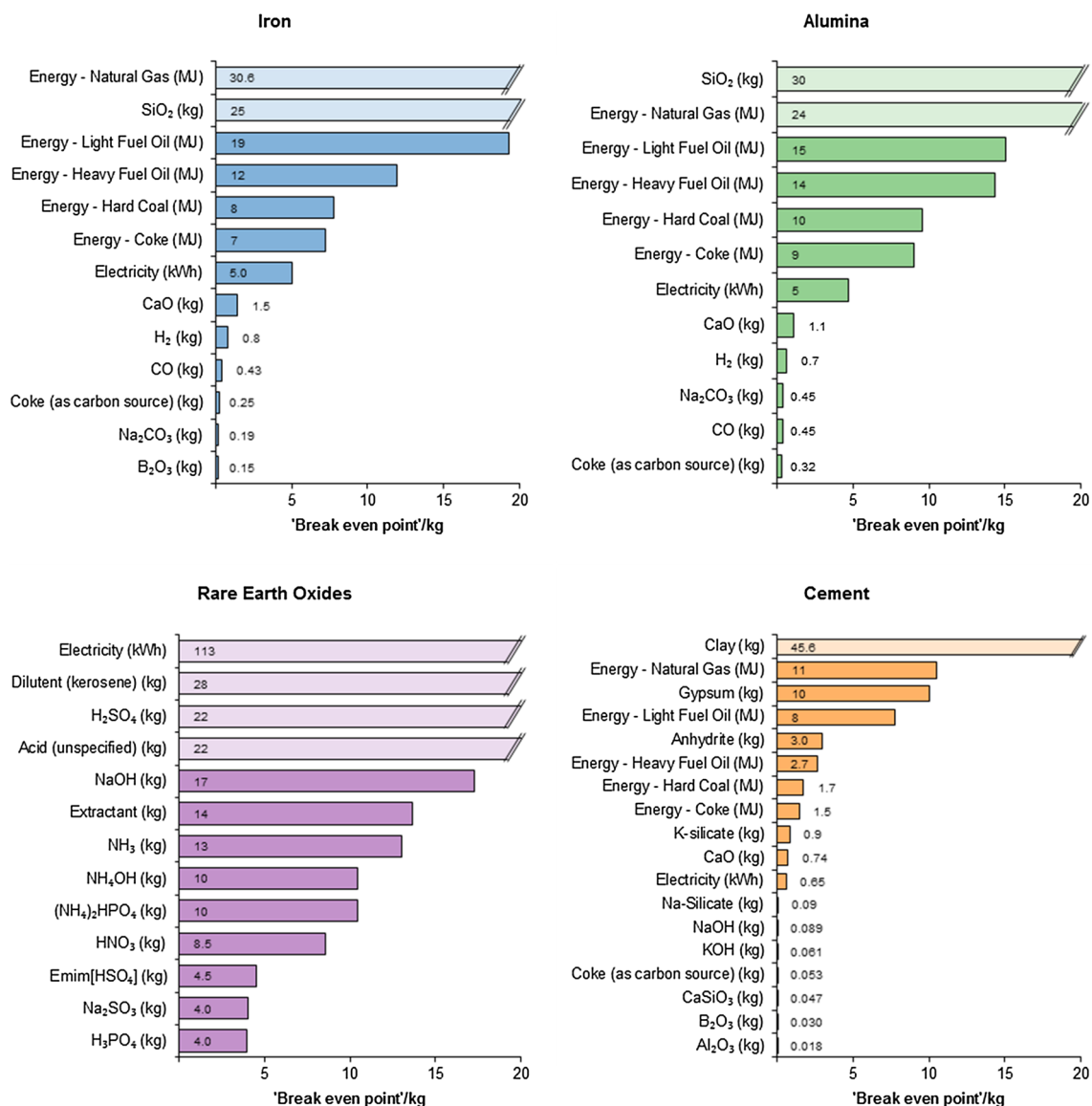


Fig. 5 Break-even values per kg of avoided product for iron, alumina, rare-earth oxides and cement. The break-even values are the minimum value across the four impacts assessed. Items with a break-even value > 20 are shown in a lighter shade (Color figure online)

