

Review

Overview on Hydrometallurgical Recovery of Rare-Earth Metals from Red Mud

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Abstract: Aluminum is produced from its primary bauxite ore through the Bayer process. Although Al is important nowadays in the development of humanity, its production leads to the generation of a huge amount of waste, called red mud. Globally, the estimation of the stock of red mud is about 4 billion tons, with about 10 million tons located in Turkey. The presence of rare-earth elements (REEs) in crucial materials such as red mud makes it a major source of these elements. A number of methods have been developed for treating red mud, which are employed globally to recover valuable products. The application of a suitable method for REE extraction from red mud is a way to overcome the supply risk, contributing to reducing the environmental issues linked to red mud pollution. The current review summarizes the research on red mud processing and examines the viability of recovering REEs from red mud sustainably, utilizing hydrometallurgy and biohydrometallurgy.

Keywords: aluminum; red mud; hydrometallurgy; leaching; solvent extraction; rare-earth elements



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1. Introduction

Aluminum (Al) is one of the most important non-ferrous metals in industrial progress. This metal is important because of its application in several sectors [1]. Aluminum is the second most malleable metal and has good thermal and electrical conductivity [2]. This metal has also a low density (2.7 kg/dm³), with a melting point of 658 °C, and has excellent corrosion resistance [3]. Due to these extraordinary features, Al is utilized in building, transportation, the electrical sector, consumer electronics, consumer products, and the defense industry [4]. As one of the most abundant elements in the Earth's crust (7.96%) [5], aluminum is produced from bauxite, which is its common primary ore [6]. The demand for primary aluminum is around 107.8 million tons, and it will increase by another 50% by 2050 [7].

Although several methods exist for aluminum recovery from bauxite, the Bayer process has been the best known and most widely used industrially. Karl Josef Bayer developed this technology in 1887 in France. The procedure essentially entails extracting Al₂O₃ from bauxite ore in an alkaline sodium hydroxide solution (NaOH) at a reasonably high temperature [8]. Figure 1 shows a simplified diagram of Al extraction from bauxite ore.

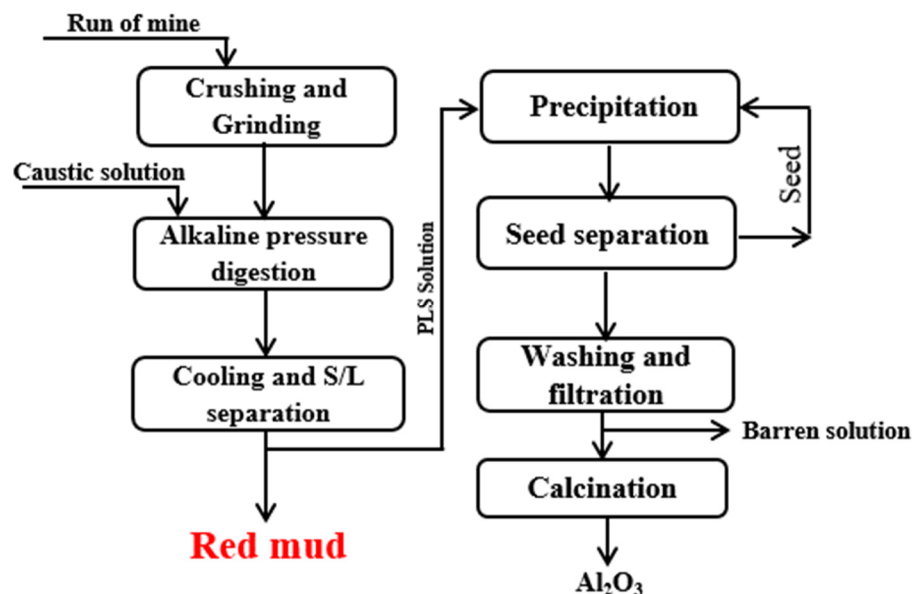


Figure 1. Simplified Bayer process flow diagram [9].

Turkey's bauxite resources are mostly found in the districts of Silifke-Taşucu, Seydişehir-Akseki, Kokaksu-Zonguldak, Słahiye-Payas, and Milas-Mula. The majority of economically valuable bauxite deposits are found in the Seydişehir-Akseki area, in the western Taurus Mountains (the Mediterranean Region of Turkey). Bauxite ores from these regions are processed at the ETI Aluminum Facility in Seydişehir, Konya, the only consolidated aluminum facility capable of producing both alumina and aluminum from bauxite ores. About 550,000 tons of bauxite ore are processed at the plant to develop 80,000 tons of primary aluminum annually, 400,000 tons of aluminum hydroxide, and 250,000 tons of calcined alumina [10]. Figure 2a shows the plant.



Figure 2. ETI aluminum plant (a) and red mud tailings dam (b) [11].

As seen in Figure 1, the Bayer process generates a second product (residue) known as red mud, which is often stored in a tailings dam (Figure 2b). For every ton of alumina produced, around 0.8–1.5 tons of red mud (RM) are produced. As of 2024, bauxite residue production increased by 135 million tons per year. There are around 4 billion tons of red mud in the world [12]. In China, over 60 million tons of bauxite residue are released each year. Rusal and Mytilineos are enormous alumina recovery organizations that produce red mud and recover precious components. In Turkey, according to the data from the ETI aluminum plant, from 2.12 tons of run-of-mine bauxite, 1 ton of alumina is produced, and 0.996 tons of red mud are generated [13,14]. In total, about 100 million tons of red mud have been generated from 1974 up to today across the world [12,15].

Red mud management remains a concern for the worldwide alumina industries, as well as regulatory agencies, due to the vast amounts created, which include minute and trace concentrations of heavy metals and radionuclides, as well as the adverse effects of its disposal. Red mud disposal is still difficult because of its high alkalinity (pH 10–13, according to Liu et al. [16]). The most significant disadvantage of the pond disposal procedure is the danger that it poses to people and wildlife when they come into contact with caustic liquor and residue (when they are not treated appropriately) [17]. Only seven of the 84 alumina refineries in the world still employ planned marine disposal due to a lack of available land. However, red mud can be used as an alternative, economic resource to several common ores, since it contains a lot of precious elements [18].

2. The Utilization of Red Mud in Various Applications

Red mud can be utilized as a raw material in a variety of applications. Red mud is stored in either dry landfills or red mud ponds. Red mud disposal becomes stable and safer because of drying. Significant efforts are being undertaken globally to identify appropriate uses for red mud so that the alumina industry produces no residue at all [19,20].

