Recent Research on Recovery of Iron and Aluminium from Bauxite Residue in China

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Abstract

Bauxite residue or red mud is a by-product of alumina extraction from bauxite. Bauxite residue is a very valuable secondary resource as it contains high alumina, iron oxide, titanium oxide, calcium oxide, silica and soda concentrations. In addition, it contains low concentrations of scandium, gallium, vanadium, and other minor elements. Extraction and utilization of metal resources such as iron and aluminum from the red mud can help solving the problem of large-scale storage of red mud, which is of great significance. This article summarizes the recent R&D focused on iron and aluminum recovery from red mud. It highlights the high degree of extraction and recovery (> 90 %) that have been achieved through high-temperature processing and leaching.

Keywords: Bauxite residue, Red mud, Iron and aluminum, Recovery.

1. Introduction

Bauxite residue or red mud is a solid by-product or waste produced during the extraction and production of alumina. This residue is essentially the components of bauxite that are do not dissolved fully in the digestion of bauxite ores and consists of sodium silicate, calcium silicate produced in the Bayer process. The production of 1t of alumina produces 1~2 t of red mud. In recent years the production of red mud in China has reached 100 million t/year. In Figure 1, the current trend covering Chinese production of red mud from 2012 to 2022 is shown. This upward trend is mainly due to the increased production rate of alumina in China. The production of red mud in the world is expected to exceed 150 million t/year, and currently, the estimated mass of stored red mud is about 4 billion tonnes.

At present, there are 53 alumina production enterprises in China, mainly distributed in nine provinces and regions of Shandong, Henan, Hebei, Shanxi, Guizhou, Chongqing, Guangxi, Yunnan and Nei Mongol. Red mud is often treated by open-air storage, which is mainly divided into wet storage and dry storage. In China, dry storage is the main method. Because a large amount of red mud has not been fully utilized for the time being, it not only occupies land for a long time, it has increased maintenance cost, and has an impact on the environment, which directly restricts the sustainable development of the alumina industry.

It is well known that red mud has strong alkalinity, which makes it difficult to process and achieve complete utilization. How to deal with it is still a worldwide problem. At present, the “green utilization” of red mud in China mainly includes extracting valuable metals [1-3], preparing cement and concrete [1,3], preparing ceramics [3], composite materials [4], and so on.

As mentioned earlier, red mud contains iron, aluminum, silicon, calcium, and a small amount of scandium, vanadium, zirconium, and other rare metals. The accumulated reserves of red mud are considerable, and this resource has a large recycling potential; thus, it can be considered a precious secondary resource. Due to the differences in the composition of bauxite ores and various practices used for the extraction of alumina in different countries, the composition of red mud produced varies considerably. Table 1 lists the main components of red mud that are produced in some countries and shows that Chinese red mud has much lower concentration of iron oxide and higher levels of silica and lime.

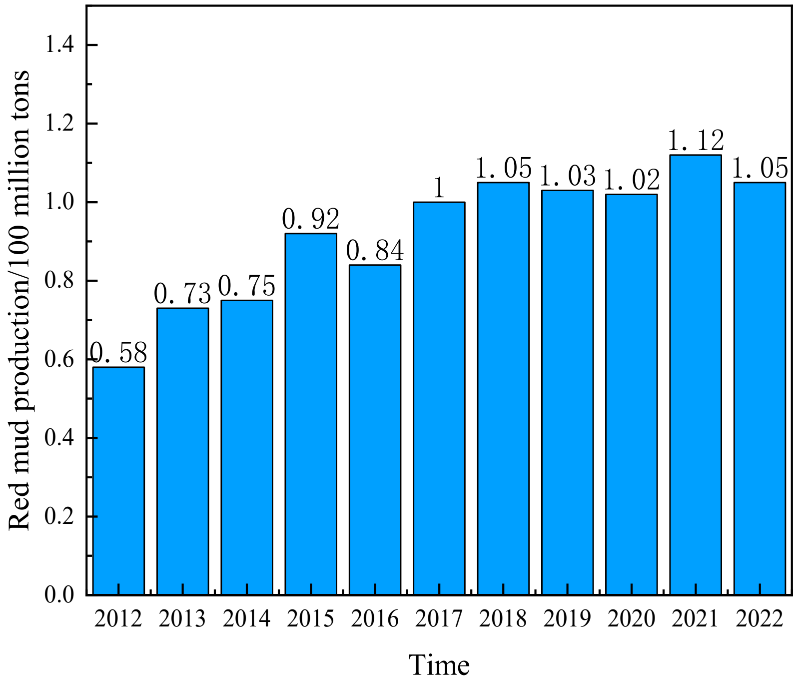

Figure 1. Red mud production in China from 2012 to 2022.

