A Sustainable and Profitable Bauxite Residues Valorization Process

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Abstract

Globally, the production of alumina generates about 175 million tonnes of bauxite residues (BR) per year, containing valuable metals such as Sc and REE, that are currently accumulating in storage ponds. With the lack of storage space, stricter environmental regulations, and the surge in REE demand, the need for a sustainable approach to valorize these residues, while minimizing waste volume is imminent. The avenue of BR valorization has been explored for decades but no approach has been successfully commercialized yet. Many routes have been studied and proposed but they either lack economic viability, and or do not reduce the waste volume sufficiently to make them attractive. In other words, environmental impacts are shifted towards effluents or other harder-to-manage waste.

A successful approach should reduce the waste volume significantly by recovering the value of both bulk (Fe, Al) and critical (Sc/REE) constituents. So called “zero-waste” approach to the BR valorization has been proposed to recover multiple compounds and to minimize waste volume and get the most value out. Main proposed processes are based on direct acid leaching and/or iron smelting, each facing limitations and serious technical challenges yet to be addressed. Direct leaching with acids creates large effluents and is expensive owed to low selectivity and extensive separation steps. Iron smelting presents technical challenges related to sodium and aluminium content in BR.

This paper provides an overview of the BR valorization approaches. Focus is placed on the proposed process by Innord Inc. which targets the recovery of bulk metals (Fe, Al) and generating valuable metal concentrate (Sc/REE), maximizing volume reduction (+70 %) and improving the process economy by producing several low value marketable by-products. Other objectives of the development are having limited number of steps, major regent recycling avoiding any regular effluent as much as feasible. The process steps consist of alkalinity and DSP removal, iron conversion and separation, and Sc/REE concentrate and reagent recycling.

Keywords: Zero-waste, Rare-earth elements, Valorization, Sustainable, Bauxite Residues.

1. Introduction

In the context of the fight against climate change, the demand for light metals such as aluminium is increasing, especially for the manufacturing of vehicles with lower fuel consumption. While this will result in decreasing greenhouse gas (GHG) emissions, aluminium production creates large amounts of waste. Bauxite residue, also known as red mud, is a major waste resulting from the alumina extraction from bauxite, mainly via the Bayer process. It is a mineralogically complex mixture composed of undigested solids from the high-pressure bauxite digestion in a hot caustic soda (NaOH) solution and precipitated aluminosilicates (a.k.a. desilication product or DSP). Up to 2 tonnes of bauxite residue are produced per tonne of alumina. BR is usually found in the form of slurry containing a variable percentage of solid residue from the process, typically between 20 and 40 wt. %.

Large spaces are required for BR stockpiling, and the extension of storage facilities necessitate periodic capital investments and permitting. In general, environmental issues associated with bauxite residue storage and disposal include its high pH (11-14), the potential for alkali (Na, K) and ecotoxic (V, Al) metals seepage into groundwater, instability of storage and the impact of alkaline airborne dust on plant life.

The annual production of alumina in 2020 was over 133 million tonnes resulting in the generation of about 175 million tonnes of BR [1]. Currently, about 97 % of this production is stockpiled, leading to over 4 billion tonnes accumulated globally to date [2]. The global volume of the stored BR is expected to double in the next decade if the rate of utilization remains the same [3].
Moreover, BR is rich in iron (Fe), aluminium (Al), titanium (Ti), as well as strategic and/or critical metals such as vanadium (V), scandium (Sc) and other rare-earth elements (REE) that are discarded during stockpiling. Assuming a 100 USD/t of BR metal value, $17.5 billion worth of material is being stockpiled annually. Valorizing BR is therefore an opportunity for the alumina refineries to achieve their waste reduction targets and take advantage of the metal value of BR.

The typical composition of BR (dry basis) is given in Table 1 below.

Table 1. Composition of dry BR (oxide basis) [4].

Oxide	**Fe_2O_3**	**Al_2O_3**	**Na_2O**	**CaO**	**TiO_2**	**SiO_2**	**V_2O_5**	**Sc***	**La***	**Ce***	**LOI**
wt. % or ppm*	30-60	10-20	6-12	2-5	5-10	5-15	0.1-0.15	21-54	20-147	30-285	5-15

A typical mineralogy of BR is given in Table 2 below. Some variations in the composition are expected, depending on the type of bauxite treated at the alumina refinery.