The production of many of the chemicals utilized in the extraction of metals from BR (strong acids, alkalis, ammonium compounds, hydrocarbon solvents) is associated with environmental impacts. Among the chemical synthesis category, the synthesis of ionic liquids is worthy of note in terms of their potential for environmental impact, due to the complex multi-stage process of organic synthesis involved in their production. Ionic liquids are regenerated as part of the metal recovery process, however, there are inevitable losses. The overall break-even point for Emim [HSO₄], a candidate ionic liquid for rare-earth element extraction from BR is 4.5 kg/kg. This suggests that it is vital from an environmental perspective that losses of ionic liquid remain under 4.5 g per gram of metal oxide produced. By way of comparison,

the overall break-even point of sulfuric acid—the conventional reagent used in acid leaching—with respect to rare-earth oxides is 22 kg/kg.

Thermal energy is required as an input in many of the proposed valorization routes, particularly the extraction of iron and aluminium and the production of building materials. For metal extraction, this thermal energy is required to drive the appropriate metallurgical reactions. In the case of building products, it is required to increase the reactivity of the minerals within the BR by physical and chemical means.

Mechanical energy is also required by many of the valorization routes, primarily for crushing, milling and grinding. These are often intrinsically linked to the efficiency of the process, for example in creating surface area for reactions,

Table 2 Potential red flag processes in the valorization of bauxite residue

Route	Important processes	Input	Approximate amount required (per kg product)	Range of break-even points
Iron recovery	Electric arc furnace smelting	Thermal energy	0–5 kWh	5 kWh
		Oxidation of carbon source	0.1–0.5 kg	0.3 kg
	Carbothermic reduction	Thermal energy	0–20 MJ	7–19 MJ
		Oxidation of carbon source	0.1–0.5 kg	0.3 kg
Aluminium recovery	Fluxing	Boric oxide	0.3–1 kg	0.15 kg
		Magnetic separation	Mechanical energy	0–10 kWh
	Soda sintering	Thermal energy	0–20 MJ	9–24 MJ
		Decomposition of Na ₂ CO ₃	> 1 kg	0.45 kg
Metals leaching (direct and secondary slag leaching)	Acid leaching	Acids	0–10 kg	4–22 kg
	Alkali leaching	Alkalis	0–10 kg	17 kg
	Ionic liquid leaching	Ionic liquids	0–10 kg	4.5 kg
Extraction and purification of metals	Solvent extraction	Solvents	0–100 kg	14 kg
		Diluents	0–100 kg	28 kg
	Stripping	Acids	0–100 kg	4–22 kg
		Ammonium compounds	0–100 kg	10–13 kg
Direct use in building products (Inorganic polymers)	High-temperature processing of precursors	Thermal energy	0–20 MJ	2–7 MJ
		Milling	Mechanical energy	0–1 kWh
	Preparation of activating solution	Alkalis	0.15–0.25 kg	0.06–0.86 kg
Use of secondary slag in building materials (Inorganic polymers)	Preparation of activating solution	Alkalis	0.15–0.25 kg	0.06–0.86 kg
Direct use in building products (Cements)	Clinkering	Thermal energy	0–20 MJ	2–7 MJ
		Mechanical energy	0–1 kWh	0.65 kWh
Use of secondary slag in building materials (Cements)	Milling	Mechanical energy	0–1 kWh	0.65 kWh

or breaking down agglomerates to release magnetic iron fractions for magnetic separation. In a future, aspirational process, this mechanical energy could be provided by renewable means, however, in the short term, the environmental impacts of these mechanical processes need to be taken into account.