Table 1. Chemical composition of red mud from different countries (%).

Country	Fe_2O_3	Al_2O_3	SiO_2	CaO	Na_2O
France	42.00	14.00	6.00	/	2.00
Greece	45.03	17.22	7.15	8.47	2.65
India	36.69	20.01	6.51	1.43	5.09
Jamaica	46.78	16.32	6.35	4.76	3.56
Romania	44.06	18.51	10.94	4.28	5.0
United States	35.29	17.15	11.22	9.64	5.07
China	16.91	15.01	17.55	23.49	4.60

In the present paper the recent research progress on the recovery of iron and alumina from red mud in China and abroad are reviewed and compared. The main processing steps include direct magnetic separation, roasting reduction-magnetic separation, melting reduction-magnetic separation and leaching iron and aluminum from red mud. The purpose of this paper is to identify a new, simple, and efficient process for iron recovery and to provide a reference for the recovery of iron and aluminum in red mud.

2. Research Status of Iron and Aluminum Recovery in Red Mud

Red mud contains a rich variety of elements and can be used as an important secondary resource for recycling and utilization. Iron in red mud mainly exists in hematite, goethite and other minerals, and aluminum mainly exists in diaspore and tridiaspore and other minerals. At present, scholars at home and abroad have conducted extensive research on the recovery of iron and aluminum from red mud. The recovery process of iron from red mud mainly includes direct magnetic separation, roasting reduction-magnetic separation, suspension magnetization roasting magnetic separation, hydrometallurgy and so on. The processes for recovering aluminum from red mud mainly include sintering, calcification carbonization, calcination and hydrometallurgy.

2.1 Recovery of Iron by Physical Separation

Li et al. [5] used gravity separation to recover iron. The recovery of total iron was between 23 % and 43 % after the concentrator was used for concentration and desliming. Under the conditions of 25 % material concentration, 35 Hz vibration frequency and 6 r/min of rotation speed, the red mud was sorted by the combination of "two-stage cyclone + suspended vibration cone separator", and the iron grade was 49 %. The gravity separation method is based on the difference in density of red mud particles and the directions and speeds of movement in the fluid medium. The gravity separation process is relatively simple, easy to operate, low investment cost, no pollution. However, due to the small particle size of the red mud particles, some agglomeration between the phases can occur, making good separation rather difficult.

The magnetic separation method uses a magnetic field to separate magnetic minerals from non-magnetic particles in untreated red mud. This is a suitable method for red mud with high magnetite content. In general industry, red mud is roughed by magnetic separation, and an iron concentrate is obtained. Wang et al. [6] pre-enriched the iron minerals in the samples by high intensity magnetic separation. The optimal conditions were as follows: the proportion of grinding samples with a particle size of 0.074 mm accounted for about 81 %, and the magnetic induction field intensity of high-intensity magnetic separation was 1.2 T. Under these conditions, iron concentrate with an iron grade of about 53 % and an iron recovery rate of about 60 % could be obtained. Generally, the magnetic separation equipment runs stably and consumes little energy. The pretreatment of red mud before magnetic separation, that is, grinding - classification - magnetic separation, can effectively improve magnetic separation efficiency. However, by using a single magnetic separation, it is difficult to capture the attached particles formed by iron and aluminum, silicon and calcium, and multiple magnetic separation costs are high, waste energy and the impurity content in iron concentrate cannot meet the standard.

2.2 Recovery of Iron by Roasting Reduction-Magnetic Separation

Due to the complex occurrence of metal ions in red mud, physical mineral processing is difficult to achieve the ideal goal, so many scholars have used pyrometallurgy process to achieve the separation of iron and aluminum. The roasting-reduction-magnetic separation process is to mix red mud, reducing agents and additives finely, and then roasting and reducing at high temperature to convert hematite in red mud into magnetite or simple iron, and finally recover iron through magnetic separation.