Table 2. Mineralogy of BR feed (adapted from [4]).

Mineral	**Chemical formula**	**Typical range wt. %**
hematite	α-Fe_2O_3	7-29 %
Goethite	α -FeOOH	7-24 %
Boehmite	AlOOH	1-10 %
Aluminated goethite	$Al_xFe_{1-x}OOH$; x=0…1	10 %-40 %
Quartz	SiO_2	0-2 %
Rutile/Anatase	TiO_2	0-5 %
Bayer sodalite (DSP)	$Na_8[Al_6Si_6O_{24}][(Y)]\cdot xH_2O$; Y= $2Cl^-$, $2OH^-$, CO_3^{2-}	2-24 %
Cancrinite	$Na_6[Al_6Si_6O_{24}]_2CaCO_3$	0-51 %
Tricalcium aluminate	$3CaO\cdot Al_2O_3\cdot xH_2O$	0-5 %
unknown	V_2O_5	<1 %
unknown	REE (Sc, Ce, La…)	0-400 ppm

The avenue of BR valorization has been investigated for decades and to this day there is no implemented large-scale process that can extract these metals and effectively reduce waste volume. Moreover, the project feasibility might not be assessed based on the price of the pure metal oxides, unless all the purification steps toward the market specification are included in the process, or conservative discounts are applied. Besides, the environmental impacts of an alternative solution to BR stockpiling should not be displaced by generation of other effluents or solid wastes that are equally or even harder to manage. This would delay the commercialization of the process owing to supplemental regulation constraints.

This paper gives an overview of the main BR valorization approaches and discusses their limitations and the processing challenges. Then, the proposed Innord's R&D process is explained and discussed. The main focus of this study is on valorization processing strategies rather than remediation & rehabilitation.

2. BR Valorization Approaches

The main approaches proposed to extract the metal value of BR are briefly described in this section.

Elements of interest are:

- Iron: main component in the BR and is a major contributor to waste volume reduction and potential revenues.
- Sc/REE: to increase profitability and provide a secure supply of these critical and strategic materials (CSM).
- Aluminium: Technologically important for lightweight vehicles and aircrafts. The aluminium content represents important losses during the Bayer process (about 20 %). Aluminium losses in BR should increase as lower-grade, higher-silica bauxites are processed, signifying higher losses via desilication.
- Titanium: Strategic metal. Titanium high-grade ores are less frequent and an enriched TiO_2 residue could be used for subsequent extraction.

2.1 Sc/REE Recovery

Sc/REE are CSM and play an essential role in defense applications, and in the transition to a low-carbon and digitized economy. These elements are qualified as critical because their production is dominated by one or a few countries, which subjects manufacturers to supply risks and price fluctuations.

These elements are present in bauxite at ppm levels. After alumina extraction, REE/Sc are enriched at a factor of two [4]. Extraction of these metals from BR is attractive considering the enrichment factor and the high price of rare-earth oxides.

Early approaches consisted in direct leaching of BR using strong mineral acids (such as HCl, HNO_3, H_2SO_4) followed by separation using solvent extraction [4]. Achieving a high recovery of Sc/REE requires breaking down the iron and aluminium minerals in which they are occluded. In addition, large volume of effluents is created if the leaching reagent cannot be effectively recycled. Besides, leaching in concentrated mineral acid requires reactor vessels made from specific alloys which increase capital costs. Another technical challenge with the direct acid leaching of BR is the formation of a supersaturated silicic acid solution that polymerizes at a low rate and forms a gel in low pH (1-3) [5, 6].