Red mud ponds, or bauxite residue disposal zones, are designed to contain debris and to be used for industrial or civil purposes when storage capacity is depleted. Red mud can be used as a landfill cover material by adding 8% sodium bentonite, contributing significantly to the construction sector [21,22]. A soil and red mud/lime sludge mixture is a practical and creative option for creating a barrier layer for open dump cleanup, preserving soil and maximizing the use of industrial waste. This approach ensures the preservation of soil and optimal utilization of industrial waste [23]. Red mud disposal can lead to alkaline mine drainage and groundwater metal pollution. However, it can also provide economic and human resource benefits by enabling the construction of facilities for bauxite beneficiation. To save transportation costs, waste facilities should be located near aluminum plants [24]. Red mud can also be used in carbothermal reduction for iron extraction, a stage in steel production, and as a reductant for recovering aluminum. This could lead to a new method for pig iron production [25].

Moreover, the elements, namely lead and chromium, may be effectively removed using red mud as an adsorbent [26]. Its potential adsorption capabilities, when activated in air, could be useful for the treatment of wastewater in a variety of sectors. The thermodynamic parameters demonstrate the viability of using the red mud adsorbent method for removing metal ions by particle diffusion [27]. The outcomes show that a wide range of Pb^{2+} and Cr^{6+} concentrations may be effectively removed using red mud. About 1% HNO_3 can be used to quantitatively elute metal ions that have been adsorbed on this material's column. It has also been observed that the presence of additional salts in effluents does not have an unfavorable impact upon using red mud as an adsorbent [23,28]. Environmental scientists are exploring the use of affordable substitutes for activated carbon in wastewater treatment [29]. Red mud has been modified to be a low-cost adsorbent that can effectively remove phenolic compounds from wastewater [28,30,31]. The adsorbent can separate 2,4-dichlorophenol and 4-chlorophenol up to 94%–97%, while removing 2-chlorophenol and phenol up to 50%–81%. It can also efficiently remove Procion orange from wastewater [32–35]. Red mud can also be used to expel fluoride from aqueous solutions from various sources, demonstrating its potential as a novel water treatment technique [24,36].

Glass-ceramic products were produced using precise combinations of fly ash, red mud, and waste pot liner, reported by Balsubramaniam et al. [37]. These products exhibit exceptional qualities and beautiful aesthetics for potential usage as ornamental tiles in the building sector. In these circumstances, sintering is one of the efficient carbo-thermal reduction procedures used within the furnace that helps produce sintered slag. Due to chemical and physical reduction processes, the amount of fly ash in the mixture of fly ash and red mud reduces with increasing sintering temperature [38]. The evolution of minerals in the sintered product and the completed product has been thoroughly studied to confirm the presence of minerals and ions following the technique [39].

Red mud is a versatile material used in various applications, including making aluminum titanate–mullite composites and wear-resistant coatings for machinery parts [40–42]. Its low waste management capacity makes it an effective replacement for commercial catalysts [43]. Red mud’s characteristics, including its ferric oxide content, large surface area, sintering resistance, toxin tolerance, and low cost, make it an attractive catalyst for various processes [44,45]. When sintered with limestone, it forms a sintering mud used to recover aluminum and to add alkali back into the main process [46]. Alternatively, advanced technology could be used to extract and recover target metals from fresh, high-quality bauxite deposits worldwide.

3. Potential of Metal Contents Present in Red Mud and Their Importance

Red mud’s mineralogical composition comprises aluminum oxide as diasporite and boehmite, ferrous minerals as hematite, limonite and goethite, anatase, rutile, calcite, dolomite, and pyrite. In addition, the extra phases produced during the Bayer process, namely sodalite and gibbsite, contribute to the red mud’s composition [47].

In general, the chemical composition of red mud is Fe_2O_3 , 30%–60%; Al_2O_3 , 18%–25%; SiO_2 , 15%–20%; Na_2O , 8%–12%; CaO , 2%–8%; and TiO_2 , 2%–5%. The residue is red in color due to the presence of oxidized iron (hematite mineral) in an average proportion of 33–48%. Red mud contains 14%–45% Fe, 5%–14% Al, 1%–9% Si, 1%–6% Na, and 2%–12% Ti [48,49].

Alongside these elements, red mud also contains critical metals that are highly researched for the development of new technologies. Red mud comprises significant amounts of REEs, including lanthanum (La), scandium (Sc), cerium (Ce), neodymium (Nd), samarium (Sm), yttrium (Y), gallium (Ga), thorium (Th), uranium (U), zirconium (Zr), vanadium (V), and others. REEs are present in red mud as the minerals synchysite ($\text{CaCe}(\text{CO}_3)_2$ F), xenotime (YPO_4), and monazite (Ce, La, Pr, Nd, Th, Y) PO_4 [23,50]. Typically, red mud consists of 121, 76, 114, 368, 28, 99, 21, 5, 22, 4, 17, 14, 4, 2, 14, and 3 g/ton of REEs, such as Sc, Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Er, Ho, Tm, Yb, and Lu, respectively. It has been shown that red mud contains 500 to 1700 ppm of REEs. It is worth noting that the Sc percentage in red mud is considerable, ranging from 130 to 390 ppm [47]. The REE concentrations in red mud are more significant when compared to the currently mined bastnäsite (carbonate-fluorides) and monazite (phosphates) ores, where the average REE levels range from 3000 to 13,000 g/t [51].

Importance of Rare-Earth Metals and Their Scarcity

This new century is the most technologically advanced period, with a significant use of rare-earth elements (REEs). The utilization of natural resources is increasing day by day as the population grows and their requirements expand. The chemical, catalytic, electrical, magnetic, and optical characteristics of REEs make them extremely valuable [52]. The demand for rare-earth metals on a worldwide scale has been rising quickly in recent years. The expected annual growth rate, which is estimated to be 8% [53], might range from 3.7% to 8.6%, depending on the scenario. About 133,000 metric tons of REEs were used globally in 2010, and it was predicted that the amount would increase to 350,000 metric tons by the end of 2023 [54]. REEs have a wide range of uses in different sectors, such as industrial, medicinal, petroleum, glass, alloys, biological, automotive, permanent magnets, automobile catalytic converters, and other applications, as well as metallurgical additives, glass polishing, alloys, ceramics, and other products [47,55,56]. China has produced the great majority of the world’s rare-earth metals and its market share was around 90%. China, however, has started a campaign to impose restrictions and has established a system of export quotas for rare-earth products. Due to the tremendous environmental harm and ecological devastation caused by local mining and smelting operations for rare-earth metals, several restrictions have been implemented [57]. Since 2005, China has gradually lowered the number of rare-earth metals it may export to other countries. In the near future, the demand for rare-earth metals is anticipated to exceed the supply, particularly for a few heavy rare-earth metals [58].