Zhu et al. [7] adopted the method of sodium carbonate roasting, reduction and magnetic separation to recover iron. They found that: when the reduction temperature was 1050 ℃, the reduction time was 80 min, the addition of sodium carbonate was 8 %, and the magnetic separation was carried out at the magnetic field intensity of 0.08 T, the total iron recovery rate could reach about 95 %. However, this method needs to be carried out under high temperature conditions, which has the problems of high energy consumption. Xu et al. [8] used hydrogen and carbon powder as reducing agents, and conducted direct reduction roasting under the conditions of 1000 ℃, reaction time 120 min and carbon/red mud mass ratio of 1: 5. Then, the roasted red mud was subjected to fine magnetic separation. The grade of iron powder obtained was about 93 %, and the iron recovery rate was about 79 %. This process is the main method to recover iron from red mud, and it has the advantages of a simple operation and low investment cost. It is difficult for direct magnetic separation to effectively recover iron from red mud, and the hematite in red mud is basically transformed into magnetite by roasting reduction. Secondary magnetic separation is conducive to improving the recovery of iron, but the powder size should not be too fine during magnetic separation, which is easy to reduce the grade of iron powder.

Rao et al. [9] added sodium sulfate and sodium carbonate as additives with a mass fraction of 6 %, and reacted at a reduction sintering temperature of 1050 ℃ for 60 min to convert goesite and hematite in the original red mud into metallic iron, and then carried out grinding and weak magnetic separation to obtain an iron concentrate with a total Fe content of about 90 % and an iron recovery rate of about 95 %. Ding et al. [10] added potassium chloride and sodium sulfate as additives, and 10 % pulverized coal as reducing agent. Under the conditions of reduction sintering temperature of 1100 ℃ and reaction time of 65 min, the direct reduction sintering method was used to reduce the weak magnetic hematite in the red mud to produce metallic iron. After 0.12 T weak magnetic sorting, about 79 % of the total Fe content was obtained from iron concentrate, with iron recovery of about 85 %. In the above two studies, although the addition of sodium salt can reduce the reduction sintering temperature and improve the grade and recovery of iron concentrate, the aluminum minerals in the red mud are not separated and recovered, resulting in high Al_2O_3 content in the non-magnetic tailings and serious waste of aluminum resources. Therefore, it is necessary to further improve the process to achieve high recovery rate and low carbon emission rate.

2.3 Recovery of Iron by Suspension Magnetization Roasting and Magnetic Separation

The suspension magnetization roasting-magnetic separation process is an innovative technology for treating high-iron red mud. During the magnetization roasting process, the weak magnetic hematite in the red mud can be reduced to strong magnetic magnetite, and then the iron concentrate can be magnetized.

Liu et al. [11] showed that under the conditions of roasting temperature 540 ℃ and roasting time 15 min, CO gas with a volume fraction of 30 % was injected as a reducing agent, and after roasting and magnetic separation, iron concentrate with an iron grade of about 56 % and an iron recovery of about 88 % was finally obtained. Zhang et al. [12] changed the CO concentration to 20 %, roasting temperature to 560 ℃, roasting time to 10 min, and finally obtained an iron concentrate with an iron grade of about 57 % and an iron recovery of about 90 %. The method has low roasting temperature, short reaction time, environmental protection and no emission, and provides a new method for the recovery and treatment of iron in red mud.

2.4 Recovery of Aluminum by Sintering Method

Chen et al. [13] studied the extraction of Al_2O_3 from Bayer red mud by low calcium sintering method. The results show that the dissolution rate of Al_2O_3 in clinker is 86 % after sintering at 1150 ℃ for 90 min. However, high sintering temperature and long sintering time will cause the clinker to combine more closely, which is not conducive to the dissolution of alumina. Xiong et al. [14] used Bayer process red mud and refractory high-sulfur bauxite as raw materials to study the alkaline roasting and recovery of Al_2O_3. The results show that under the optimal roasting and dissolution conditions, the dissolution rate of Al_2O_3 can reach 92 %. However, the concentration of NaOH required in this method is 18 g/L, and the alkali consumption is large. Fan et al. [15] studied red mud melting by high temperature method, and then leaching aluminum from molten slag with sodium carbonate. The results showed that under the conditions of mass concentration of sodium carbonate of 100 g/L, liquid-solid mass ratio of 10/1, temperature of 80 ℃, and melting time of 90 min, the leaching rate of Al_2O_3 could reach 91 %, and the tailings after leaching of aluminum could be used for cement production.