Main Approaches are:

- Sulfuric acid baking [7]: in this process, the BR is mixed with 98 % sulfuric acid at an acid/BR ratio of 1-2 and is heated to high temperatures (500-700 °C) to convert the REE/Sc to water-soluble sulfates. The baked residue is leached with water. Besides,

sulfuric acid baking is a known technique to fix reactive silica and converted it to an acid-insoluble form [8] which avoid the gelation and subsequent filtration issues [7]. Acid baking prior to water leach increased the REE/Sc extraction rate with a reduced overall acid consumption compared to direct acid leaching. The disadvantages of the baking process are SO_x generation requiring capture and an iron-rich acidic residue that requires additional treatment prior to disposal, or additional steps for the extraction of the iron value.
- Sodium bicarbonate leaching. This process is currently used at the commercial scale by RUSAL [9], to selectively extract scandium from BR. The main principle of the process is based on the formation of scandium bicarbonate water-soluble compound. This is achieved by bubbling CO_2 in the alkaline solution present in the bauxite residue to form $NaHCO_3$ that dissolves scandium selectively against the other REE as lanthanides do not form soluble bicarbonates. After multiple processing steps (sorption/desorption on an ion-exchange resin, hydrolysis) scandium is concentrated to obtain a 99 % pure oxide. Other REE do not form soluble bicarbonates as readily as scandium and are thus not recovered.

While the Sc/REE extraction from BR can be profitable, depending on the grade of the feed, it must be incorporated in a comprehensive metal extraction flowsheet. Otherwise, waste volume reduction is economically marginal, and the sustainability problem is not solved.

2.2 Iron Recovery

Since the iron content of BR can reach up to 50 wt. %, iron recovering is the key to reducing waste volume significantly.

Different approaches, mainly derived from conventional iron pyrometallurgy, have been considered to separate and valorize this element, with the advantage of reducing the overall acid consumption on the following hydrometallurgical section.

The most investigated route is to smelt the BR to produce metallic iron and a slag containing oxides of aluminium, titanium and REE. The slag could then be acid-leached to recover REE and/or aluminium values. However, smelting in a blast furnace presents several technical challenges such as:
- The sodium content in BR can damage the refractories of the furnace by the formation of sodium nests [4].
- The content of aluminium oxide makes the slag more viscous than in conventional operations. This in turn increases energy and/or fluxes consumption to maintain an acceptable fluidity [10].

Therefore, iron smelting using BR was not commercially successful owing to increased operation costs compared to using conventional feeds.

The solid-state reduction of iron has been considered as well to either obtain magnetite or DRI (Direct Reduced Iron) that can be isolated from other materials via magnetic separation. The separation efficiency of the magnetic separation step is a known challenge owing to the coexistence of different elements in minerals [11,12]. Sc/REE losses in the iron fraction can impact the recovery of these elements. Besides, contamination of the iron fraction with gangue minerals may affect the selling price of the product obtained.

2.3 Alumina and Sodium Recovery

The alumina content in BR may reach 20 %. This represents about 20 % losses in the Bayer process assuming BR to alumina mass ration of 1:1. The alumina in the BR is either in an aluminosilicate form, mainly in the desilication product (DSP) or in non-digested minerals from the original bauxite, such as boehmite (AlOOH). The loss of aluminium to aluminosilicates is likely to increase with the growing usage of low grade, silica-rich bauxite ores.

The aluminosilicate portion can be leached in acids, but reactive silica is extracted and in acidic solutions silicic acid is formed, which polymerizes and causes gelation issues in the process [7]. The high-temperature (T > 260 °C) and high alkalinity ($Na_2O:Al_2O_3 > 10$) variant of the Bayer process was tested on BR with the idea to recover alumina from the more stable phases. This approach showed limited leaching efficiencies [10]. The presence of aluminium substituted in goethite, as it has been observed in BR, can explain the limited recoveries. The overall recovery of aluminium is dictated by the partition in different phases: Al-substituted goethite, DSP, and hydrated alumina phases like boehmite or gibbsite.