As a result, various strategies, such as reopening old REE mines or recycling REEs from secondary sources, such as red mud, fluorescent lamps, Nd hybrid magnets, scrap alloys, e-waste, fly ash and other waste sources could be interesting and efficient ways to overcome this situation, while also reducing the environmental issues associated with mining [59]. The best course of action when it comes to recycling is always to create as little waste as possible, followed by minimizing it further. The second strategy may involve reusing diminished resources through energy recovery and recycling processes. Disposal at a suitable area, with adequate treatment, is the least favored option.

In this aspect, the world is looking for alternative REE sources. Bauxite residues are a potential source of REEs if appropriately exploited [60,61]. The lack of/rapid depletion of primary resources, industrial relevance, production chain bottlenecks, the significance of green energy, and the shortage of REEs discussed above has made it vital to develop a sustainable green process for REE recovery from red mud. The REE economy is highly concentrated, being susceptible from a geopolitical standpoint due to the absence of diversity in the REE mineral deposits. The wide application of REEs in various sectors is reflected in Table 1.

Table 1. REE requirements by application and total consumption rates (adapted from [53,62–64]). Note: LREE: light REEs and HREE: heavy REEs.

REE Application	Total REE Consumption (%)	LREE							HREE			Others%
		La%	Ce%	Pr%	Nd%	Sm%	Eu%	Gd%	Tb%	Dy%	Y%	
Magnets	23	–	–	23.4	69.4	–	–	2	0.2	5	–	–
Battery alloys	8	50	33.4	3.3	10	3.3	–	–	–	–	–	–
Metal alloys	8	26	52	5.5	16.5	–	–	–	–	–	–	–
Auto catalysts	5	5	90	2	3	–	–	–	–	–	–	–
Petroleum refining	18	90	10	–	–	–	–	–	–	–	–	–
Polishing compounds	12	31.5	65	3.5	–	–	–	–	–	–	–	–
Glass additives	7	24	66	1	3	–	–	–	–	–	2	4
Phosphors	2	8.5	11	–	–	–	4.9	1.8	4.6	–	69.2	–
Ceramics	6	17	12	6	12	–	–	–	–	–	53	–
Other	10	19	39	4	15	2	–	1	–	–	19	–

In 2007, there were 2.7 billion tons of red mud in the world’s inventory; by 2014, there were 3.5 billion tons [17,65]. By 2024, the total red mud inventory might be 4 billion tons, assuming a production rate of 135 million tons per year. It was examined that the total metal values (mainly REEs), contained in around 4 billion tons of red mud were accounted for based on the metal prices for the second quarter of 2019. Up to 4.3 trillion dollars’ worth of economic potential can be unlocked with the optimal recovery of REEs from red mud. The list of CRMs has recently been amended by the European Commission (EC), moving from 30 in 2020 to 34 in 2023 [66,67]. Bauxite is now included in the updated list of CRMs, which is given in Table 2, along with Li, Ti, and Sr [67]. Therefore, from the standpoint of the circular economy, which potentially opens up billion-dollar prospects from hoarded red mud globally, the quantitative valuation of red mud on an industrial scale might be substantial.

Table 2. Globally production of REEs from 2017–2023 (in tons) [68–70].

Country	2017	2018	2019	2020	2021	2022	2023
United stateS	-	18,000	26,000	39,000	43,000	38,000	43,000
Australia	19,000	21,000	21,000	21,000	22,000	17,000	18,000
Brazil	1700	1100	1000	600	500	1000	80
Burma	NA	19,000	22,000	31,000	26,000	30,000	38,000
Burundi	-	630	600	300	100	500	-
China	105,000	120,000	132,000	140,000	168,000	140,000	240,000

Table 2. Cont.

Country	2017	2018	2019	2020	2021	2022	2023
Madagascar	-	2000	2000	2800	3200	8000	960
India	1800	2900	3000	2900	2900	3000	2900
Russia	2600	2600	2700	2700	2700	2700	2600
Thailand	1300	1000	1800	3600	8000	2000	7100
Vietnam	200	920	900	700	400	1000	600
World total	132,000	190,000	210,000	240,000	280,000	243,300	350,000

NA: Non Available.

4. REE Extraction from Red Mud and Sustainability

Rare-earth elements, as their name suggests, are not that rare in the earth crust, since they are more abundant than some base metals [71]. However, the group of these elements has been classified as critical materials due to their economic importance and supply risk. Figure 3 illustrates the critical materials in the EU. The supply risk is because it is produced in fewer countries (China is the main producer) [66].

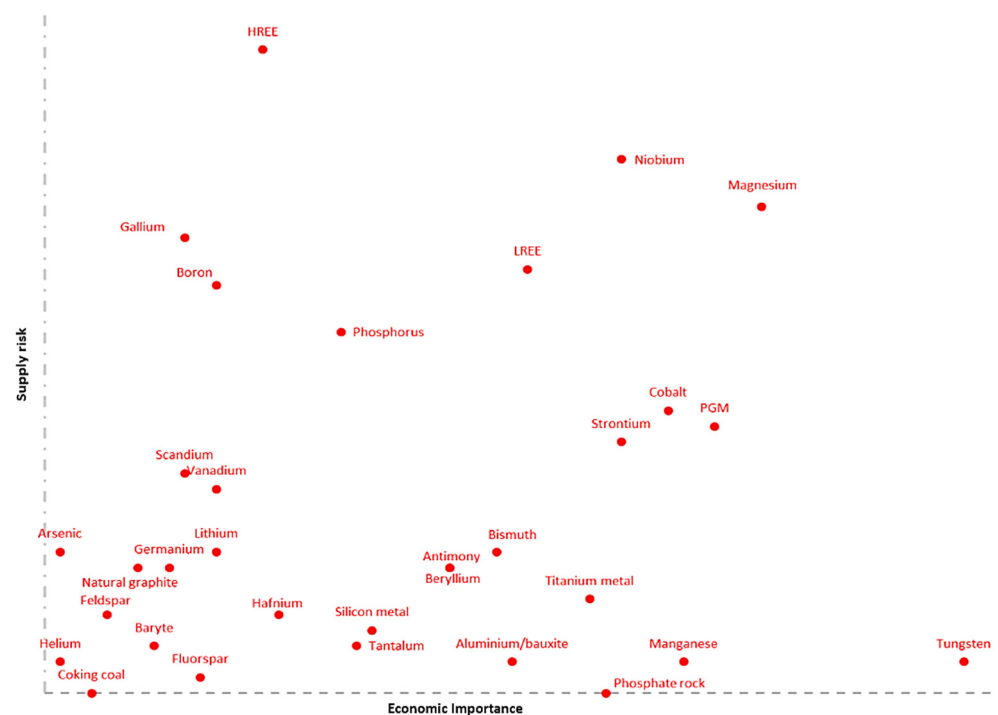


Figure 3. Supply risk results and economic importance of 2023 critical materials [67].