At present, the fire process is relatively mature, with the advantages of simple process, large processing capacity and strong applicability, but there are also problems such as high energy consumption, large amount of waste residue and heavy pollution.

2.5 Recovery of Aluminum by Calcification and Carbonization

Calcification and carbonization method for extracting aluminum from red mud is a new method developed in recent years. By calcification transformation and dealkalization of red mud, silicon containing minerals are transformed into sodium free hydrated garnet. The transformed red mud reacts with CO_2 (carbonization decomposition), and then aluminum oxide is extracted through alkaline solution aluminum process.

Zhu et al. [16] studied the recovery of aluminum from red mud using the calcification and carbonization method. The results showed that under the conditions of calcification temperature of 120 ℃, calcium silicon ratio of 1.5, liquid solid ratio of 4/1, calcification for 120 minutes, and carbonization temperature of 80 ℃, CO_2 pressure of 1.2 MPa, and liquid solid volume ratio of 5/1 for 120 min, the recovery rate of aluminum oxide in red mud was 76 %. Xie et al. [17] found through a comparative study of the traditional calcification carbonization method and the continuous method (after calcification, the solution is directly carbonized without solid-liquid separation) that the continuous method can greatly simplify the production process. In order to improve the recovery of alumina extracted from red mud by the calcification carbonization method, Liu et al. [18] conducted a wet grinding study on calcified slag, The results show that wet grinding can destroy the dense structure of spherical water garnet, and prolonging the grinding time and accelerating the grinding rate are helpful for the recovery of Al_2O_3. The calcification-carbonization method can effectively recover aluminum, significantly reduce the alkali content in tailings, and facilitate the direct preparation of building materials from tailings, in order to achieve repeated utilization of red mud and reduce environmental pollution.

2.6 Recovery of Iron and Aluminum by Hydrometallurgy

Metallic elements in red mud generally exist in the form of oxides and are usually leached from iron and aluminum in red mud using inorganic or organic acids. Many researchers have conducted extensive acid-leaching experiments on red mud. Considering that red mud contains a number of alkaline metal oxides, when using the acid method to treat high iron red mud, it is usually to leach and recover iron oxide together with one or more other alkaline metal oxides. Currently, the acids used for separating and recovering iron and aluminum resources from iron red mud mainly include sulfuric acid, hydrochloric acid, nitric acid, oxalic acid, etc.

Pepper et al. [19] studied the leaching rate of red mud under different acid concentrations by using the acid leaching method of nitric acid, hydrochloric acid, sulfuric acid and phosphoric acid. The experimental results showed that the leaching rates of iron were the highest after treatment with phosphoric acid and hydrochloric acid, ranging from 76 % to 78 %, and the recovery rate of aluminum was the highest after treatment with phosphoric acid, reaching 50 %. But this method requires a high acid concentration, consumes a large amount of acid, and has a long reaction time. Lu et al. [20] used hydrochloric acid leaching to recover Al_2O_3 and Fe_2O_3. Through research, it was found that Al_2O_3 and Fe_2O_3 in red mud after acid leaching reaction exist in the form of hydrated ions in hydrochloric acid solution. Increasing the concentration of hydrochloric acid, acid leaching temperature, reaction time, and liquid-solid ratio are beneficial for promoting the leaching reaction of Al_2O_3 and Fe_2O_3 in high-speed iron red mud. Under the optimal acid leaching conditions, the leaching of alumina and iron oxide can reach over 90 % and 95 %, respectively. Although this method has a high leaching rate, the simultaneous leaching of multiple metals is not conducive to the subsequent separation of iron and aluminum.