Streamlined LCA Case Study

The iron/inorganic polymer case study highlights the importance of all four of the red flag categories mentioned above. Figure 6 shows the major contributors to the impacts incurred by the valorization pathway. Direct emissions of carbon dioxide from the carbothermic reduction of iron are a significant contributor to the climate change impact, accounting for 36% of the total impact of the valorization system. Chemical synthesis, in particular the production of boric oxide, is also a substantial part of the impact incurred by the valorization pathway. The production of boric oxide is the second largest contributor to climate change impact (34%), and the largest single contributor to acidification (73%), freshwater eutrophication (68%) and freshwater

ecotoxicity (77%) impact categories. The production of sodium silicate also makes a noteworthy contribution to freshwater eutrophication (9%) and freshwater ecotoxicity (8%).

Thermal energy, for the high-temperature smelting of the augmented BR mix, makes a significant contribution to the climate change (16%) and acidification impact (17%) categories, through the production of fuels and the emissions released by their combustion. Mechanical energy for milling, pressing and kiln rotation, provided by electricity, can be seen to contribute to the freshwater eutrophication impact of the valorization pathway, making a combined contribution of 12% of the total impact, second only to the production of boric oxide.

Figure 7 shows the overall net environmental balance of the iron/inorganic polymer valorization pathway, including both the impact incurred and the impact avoided through the displacement of the requirement for landfilling of BR and the primary production of pig iron and concrete pavers.

For climate change, freshwater eutrophication and freshwater ecotoxicity, there is a net environmental *benefit* from the valorization of BR. The avoided production of pig iron

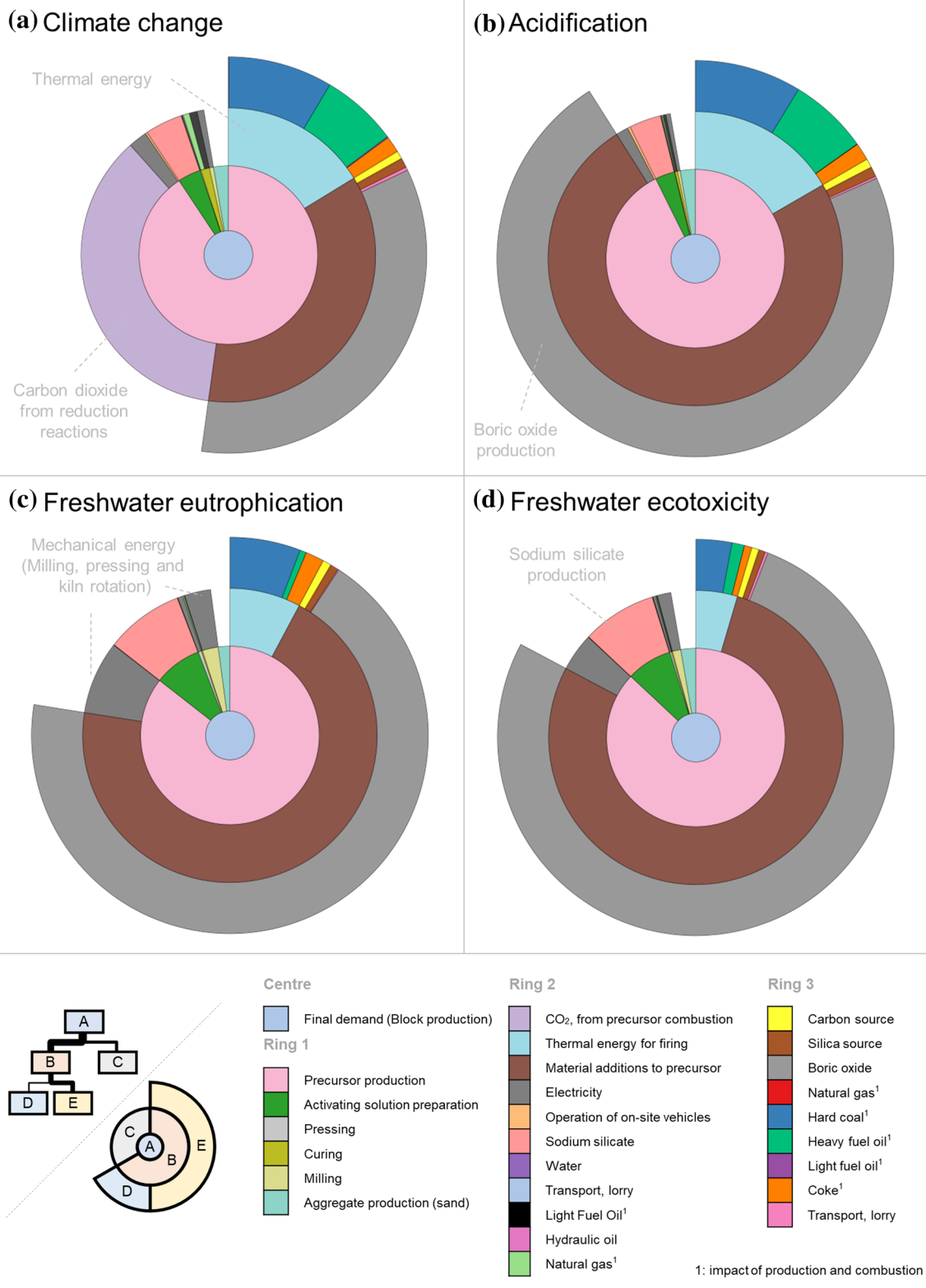
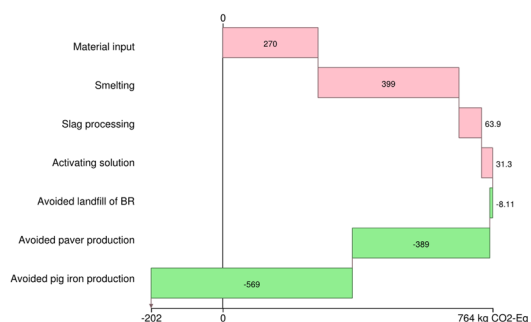