One solution is to circumvent these issues is to convert the alumina to a leachable form. The Deville process or a variation thereof consists in alkali roasting at T= 800-1200 °C using Na_2CO_3 to form sodium aluminate leachable in water according to the following reactions [14].

$$Na_2CO_3 \rightarrow Na_2O + CO_2 \tag{1}$$

$$Al_2O_3 + Na_2O \rightarrow 2NaAlO_2 \tag{2}$$

The sodium aluminate solution can then be sent to Gibbsite precipitation, either via seed-induced crystallization (Bayer) or carbonatation (Pedersen) [10].

$$NaAlO_2 + 2H_2O \rightarrow NaOH + Al(OH)_3 \tag{3}$$

$$2\,NaAlO_2 + 3\,H_2O + CO_2 \rightarrow Na_2CO_3 + 2\,Al(OH)_3 \tag{4}$$

To prevent the formation of soluble silicates, lime is added to form insoluble $CaSiO_4$. In some variations of this process, coke or other carbonaceous materials are used to produce metallic iron and prevent the precipitation of sodium and aluminium ferrites. The iron must then be separated via magnetic separation, which bears the limitations mentioned above.

While the process allows to recover the sodium value of the BR, by forming a caustic solution or soda ash that can be recycled in the process, the energy consumption is two to three 3 times higher than the Bayer process.

2.4 Multi-product Valorization

There has been a growing interest for “complete” or “zero-waste” valorization of BR. The recovery of as many components as possible can minimize waste and get the most value out of the feed. The recovery of multiple products has many advantages such as:

- Significant volume reduction which is the key element to solve the sustainability problem, ensuring long-term viability of alumina refineries.
- Distribution of the process environmental impacts to multiple by-products [15].
- Selling multiple products allows to maximize revenues and to average market price fluctuations, especially when more volatile commodities such as Scandium and REE are involved.

Different combinations of pyrometallurgical and hydrometallurgical steps have been proposed in the literature [8]. Borra *et al.* [16] presented multiple combinations of iron smelting or reduction roasting/magnetic separation followed by slag or non-mag fraction leaching to recover the REEs and possible alumina. To increase the revenues, reduce the acid consumption, and improve the smelting performance, an alumina recovery step, such as alkali roasting/water leach, can be performed prior to smelting. The most promising option, according to the authors [16] is the combination of alkali roasting – smelting – quenching – leaching. However, the CAPEX is not considered in this preliminary techno-economic evaluation.

The Orbite process [17] consists of pressurized leaching of BR in concentrated hydrochloric acid. Aluminium, iron as well as REE and the alkaline elements (Na, Ca, K…) are leached while leaving behind a silica residue [15]. Aluminium is recovered by saturating the solution with HCl to induce crystallization of aluminium chloride hexahydrate that can be thermally decomposed to form Al_2O_3 and regenerate HCl. The iron chloride-rich solution is hydrolyzed in hydrothermal conditions (150-170°C) to precipitate hematite. REE are recovered from the solution using solvent extraction. The difference with direct acid leaching, is that the various metal recovery steps allow to recover HCl to reduce the reagent consumption and reduce effluents. Considering the price of HCl, it must be regenerated from alkali and alkaline-earth chlorides for the process to be economically viable. Sulfuric acid is used to precipitate calcium sulfate from calcium chloride. In the process presented, it is envisioned that HCl is regenerated from sodium chloride by the chlor-alkali process.

Several limitations of the HCl-leaching process are identified:
- All the equipment used must be corrosion-resistant and pressure-rated which significantly increases the equipment costs.
- Energy-intensive and expensive HCl recovery steps (hydrolysis, chlor-alkali process).

3. Development Challenges

The goal of a successful BR valorization process is to combine sufficient waste volume reduction and economic viability. The main challenge is to ensure the successful extraction of bulk (Fe, Al) and strategic (Sc/REE, Ti) metals in a marketable form at a reasonable operating and capital costs. Therefore, producing the simplest marketable product at minimum number of steps has the most positive impact on the process economy.

The technical challenges and benefits of the recovery of each product are summarized in the Table 3 below:

Table 3. Benefits and challenges with metal extraction from BR.