Based on the type of metal in demand, the price of the major REEs fluctuated between 2014 and 2023 on a worldwide scale. Neodymium oxide increased from 71,180 US dollars per metric ton in 2014 to 50,000 US dollars per metric ton in 2023, and Yttrium oxide was estimated to cost 21 US dollars per kilogram in 2014 and 31 US dollars per kilogram in 2023. Lanthanum oxide was predicted to cost \$5955 USD per metric ton in 2014, rising to \$3700 USD per metric ton in 2023. Conversely, the expected price for cerium oxide in 2014 was 8847 US dollars per metric ton, but it subsequently decreased to 1013 US dollars per metric ton in 2023 (<https://www.scrapmonster.com/metal-prices/rare-earth/cerium/921>, accessed on 12 April 2023).

The conventional metal extraction from primary ore has followed a linear economy approach (take, make, waste) (Figure 4a). This approach has been considered an uncompleted process since, at the end, an amount of waste material is left at the tailings dump. However, the recycling of waste at the end of the process in a closed loop, known as circular economy, is a sustainable approach [72]. The procedures suggested for extracting

metal values from red mud are relatively restricted, according to a thorough literature review. Several authors have reported on or talked about the commercial side of bulk-scale management [47,55,56]. In general, for a number of reasons, the industrial-scale recovery of metal values and efficient red mud management outside of dry stockpiles have been neglected for many years. Even if it were possible to extract red mud and meet the demand for these metals, the value of red mud, in particular through metal value recovery, would remain a significant circular economy problem. The limitations that prevent red mud from moving into a circular economy are a lack of appropriate technology and inadequate waste management procedures. Therefore, the application of the circular economy to red mud contributes to reducing environmental impact, while at the same time increasing its value through REE extraction [73]. The worldwide demand for metal value recovery from RM in general, and REE valorization in particular, has been mounting, owing to its constituents, inventories, and continuous generation as secondary resources. The recovery of REEs from RM has been receiving significant attention from the global scientific and industrialist communities in order to overcome challenges such as ensuring a steady supply of REEs for industrial economies, addressing geo-political instability, and considering the perspective of strategic importance [71,74].

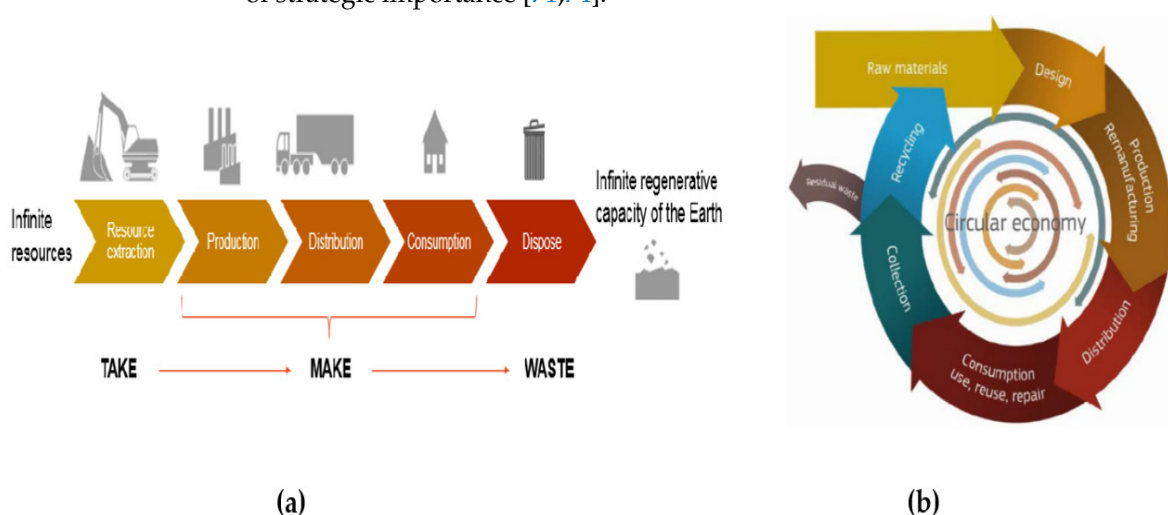


Figure 4. (a) Linear economy [72] and (b) circular economy in mining activities [75].

Several processes/methods have been undertaken to meet the circular economy approach. The main application processes in the metallurgical extraction of REEs from red mud are pyrometallurgy [49,76], hydrometallurgy [71,77,78], and biohydrometallurgy [79]. However, the last two processes are more economically and environmentally acceptable than pyrometallurgy [76]. Pyrometallurgical procedures frequently have significant capital costs, require a lot of energy, and go against the objective of reducing the carbon footprint for value recovery from red mud. However, hydrometallurgy provides a number of benefits, including cheaper capital costs, flexibility in handling difficult secondary resources, diverse by-product recovery, relatively low energy consumption, and the ability to process closely related wastes and even extremely low-grade materials. For the separation, purification, and concentration of the metals, a variety of hydrometallurgical processes, including precipitation, cementation, solvent extraction, adsorption, and ion exchange are available [59,80]. These methods have several drawbacks compared to heterogeneous reactions (such as longer operating times), and in most instances, the goal is to remove metals rather than recover them [54,71]. In comparison to the pyrometallurgical method, hydrometallurgical processes are frequently thought to be more effective, versatile, and flexible when it comes to recovering valuable metals from waste, separating, purifying, and concentrating metals from its mixed leachate/solutions [74,81]. The biohydrometallurgical technique has been gaining attention in recent years for the recovery of rare-earth elements (REEs), and new strategies are being explored for a successful application of biotechnology

to industrial sectors for REE recovery. The literature shows that the recovery of rare-earth metals from red mud, either through hydrometallurgical or biohydrometallurgical techniques, has been discussed. However, a comprehensive review combining both methods has not been explored so far in the literature. In this regard, the present review article focused on the critical examination of biohydrometallurgy and hydrometallurgy aspects of the recovery of rare-earth from red mud.

4.1. Hydrometallurgical Recovery of REEs from Red Mud

The goal of hydrometallurgical operations is to selectively separate or leach rare-earth metals from red mud or bauxite residue [82]. The kind of bauxite residue shows which acid is best adapted for leaching, and research shows that the results of acid leaching are highly dependent on the type of bauxite residue. As a result, a rare-earth element recovery technique from bauxite residue must be customized for a particular type of bauxite residue. The hydrometallurgical (chemical leaching) recovery of REEs from red mud samples has been the subject of several laboratory-scale experiments [47,48,51,83,84]. It has been shown that rare-earth elements may be easily extracted from bauxite residue using diluted mineral acids, such as nitric, sulfuric, hydrochloric, acetic, and citric acids [55]. Additionally, other techniques, including carbonization, leaching in concentrated sulfuric acid, and using extremely acidic ionic liquids, have produced notable outcomes for the selective leaching of REEs [85–87]. The schematic representation of the hydrometallurgical recovery of REEs from red mud is depicted in Figure 5.