In addition to the commonly used acids mentioned above, waste acid generated by the steel industry can also be used as leaching solution to achieve waste utilization and save costs. Liu [21] proposed using H_2SO_4, a byproduct from the sintering process in the steel industry, as a leaching agent to leach red mud. The recovery of Fe and Al was achieved through steps such as leaching, co-precipitation, Fe/Al separation, and calcination. The final recovery rate of Fe is 73 %, and the recovery rate of aluminum is 91 %. This method comprehensively utilizes two types of waste to turn waste into treasure, but the separation and recovery process is complex, and the amount of H_2SO_4 added has the greatest impact on the leaching rate of aluminum and iron, which is not conducive to further industrial application.

2.7 Combining Different Processes to Recover Iron and Aluminum

Xiao et al. [22] developed a process based on alkali lime sintering, leaching, and magnetic separation, which achieves the recovery of aluminum and iron through the simultaneous treatment of red mud and phosphogypsum. When the sintering temperature is 1100 °C, the sintering atmosphere is N_2, the sintering time is 30 minutes, the C/S ratio (molar ratio of CaO to SiO_2) is 2.0, and the N/A ratio (molar ratio of Na_2O to ($Al_2O_3 + Fe_2O_3$)) is 1.3, the aluminium recovery rate, iron recovery rate, and iron grade are 69 %, 78 %, and 84 % respectively. The simultaneous treatment of red mud and phosphogypsum can reduce waste emissions, but due to the small particle size of the reduction product under N_2 - CaO conditions, separation is difficult, and the grade of iron decreases. Zhao et al. [23] studied the roasting modification of Bayer process red mud at 900 °C, followed by sulfuric acid leaching of aluminum and iron. The results showed that under the conditions of H_2SO_4 concentration of 45 %, liquid-solid volume mass ratio of 9 mL/g, and temperature of 100 °C for 60 minutes, the leaching rate of Al_2O_3 was 93 %, and the leaching rate of Fe_2O_3 was 90 %. The leaching solution can be used to prepare polymeric aluminum iron sulfate, achieving comprehensive recovery and utilization of red mud. Yu. et al. [24] studied the leaching of red mud with industrial waste sulfuric acid, the reduction of Fe^{3+} with waste iron, and the addition of ammonia to precipitate iron and aluminum in the form of hydroxides, The results showed that under the conditions of H_2SO_4 concentration of 40 %, liquid-solid volume mass ratio of 3.5/1, and temperature of 110 °C for 300 minutes, the leaching rates of iron and aluminum were 96 % and 94 %, respectively.

This method utilizes two types of waste to turn waste into treasure, but the separation and recovery process is complex. The amount of H_2SO_4 added has the greatest impact on the leaching rate of aluminum and iron, which is not conducive to further industrial application.

Table 2 summarizes the different methods involved in recovering iron and aluminum from the above red mud and gives representative process conditions and recovery effects.

Table 2. Recovery technologies of iron and aluminum from red mud.

Method	Technique	Technological condition	Product	Reference
Physical	Gravity separation	Concentration 25 %, vibration frequency 35 Hz, 6 r/min	TFe: 49 % IR: 23–43 %	[5]
	Magnetic separation	Grinding: -0 074 mm accounted for 80 75% MFI:1.2T	TFe: 53 % IR: 60 %	[6]