Component	Synergistic benefits of recovery	Challenges
Fe	• Improves downstream recovery of REE	• High reagent consumption • GHG emissions during carbothermic reduction
Al	• Prevents technical issues with iron smelting	• Harsh conditions (acidic, caustic, roasting) required for extraction
Sc/REE	• High economic value per tonne	• Low grade • Recovery from dilute solutions
Other	• Maximize volume reduction • Prevent contamination of other by-products • Avoiding residues and effluents	• Identifying and obtaining marketable products without extensive purification circuits

Preliminary technoeconomic studies published in the literature [16, 7] show that the main reagent consumption (generally the acid) contributes the most to the operating costs. Three additional goals can be identified:

- Main reagent recycling: to reduce operating costs, water consumption and avoid effluents.
- Minimal effluents: to reduce wastewater treatment costs and avoid displacing environmental impacts.
- No net direct GHG emissions: to stay in line with the GHG reduction targets.

4. Innord's Process

Innord Inc has developed at a bench-scale a process in an attempt to find and answer to some of the challenges previously mentioned. The proprietary process is schematized in the Figure 1 and a high-level description of the steps is described below.

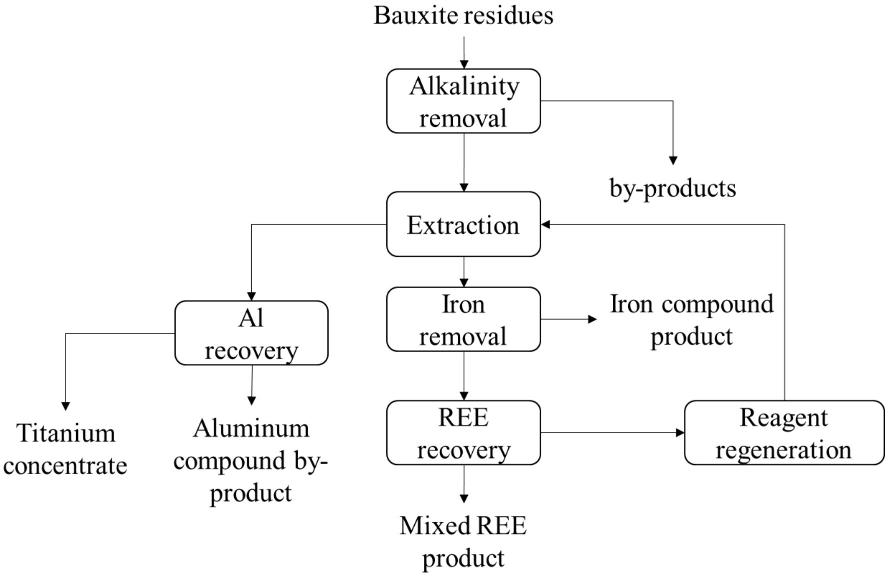

Figure 1. Innord's proposed route for the valorization of bauxite residues.

1. The BR is neutralized to remove the alkalinity; The neutralization reagent is recycled to avoid effluents. By-products are obtained at this stage.
2. Iron and REE are extracted in a proprietary reagent, leaving behind an aluminium-enriched residue that can be further treated to recover an alumina precursor and a titanium concentrate.
3. Iron is separated and recovered as a by-product.
4. REE are recovered from the iron-depleted medium as a mixed concentrate (70% REO equiv.).
5. The extraction reagent is regenerated and is recirculated to the extraction circuit.

The advantages of the process are:

- Multiple products recovery
    - Iron compound product: 80% recovery achieved at bench scale at 99% purity
    - Aluminium compound product: 90% recovery at 95% purity
    - Titanium product: as a concentrate in residue (50-60%)
    - Sc/rare earths: 85% recovery present as a concentrate (70% REO equivalent)
- Minimal water usage (less than 1 m^3/t BR); based on process mass balance
- Minimal effluents

- Potentially up to 90 % volume reduction
- Regeneration and recycling of main reagents

The heat and mass balance of Innord's proposed process was calculated using Aspen Plus® simulation software and bench-scale testwork data. The profitability and the environmental impacts were assessment by calculating the operating costs and the impact on climate change (GHG emissions calculations).

4.1 Operating Costs Estimation

Based on the current state of the technology, the operating costs of the BR process were estimated. The output from the heat and mass balance model and the pricing assumptions were used to calculate the reagents and energy requirements of the process and the operating costs.

The estimated plant operating costs (OPEX) for the valorization of BR through the proposed process is broken down in the Table 4 below. The estimation of the direct OPEX is based on the calculated heat and mass balance and based on a general BR composition.