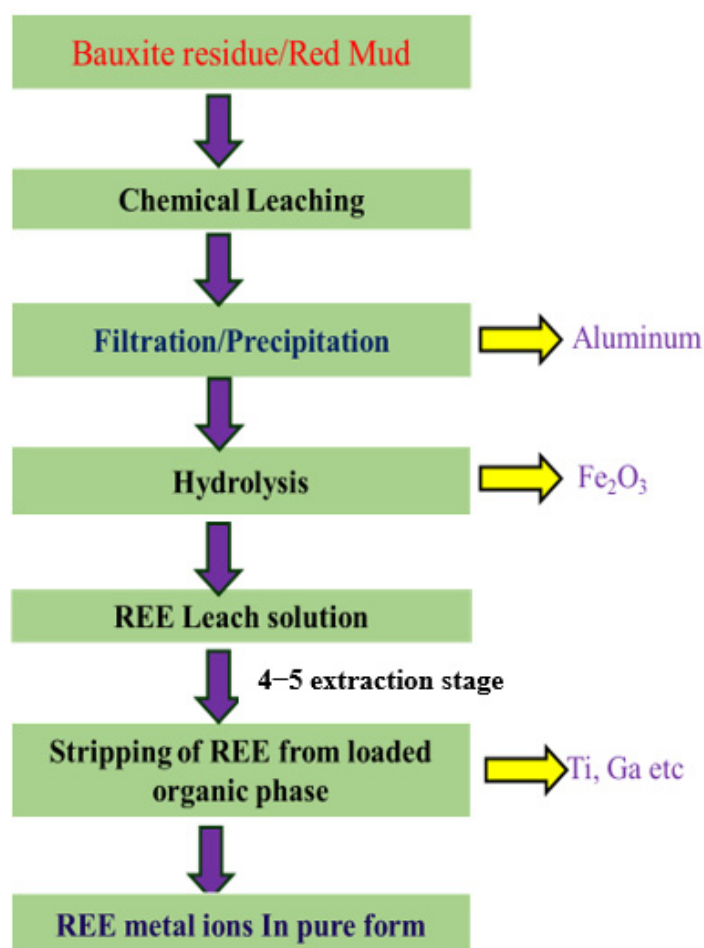


Figure 5. Flow sheet for the recovery of rare-earth metals from red mud using the chemical hydrometallurgical approach.

4.1.1.1. Extraction of Scandium from Red Mud

The most extensively studied REE, scandium, can occasionally be nearly two times as abundant in red mud than it was in the initial ore [88]. It accounts for more than 95% of the REEs' commercial worth in bauxite residue. Ores having scandium contents between 0.002 and 0.005% (20 to 50 mg/kg) can be regarded as significant sources of the metal and should be fully used [47]. Due to the extreme difficulty in isolating it from the other REEs, scandium is the most costly REE in terms of chemical yield. This results from the chemical reaction that occurs between Sc and the organic materials utilized in liquid–liquid extraction. It is quite difficult to break this chemical connection because of its tremendous stability [89]. However, because of the high amounts of primary components, particularly Fe, Al, Ti, it is challenging to directly extract scandium from red mud. Derevyankin et al. [90] reported the technique for extracting scandium from red mud, and the authors acquired the separation of scandium from yttrium in RM. Generally, the leaching technique using diluted HNO_3 has been improved in order to recover scandium from bauxite residue and was carried out at a pilot scale by Ochsenkühn-Petropoulou et al. [91], revealing that a 78%–80% recovery rate of Sc is possible. Ochsenkühn-Petropoulou et al. [91] also studied the optimum leaching conditions, by comparing leaching with various acids (HCl , HNO_3 , or H_2SO_4). They found 0.5 M HNO_3 to be the most effective, and the procedure may be carried out at room temperature and pressure. This is due to the high oxidation potential of nitric acid compared to other acids [92]. The issue with leaching with HNO_3 is the challenge of recovering nitrate ions adsorbed to bauxite residue [93]. In contrast, the leaching process proved less effective for recovering the light lanthanides, i.e., achieving 30%–50% recovery for scandium and 96% for yttrium. Although leaching with HCl is quite effective for recovering rare earths, one major drawback is that significant quantities of iron are co-dissolved [55]. These impurities can be separated using suitable experimental circumstances, such as pH or potential, and the details are presented below. Studies have shown that the exploitation of the dry digestion method, subsequently followed by water leaching, shows good extraction efficiencies for Sc and Ti from bauxite residue using H_2SO_4 [94]. Borra et al. [55] reported the removal of iron from red mud using graphite (5 wt%) as a reducing agent and 20 wt% wollastonite as a flux. It was discovered that the bauxite residue might be used to recover more than 95% of the iron. High temperatures enhanced the extraction yields compared to room temperature leaching of the slag sample. At 90 °C, HCl and HNO_3 may be used to leach all of the scandium, the majority of other REEs, and almost 70% of the titanium. With H_2SO_4 leaching, scandium is more selective than other REEs [95]. Recently Ding et al. [96] explored a combined sulfation–roasting–water leaching process for the selective removal of Sc from red mud. The investigation showed that 91.98% of the Sc leaching efficiency was achieved under optimal conditions [96]. It is important to understand that when bauxite residue is leached with acids, some of the acid is used to neutralize the extremely alkaline bauxite residue. Habilbi et al. [97] investigated methods to dissolve titanium and rare-earth elements (REEs), concentrating on sulfuric acid, and then hydrolyzing the titanium to precipitate the metal. A solvent extraction procedure was used to concentrate the REEs in the solution. Ti and Sc were found to have dissolving efficiencies of 91% and 93%, respectively. The final TiO_2 product had a purity of 92%, with the mixed iron and titanium oxides serving as the primary contaminants. The Ti precipitation efficiency was around 86%. Following Ti recovery, solvent extraction was used to study the Sc concentration. Sc was concentrated from 69 ppm in the red mud to 1125 ppm in the precipitate that was produced [97]. Following leaching, the rare earths can be extracted from the leachate via solvent extraction or by selectively precipitating them as oxalate. After leaching bauxite residue with sulfuric acid, several ion-exchange resins have been explored to selectively remove scandium from the leachate [98,99].