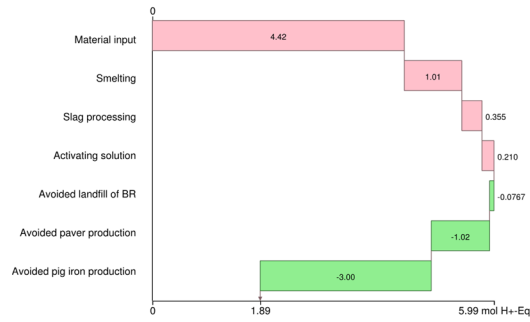
Fig. 6 Bulls eye hotspot diagrams for each of the four impact categories considered. These are hierarchical pie charts, with and should be read as outlined in the diagram to the left of the key. Each ring represents

a level of the system tree, with the arc angle representing the contribution to overall impact (as in a traditional pie chart). Selected hotspots are shown as annotations (Color figure online)

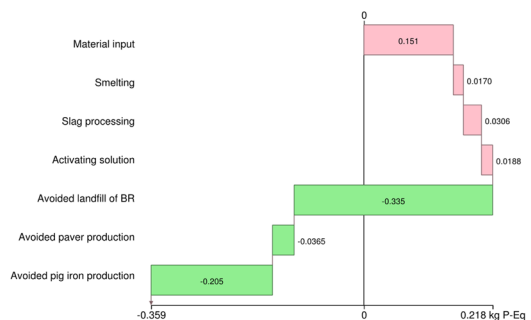
(a) Climate change



(b) Acidification



(c) Freshwater eutrophication



(d) Freshwater ecotoxicity

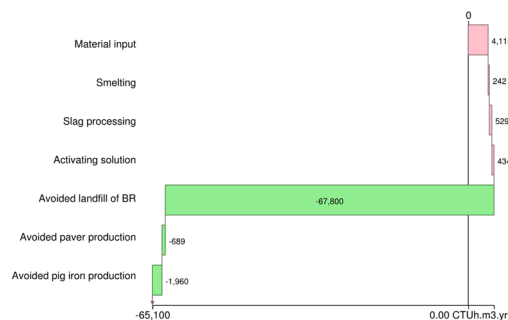


Fig. 7 Net environmental balance of a combined iron extraction and inorganic polymer synthesis valorization route for four environmental impact categories, broken down by contributing processes. Red bars indicate incurred impact from valorization processes, green bars indi-

cate avoided impact through the displacement of products and processes. Total net impact is indicated by the arrow and value on the x-axis (Color figure online)

and pavers are the most significant aspects of the climate change benefits of the valorization pathway (Fig. 7, panel a). For the freshwater categories, the most significant aspect of this environmental benefit is the avoidance of landfilling of BR (Fig. 7, panels c and d), particularly in the case of freshwater ecotoxicity. For freshwater eutrophication, the avoided production of pig iron also results in a substantial environmental benefit.

For climate change, the net benefit depends on all three avoided aspects to be realised. The addition of boric oxide (as a boron source) is the most significant difference between this combined process and a single-step process producing only inorganic polymers (such as that presented in Joyce et al. [48]). A source of boron is added to the BR to enhance the separation of metallic iron from the glassy slag produced in smelting. The climate change impact of the high temperature processing, slag processing, and activating solution steps, which would remain broadly unchanged in a single step process for producing inorganic polymers, are higher than the impact which could be avoided through paver production (Fig. 7, panel a). Similarly, the impact resulting from the production of boric oxide, and the high-temperature

smelting step, which would be required to produce iron alone, is greater than the impact which could be avoided by the production of pig iron (Fig. 7, panel a). However, because the high-temperature processing step is common to both iron and inorganic polymer production, the impact of the combined valorization process is lower than the avoided impact through the displacement of pig iron and pavers, resulting in a net environmental benefit. This is significant; as it means that from a climate change perspective a multi-output process is necessary to achieve an overall net benefit.

For acidification there remains a net environmental impact from the combined valorization process (Fig. 7, panel b). This is primarily due to the boric oxide requirement and the energy and emissions from smelting.

The substantial contribution of boric oxide to all impact categories highlights a potential issue with the detailed environmental assessment of emerging technologies only proven at lab scale. Boric oxide is, in this case, an analytical grade reagent, and while it is required to demonstrate proof of concept it is unlikely to be the actual boron source used at industrial scale. Importantly at this stage, the addition of boric oxide as a fluxing agent demonstrates that it is possible

to combine iron smelting and inorganic polymer precursor generation in a single step from which it is possible to recover both products. This opens the door to future research into the identity and quantity of a fluxing agent which could optimise the process both technically and environmentally.

Altering a process to reduce its environmental impact may also, however, reduce its efficiency, which in turn reduces the benefit from avoided production. Changing the process conditions to reduce the amount of fluxing agent required may lead to suboptimal iron recovery, but if the percentage decrease in iron recovery is lower than the percentage decrease in fluxing agent use this may lead to increased environmental benefits. It is therefore important to quantify this balance.

A potentially promising option is the use of boron waste (from the mining and refining of borax) as a substitute fluxing agent. This has the additional benefit of constituting a use for another industrial waste [57]. Substantial and growing stockpiles of boron waste exist in Turkey and China [57, 58]. Again, the environmental impacts associated with the extraction and transport of this waste, and any concomitant reduction in the efficiency of the process or the quality of the outputs would need to be accounted for in the environmental balance.