Table 4. Breakdown of the estimated direct operating costs of Innord's proposed BR valorization process.

Item	**Unit/t dry BR**	**Unit**	**Cost (USD/unit)**	**USD/t of dry BR**
Reagents				48.3[1]
Electricity	685	kWh	0.024	16.4
Net Heat duty	1,198	MJ	0.006	7.2
Labor	0.4752	Person-hour	40	19.0
			total	90.9

[1] Reagents pricing sourced from [18]

4.2 Prospective Life-cycle Impact on Climate Change

Prospective life-cycle impact assessment (LCIA) at the development stage can help to identify potential hotspots and influence the decision making [15]. This preliminary study was performed on the proposed process developed by Innord. The material inputs based on the test results and energy requirements based on the simulation results were used to quantify the impact of the process on climate change using life cycle thinking. The treatment of BR is a multifunctional process achieving both waste removal and the production of metal concentrates. For this reason, the impacts were determined on a basis of one tonne of BR feed treated, as it is a common practice for waste valorization processes.

To calculate the life-cycle impact on climate change, these material and energy requirements were combined with the process inputs impacts on climate change. To account for the multifunctionality of the process, credits for the avoided impact of by-products primary/conventional production and BR landfilling were attributed as "avoided processes", as explained by the authors [15]:

'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.'

The system boundaries considered for the process developed by Innord's is schematized in the Figure 2. The functional unit considered is 1 tonne of BR (dry basis) treated using Innord's proprietary valorization process.

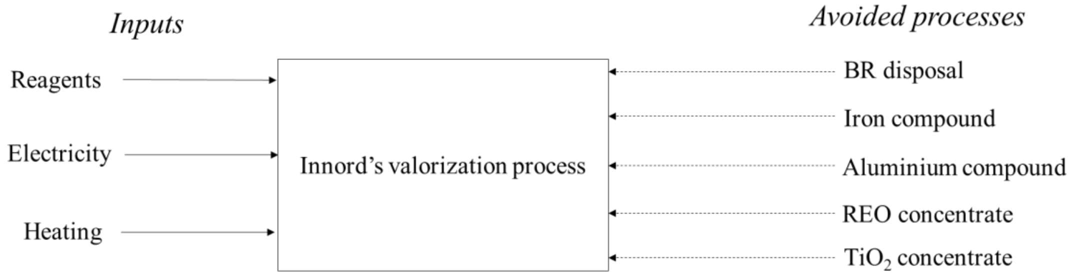

Figure 2. System boundaries for the GHG quantification.

The environmental impact associated with the disposal of BR, the production of the inputs (reagents, energy) and each avoided product was taken from the literature [15] or approximated from available emissions factors [19, 20].

The estimated impact on climate change of the process, are shown in Table 5.

Table 5. GHG assessments of Innord's process.

Inputs	**Unit/t BR**	**unit**	**kg CO_2eq/unit**	**kg CO_2eq/t of dry BR**
Reagents				167.19
Electricity consumption	685	kWh	0.035	28
Heating	1198	MJ	0.054	129.6
Credits				
BR disposal	900	kg	0.0081	-7.29
Iron compound				-5
Aluminium compound				-207
Titanium concentrate				-307.8
Other products				-118.8
REO concentrate				-9.59
Total				-399.6

According to this GHG impact assessment, the project will not create GHG emissions as the impacts are distributed along the multiple by-products obtained.

5. Conclusion

The valorization of bauxite residues has been a studied topic for many decades and there is yet no commercial-scale process that can extract the metal value of BR profitably and without shifting environmental impacts. An overview of the previous and current proposed approaches to extract the metal value of bauxite residues allowed to identify the processing challenges to address for a successful BR valorization technology. These challenges comprise:

- Effective waste volume reduction
- Minimal effluents and emissions
- Low operating costs
- Marketable products in a minimum number of steps

Innord has proposed a process in an attempt to address most of these challenges by obtaining multiple product streams:

- Iron and aluminium products
- Strategic metal concentrates (Sc/REE, Ti)
- other by-products

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