By employing solvent extraction with an acidic extractant, such as D2EHPA (Di-(2-ethylhexyl) phosphoric acid), Wang et al. [100] were able to recover scandium from a synthetic red mud leaching solution through a cation exchange mechanism at a lower acid medium. Further, a solution of D2EHPA and tributylphosphate (TBP) at pH 0.4 allowed for

the extraction of scandium from the leachate. The leach solution was then extracted once more using a solvent that contained two chelating agents, an organophosphorus acid, such as Primene JMT (through ion pair principle), a carboxylic acid (Versatic 10), an ethylhexyl phosphonic acid mono-2-ethylhexyl ester (ion exchange mechanism), and an organophosphorus acid, namely Cya LIX 984N–LIX 860, LIX54-100- β -diketones and Cya ketoxime LIX 84N. Using the stated procedure with 0.05 mol/L D2EHPA and 0.05 mol/L TBP in 100% aliphatic diluent (Shellsol D70) as the extractant, the recovery of scandium was 99% at different experimental conditions such as pH, aqueous to organic ratio, etc. [101]. Other recent developments involving hydrometallurgical routes include direct leaching with ionic liquids, which has been explored for the separation of valuable metals from RM and has provided assuring results concerning Sc extraction [102,103]. Complex processing refers to the employment of pyrometallurgical and hydrometallurgical processes in combination to extract metal values from bauxite residue [104].

Hubicki [105] has also had success using selective ion exchangers to separate Sc^{3+} from other REEs. Employing 6 M HCl, the amount of Sc^{3+} may be increased to $500 \text{ g}\cdot\text{L}^{-1}$. In addition to the traditional adsorbent, it is important to highlight that functional porous hybrid materials show improved selectivity for the HREEs [106,107]. Ochsenkühn-Petropulu et al. [92] discovered a combination of solvent extraction and ion exchange methods. Red mud components were thoroughly mixed with $\text{NaKCO}_3/\text{Na}_2\text{B}_4\text{O}_7$ (1:1) under different experimental parameters. The remaining half of the leachate was then put through a column study with Dowex 50W-X8 resins. After at least 5 min, 2 M NaOH quantitatively stripped away 93.5% of the scandium as $\text{Sc}(\text{OH})_6^{3-}$ in the aqueous phase.

4.1.2. Extraction of Scandium in Presence of Iron from Red Mud

The link between the breakdown of iron and the extraction of scandium demonstrates the intimate relationship between scandium and iron minerals [55]. Without introducing iron into the solution, it is possible to recover around 50% of Sc from the bauxite residue samples. However, attempting to recover more than 50% of the scandium invariably causes a significant portion of the iron to dissolve [108]. Without dissolving the iron, it is impossible to extract all Sc found in the bauxite residue. By digesting red mud with a weak acid solution made by saturating water with SO_2 , the REEs can be selectively dissolved while leaving the majority of the iron undigested [47,109]. After it is removed, a rare earths solution is produced, from which the REEs may be extracted using a solvent. In one variation of the procedure, the bauxite residue is leached in three phases at 50°C for two hours, with solutions of Na_2CO_3 and/or NaHCO_3 (5%–12%) at an S/L ratio of 1:2.50 to 1:5. The scandium is recovered from the effluent by adding NaAlO_2 or Na_2ZnO_2 , and then heating the mixture for two hours at 80°C [110]. The resultant precipitate is then removed from the solution, followed by filtering and washing with 15% NaOH solution, while being heated simultaneously till boiling. The material is then dissolved in a 1 to 5% aqueous HCl solution, and the leftover solid is filtered out. By adding 10%–25% of an aqueous NH_3 solution to the filtrate, Sc is precipitated as $\text{Sc}(\text{OH})_3$ [111]. In an updated version of the procedure, the mixed $\text{Na}_2\text{CO}_3/\text{NaHCO}_3$ solutions are passed through a gas mixture including air and 10%–17% CO_2 to carry out the leaching process [112]. Later, titania is first precipitated from the leachate using an organic flocculant, then the impurities are removed using electrolysis, and lastly, scandium is precipitated using sodium zincate in conjunction with an organic flocculant. The process is further enhanced by passing a flow of flue gases from the CO_2 -containing bauxite sintering or calcination furnaces through a series of leaching processes using sodium carbonate and sodium hydrogen carbonate mixes. The drawback is that the other REEs have a far lesser propensity to form soluble carbonate. Recently, Zhang et al. [113] studied concentrated hydrochloric acid dissolution proceeded by coordination-solvent extraction of iron by Aliquat 336 and the utilization of P204 as an extractant to concentrate scandium in a novel method for recovering iron and REEs from red mud. They discovered that Fe, Al, Ti, Sc, La, Ce, Nd, and Y had leaching efficiencies of up to 95.9%, 82.1%, 68.3%, and 93.3%, respectively. The authors extracted Sc in one step

using P204 at a concentration of 5%, with an extraction efficiency of around 100%. Similarly, Zhou et al. [114] studied the preferential leaching of Sc and Fe from red mud and computed 79.6% and 6.12% of the leaching effectiveness of Sc and Fe, respectively. They performed this by using EDTA as a complexing agent to re-distribute the forms of Sc and Fe ions in the removal process. Salman et al. [115] investigated the use of hydrometallurgical and pyrometallurgical processes to remove Fe and Sc from RM. He discovered that utilizing D2EHPA was selective in recovering 95% of Sc and 65% of Ti following Fe at pH 1. This was achieved with 7 mol/L HCl at an S/L ratio of 1/25 for 2 h at 75 °C under reflux circumstances, resulting in 75% of Sc being leached. With various experimental conditions, the Sc extraction efficiency can reach 97%. TBP can increase the extraction efficiency of Sc to 99%, and diethyl ether can increase the extraction efficiency of Fe to 99%. Furthermore, the authors used 1 mol/L HCL to achieve 92.85% of Sc and 100% of Fe [115].

4.1.3. Extraction of Other Rare-Earth Metals from Red Mud

Red mud also contains scandium, as well as very small, but important, levels of other rare earths and radioactive elements, such as yttrium ($60\text{--}150\text{ g}\cdot\text{t}^{-1}$), uranium ($50\text{--}60\text{ g}\cdot\text{t}^{-1}$), thorium ($20\text{--}30\text{ g}\cdot\text{t}^{-1}$), and other elements [71,116]. An alternate way of extracting scandium and uranium from red mud is the sulfuric acid sorption procedure [116]. The ions of uranium (U) and scandium were adsorbed using the amphoteric resins of AFI-21 and AFI-22, which contain nitrogen and phosphorus. As a result, in addition to the 50% Sc recovery, some radioactive U and thorium(Th) were also extracted from the red mud's H_2SO_4 pulp. In order to selectively recover rare-earth metals with atomic numbers of 57 to 71, as well as Sc and Y, from Bayer red mud without simultaneously dissolving Fe and Ti to the leachate, Fulford et al. [109] designed a leaching technique that involves the introduction of sulfur dioxide. He suggested that a diluted sulfuric acid solution was used first to digest the red mud slurry. A final digesting solution, including a few contaminants and rare-earth elements was developed by adjusting the pH values between 1.8 and 3.2. REEs and more sodalite-type compounds were leached out when the pH dropped to around 1.8–2.5. Leachate containing some rare earths and, particularly, few contaminants completed three leaching stages by dividing the second leach into pH levels of 1.5–2.4. The alumina, soda and silica impurities were dissolved together with the rare-earth metals in the one step process (slurry pH 2.0). Using various ammonium hydrosulfate concentrations, Doronin et al. [117] performed the leaching of red mud. They noticed that the main elements and the rare-earth elements were dissolved by leaching with medium concentrations of ammonium hydrosulfate (8.45%) and high concentrations of ammonium hydrosulfate (58.7%). A 58.7% reagent concentration was obtained after 60 min of dissolving at an S/L ratio of 14, and the rare-earth metal extraction was 70%–95% at boiling point. However, it also caused calcium (78.61%) and iron (60.25%) to dissolve in the solution along with the REEs.