Roasting reduction-magnetic	Reduction roasting magnetic	Reductant: sodium carbonate 1050 ℃, 80 min, Na_2CO_3 addition: 8 %, MFI: 0.08 T	TFe: 96 %	[7]
	Reduction roasting magnetic	Reductant: hydrogen, carbon powder, 1000 ℃, 120 min, C:red mud = 1:5	TFe: 93 % IR: 79 %	[8]
		Additive: 6 % sodium sulfate and sodium carbonate 1050 ℃, 60 min	TFe: 90 % IR: 95 %	[9]
		Additives: potassium chloride, sodium sulfate, reducing agent: 10 % coal powder, 1100 ℃, 65 min	TFe: 79 % IR: 85 %	[10]
Suspension magnetization roasting magnetic	Suspension magnetization roasting magnetic	540 ℃, 15 min, CO: 30 %	TFe: 56 % IR: 88 %	[11]
		560℃, 110 min, CO: 20 %	TFe: 57 % IR: 91 %	[12]
Sintering process	Low calcium reduction sintering	1150 ℃, 90 min	Dissolved alumina: 86 %	[13]
	Alkaline roasting	Roasting: 1100 ℃, dissolution: 80 ℃, 25 min, L/S=10, NaOH: 18 g/L, Na_2CO_3: 8 g/L	Dissolved alumina: 92 %	[14]
	Smelting leaching	Na_2CO_3: 100g/L, L/S=10, 80 ℃, 90 min	Dissolved alumina: 91 %	[15]
Calcification carbonization	Calcification carbonization	Calcification: 120 ℃, 120 min, C/S=1.5, L/S=4 Carbonization: 80℃, 120 min, CO_2 1.2 MPa, L/S=5	AR: 75 %	[16]
	Continuous method	NaOH: 100 g/L, 120 ℃, 90 min	AR: 36 %	[17]
	Wet grinding	80 rpm, 60 min, L/S=2, ball-to-powder ratio: 12	AR: 44 %	[18]
Hydrometallurgy	Acid leaching	Phosphoric acid: 1 mol/L, 25 ℃, 1440 min	IR: 76 % AR: 50 %	[19]
	Hydrochloric acid leaching	HCl: 6 mol/L, L/S=2, 109 ℃, 60 min	IR: 98 % AR: 89 %	[20]
	Waste sulfuric acid leaching	85 ℃, 360 min, L/S=30	IR: 73 % AR: 91%	[21]

Combination of different processes	Sintering leaching	1100 ℃, 30 min, C/S=2.0	IR: 78 % AR: 69 %	[22]
		H_2SO_4: 45 %, 100 ℃, 60min, L/S=9	IR: 89 % AR: 93 %	[23]
		H_2SO_4: 40 %, 110℃, 300min, L/S=3.5	IR: 96 %, AR: 94 %	[24]

MFI: magnetic field intensity; IR and AR: iron and alumina recovery; C/S: molar ratio of CaO to SiO_2; L/S: liquid-solid ratio.

3. Conclusion

There are mainly pyrometallurgical and hydrometallurgy processes for recovering aluminum and iron. The following conclusions could be arrived from the present review:

(1) When recovering iron from red mud, the physical separation treatment of red mud has low emission, large processing capacity and is easily managed. However, the obtained iron concentrate has low iron grade, high impurity content, and needs to be broken and pulverized several times to treat the tailings. This method is more suitable for raw material pretreatment or tailings post-treatment.

(2) The roasting reduction-magnetic separation method for iron recovery can improve iron grade, but this method needs to be carried out under high-temperature conditions; there are issues with large raw material and energy consumptions, which do not meet the green production concept.

(3) As an emerging technology, suspension magnetization roasting and iron recovery technology has the advantages of high efficiency, large production capacity, and good utilization of heat energy. This method can achieve a double improvement of iron grade and recovery rate and has a good application prospect, which is worthy of further attention.

(4) When using pyrometallurgy to recover aluminum, it is generally necessary to mix red mud with certain additives, sintering in a high temperature and pressure environment, and then to use alkali dissolution or carbonization for recovery. This method has the advantages of a simple process, large processing capacity and strong applicability, but it also has some problems, such as high energy consumption, large amounts of waste residue and heavy pollution, which does not meet the current requirements of environmental protection.

(5) The hydrometallurgical process can avoid the above problems compared to the pyrometallurgical process. The acid leaching process has the advantages of high recovery rate of iron and aluminum, low energy consumption and simple process, but a large number of other ions are leached at the same time during the acid leaching process, and how to effectively separate iron and aluminum from other metal ions is also a challenge. In addition, red mud is strongly alkaline, requiring a large amount of acidic solution for neutralization and high requirements for acid resistance of equipment and large investment.

(6) The use of waste acid leaching is a better idea. Combining the two kinds of waste can achieve waste utilization, improve the recovery rate of aluminum and iron, and not only save costs but also reduce waste emissions. How to use waste acid to treat red mud and develop a set of technology that can separate metal ions in the leaching solution is still the direction of research.

In conclusion, suspension magnetization roasting technology is a new technology, which has the characteristics of high efficiency and high recovery of iron from red mud, and has good application prospects. Recycling iron and aluminum from red mud with waste acid can realize the comprehensive utilization of waste and has the advantages of low energy consumption and high alumina leaching rate, which is worthy of further study.

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