Conclusions

Assessing the potential environmental impact of emerging technologies in research and development is challenging. The completeness and accuracy of available information increases as the technology develops, but the scope to make changes to the process design decreases at the same time [59]. Therefore, the earlier in the process that environmental information can be taken into account, the more impact it can have on decision-making. In this paper we have demonstrated that a Life Cycle Thinking approach can provide useful insights into the potential environmental balance of valorization processes even in the absence of detailed information about the particulars of the process.

At the earliest possible stage, an appreciation of the possible environmental benefit which could be realized from displacing the production of different target materials can help shape the overall aim of a research project; for example, the possibility of tailoring of residues from metals extraction for use in construction materials as a way to reduce climate change impact.

The red flags assessment demonstrates that once early draft processes have been developed, and possible inputs have been chosen, potential hotspots of environmental impact can be identified, even in the complete absence of quantitative data about the process. This can act as a screening stage, either to disqualify certain inputs, or to direct

research towards finding substitutes. It can also help direct research towards enhancing the recovery of environmentally significant inputs, for example ionic liquids.

As the technology develops, and basic quantified flow sheets become available, streamlined LCA studies such as the case study carried out here, can provide additional, more specific insights into the potential hotspots of environmental impact that exist in the process. This can point to specific areas in which the process can be altered to reduce the impact incurred.

The streamlined LCA study presented here also validates a number of the higher level inferences that were gained from the first two analyses. Most importantly, it demonstrates that combining processes to yield multiple products can have a significant environmental benefit over single product valorization pathways. The combined iron/inorganic polymer process had a net benefit in terms of climate change, which would not have been the case for either of the single product valorization routes. It also confirmed that red flags identified at the screening stage can translate to hotspots of impact in the more detailed analysis. Chemical synthesis and thermal energy provide the best examples of this.

The results presented in this paper suggest a number of important considerations which researchers developing valorization pathways for bauxite residue should take into account. In addition to paying attention to the specific red flag reagent and processes identified here, three suggestions stand out. First, synergies are important. Combining valorization processes, either as a pretreatment or as a combined, multi-output single step can lead to environmental benefits. At the very least, the modification of process residues for use in construction materials should be considered. Second, reagent recovery has important environmental implications. The synthesis of chemical reagents required to extract metals from bauxite residue can lead to significant environmental impacts, and as such minimizing the amount required is an important consideration. Once proof of concept for the use of a given reagent has been established, optimizing the recovery of this reagent should be prioritized during the scaling up process. Third, even quick streamlined environmental assessments can yield important conclusions. Such assessments should be incorporated into the technology development process to help identify both important hotspots of impact and new avenues of research.

The methods presented here can make a useful addition to the suite of assessment methodologies that should be used to guide the future valorization of bauxite residue. These include technical and economic assessments (e.g., [60]), as well as the assessment of Naturally Occurring Radioactive Materials (NORM) in the valorization process, both from a process perspective [61] and a product life cycle perspective [62, 63]. An additional side benefit of incorporating an environmental perspective into the early stages of research

and development, specifically in European Training Network projects such as REDMUD, is that it fosters environmental and life cycle thinking among researchers who may not have previously been trained in this area. These early-stage researchers can then take this approach forward into their careers.

The sustainable management of bauxite residue is a key aspect of the long-term future of the alumina sector. The current evidence suggests that innovative valorization solutions will be a burgeoning area in this respect. The success of the first two *Bauxite Residue Valorisation and Best Practices* conferences in 2015 and 2018 are testament to this fact. Close collaboration between academia and industry in the tailoring of valorization processes to specific refineries, alongside a clear and explicit focus on the environmental aspects of bauxite residue valorization, are likely to important features of future research in this area. The commitment demonstrated through the EU Horizon 2020 RemovAl project [64], which began in 2018, and the inclusion of plans for the environmentally sustainable valorization of bauxite residue at the new EGA alumina refinery in the UAE prior to startup [65] are just two examples of the bright future which potentially lies ahead. The methods presented in this paper have the potential to help provide a scientific basis for the assessment of environmental aspects of future bauxite residue valorization schemes.

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