Abhilash et al. [84] studied the effects of H_2SO_4 leaching on a bauxite residue sample that included 110 ppm of cerium and 70 ppm of lanthanum and revealed that a 3 M H_2SO_4 solution produced the highest levels of recovery. Efficient recovery (99%) of REEs such as Ce and La from red mud using H_2SO_4 solutions has been reported [118]. Abdulvaliyev et al. [119] explored on use of the Bayer–Hydrogarnet treatment for the processing of red mud to recover the contaminated metal values, such as Na_2O , Al_2O_3 , Ga, and V_2O_5 by using autoclave leaching with lime [119]. The results showed the recovery of vanadium as ammoniummetavanadate by using precipitation with ammonium sulphate and sulfuric acid solutions. The same authors continued their investigations on the extraction of vanadium and gallium from solid waste by-products of vanadium sludge and calcination plant containing electrofilter dust of the Bayer process [120]. In addition, Chenna Rao and colleagues conducted a series of tests on the leaching of Greece red mud/bauxite residue for the recovery of rare-earth elements using different acids, such as HCl, H_2SO_4 , HNO_3 , acetic acid, methanesulfonic acid, and citric acid. Under the same experimental settings, the extraction or dissolving capacities of these studied systems toward the REEs were

effectively compared. With HCl acid, the maximum REE extraction efficiency was attained at 6 M, 25 °C, and 24 h of reaction time [55].

The rare-earth elements were then extracted via solvent extraction from the resultant leachate using different pH levels and organic phases. The solvent extraction included D2EHPA, 2-ethylhexyl phosphonic acid mono-2-ethylhexyl ester (EHEHPA), phosphonic acid esters (Cyanex 272), thiophosphinic acid esters (Cyanex 301), and mixtures of the aforementioned organic phases with tributyl phosphate (TBP), trioctylphosphate [121]. With the exception of yttrium and HREEs, TOPO typically reduces the extraction level of light and intermediate REEs [54,109]. However, lanthanides, which contain roughly 96% yttrium and 80% scandium, as well as approximately 60% heavy REEs, i.e., Dy, Yb, Er, 50% middle REEs, such as Nd, Eu, Sm, Gd, and 30% light REEs such as La, Pr, Ce, can be selectively extracted out of Bayer red mud with diluted nitric acid (0.5 M) under moderate dissolution conditions, without using any prior treatment. This was investigated by several authors [55,92,122].

4.2. Biometallurgical Recovery of Rare-Earth Metals from Red Mud

A branch of hydrometallurgy and biotechnology known as “biohydrometallurgy” uses microorganisms to extract metals and valuable products from minerals and secondary sources. Resource conservation, pollution avoidance, and environmental remediation are among the environmental issues that biohydrometallurgical technology is being developed to address at the moment [123,124]. Examples include the separation of radionuclides and metal values from heavy nuclear waste and other waste sources [125], the elimination of dispersed oil from oily wastewater, the recycling of plastics, the recycling of waste paper to create clean cellulose products, and the production of clean coal, which all serve to highlight the value and necessity of developing improved biohydrometallurgical technology [126,127]. The usage of this method is likely most notable in the bauxite residual mineral business, where over 800,000 tons of bauxite minerals are treated by the Bayer process. In Turkey alone, 1.5 Mt of red mud is produced, and 10 Mt of bauxite residues are stored in the country’s tailings ponds [128].

Biohydrometallurgy and bioleaching are based on microbiological processes that result in the eventual recovery of metals. In actuality, abiotic physical and chemical processes are often used to process metal-bearing minerals, whereas biohydrometallurgy makes use of the fact that microorganisms are influencing mineral transformations [49,124]. Chemical transformations and transport processes may be amplified by interactions between microorganisms and minerals [129]. It is determined which microbes control biologically enhanced changes. It is possible to apply biological processes to create novel techniques for bioprocessing minerals. All metals may be used in biohydrometallurgical processes [130]. In the past, copper was passively recovered by biohydrometallurgy for centuries, without much understanding of the precise nature of the phenomenological process. There are two primary methods of bioleaching: “direct bioleaching”, which involves extraction/leaching through metabolic activity, and “indirect bioleaching”, which depends on the action of metabolic chemicals generated during microbial development. Due to the increased pH and lack of electron donors and energy sources such as reduced iron or sulfur, autotrophic bacteria are not appropriate for recovering metals from RM [131]. Metal leaching from red mud has been accomplished by heterotrophic bacteria. They also provide a number of benefits over other methods [132]. They may survive in extremely alkaline conditions, releasing metabolites such as organic acids, amino acids, and proteins at the same time. These metabolites can form complexes with the poisonous metal ions found in RM [133]. As a result, there is less harm caused to the microbes’ metabolic processes [134,135]. It has been demonstrated that a number of heterotrophic microbial strains are effective at recovering REEs from a variety of solid matrices [136]. Numerous research studies in the last several decades have concentrated on how microorganisms interact with REEs, including REE mobilization from solids via metabolic interactions and REE immobilization from liquids via sorption. Rasoul-nia et al. [137] extensively reviewed the recovery of rare-earth metals from various sources

with their detailed mechanisms, such as complexosis, acidolysis, redoxolysis, etc., during the bioleaching process [137]. The biohydrometallurgical processes provides a low-cost, energy-efficient “green approach” to REE recovery [39]. The schematic representation of rare-earth metal recovery using the biohydrometallurgical process is shown in Figure 6.

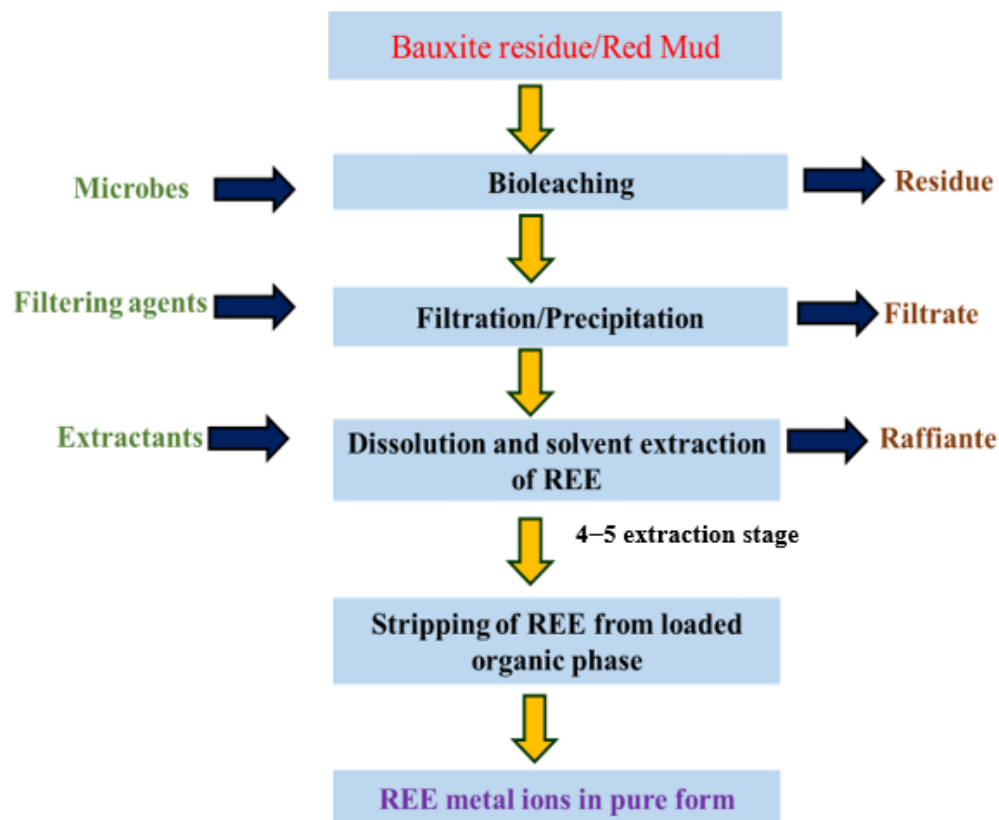


Figure 6. Schematic flow sheet for the recovery of rare-earth metals from red mud using a biohydrometallurgical approach.

The capacity of three heterotrophic fungi, *Aspergillus niger*, *Penicillium tricolour*, and *Penicillium crustosum*, to extract metals from RM was examined [138–140]. So far, three methods have been discussed: (1) one-step bioleaching, in which microorganisms are grown in the presence of RM; (2) two-step bioleaching, where fungi are grown first without RM, then RM is added; and (3) cell-free wasted media, in which the solution is filtrated and then added to RM. According to recent research by Qu and Lian [134], employing the fungus *Penicillium tricolour*, REEs and radioactive elements were recovered from red mud in both steps of bioleaching tests. The two-step bioleaching technique generated the highest extraction yields, with a pulp density of 10%. Furthermore, heavy metal bioleaching from red mud has been connected to *Aspergillus niger* [49,141]. The organic acids from fungi’s metabolic products considerably aided the bioleaching procedure through chelation or complexation. The two-step approach, which entails first pre-culturing the fungus in a medium of sucrose for 72 h to produce a visible increase in biomass before adding sterilized red mud, offers the highest separation yields, from 20% to 75%, at a pulp density of 10% (*w/v*). The wasted medium bioleaching methodology outperformed the one- and two-step bioleaching procedures for leaching heavy metals when *A. niger* was used as the leaching fungus. Additionally, the need to separate the cells from the REEs following the bioleaching process in one- and two-step processes might result in a loss of REEs [49].

A more recent study demonstrated the capacity of the *Aspergillus* strains *A. niger* and *A. flavus* to efficiently extract significant quantities of REEs from lower carboniferous carbonaceous shale [142]. To extract Lu, Y, Sc, and Th, a chemo-heterotrophic bacterium that was isolated from RM was tested as a leaching agent [143]. Organic acids, namely

acetic, malic, oxalic, and lactic acids that were released into the medium in this instance acted as the leaching agents. The separation of Sc, Y, Ce, and from RM was evaluated using a thermophilic autotrophic archaeon such as *Acidianus manzaensis*, although RM does not provide an energy source for autotrophic bacteria [49]. In order to give *A. manzaensis* a source of energy, pyrite was introduced to the medium, along with RM during the batch mode of the experiment [144]. *A. niger* and *P. simplicissimum* have recently been studied using sugarcane molasses as a carbon source high in sucrose. The findings show that waste sludges may be employed as a substrate for RM leaching, using fungus to recover alumina [141]. Due to the existence of an annotated genome sequence, Reed et al. [145] reported that the strain of *G. oxydans* (B58) is more efficient at leaching REE than the isolates. Furthermore, the author stated that to improve acid production and obtain even better REE recovery, genetic alterations and/or adjustment of culture conditions would be taken into consideration [145]. Recent research by Kiskira et al. [146] examined the bioleaching of red mud using various microbial cultures and solid-to-liquid ratios. Using *Acetobacter tropicalis* in a one-step bioleaching procedure at 1% pulp density, the highest Sc extraction yield was 42% [146]. When evaluating the economic worth of the recovered chemicals, the value of the electron donor should be taken into consideration. However, using inexpensive trash as fuel allows for the recovery of valuable chemicals, which is in line with sustainability principles.

5. Recommendation and Conclusions

The production of red mud is increasing in the world due to the high need of aluminum. Since the residue from this process contains an important quantity of REEs, which is, in some cases more than 1000 ppm. The idea of producing green technology devices makes REEs more attractive and critical to the industry. However, the recycling of red mud for REE extraction by using a low-cost and green process would be interesting.

REE extraction from this bauxite residue will contribute to reducing the importation of these elements worldwide. Therefore, in-depth studies could be performed to fully understand these residues' mineralogical and chemical composition. This will help in choosing a suitable method of treatment. However, existing methods are generally less effective and time-consuming. Research could be carried out in this direction in order to set up an efficient and economical technology for REE extraction from red mud. Chemical leaching has been shown to be an efficient process in terms of time and metal recovery; however, the using of toxic chemicals makes residue management complicate. Using an ecofriendly reagent such as light or organic acid could be tested instead of using a strong acid and an alkaline chemical. On the other hand, bioleaching has been shown to be an ecofriendly and low-cost process; however, the slow leaching time and low pulp density make its application complicate at a large scale. Since red mud is a highly alkaline material and contains fewer sources of energy for acidophilic microorganisms, research could be directed towards heterotrophic microorganisms to choose a suitable one. In addition, bioengineering studies can be undertaken to improve their acid production. For each process, the application of life cycle assessment (LCA) is important to evaluate their effect on the environment.

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