

# Iron Recovery Technology of Red Mud—A review

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**Abstract:** RM (red mud), which comes from the Bayer process, has a huge annual output and is harmful to the environment. Because of the high iron content in RM, the process of iron recovery from RM can reduce the amount of RM well and create economic benefits, so it is a promising process. The paper focuses on the review on the research of the iron recovery method from RM, which includes the physical recovery method, chemical recovery method and emerging recovery method. By comparing the advantages and disadvantages of these processes, it is concluded that the fluidized bed reduction is a promising process that can be rapidly applied to the industry, but it still needs more investigation to overcome the current technical difficulties in the near future.

**Keywords:** red mud; iron recovery; pyrometallurgical method; fluidized bed reduction



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## 1. Introduction

As global aluminum production continues to increase, as the main by-product, the output of RM has also risen. On average, 1 to 1.5 tons of RM are produced for every ton of aluminum produced [1], and about 67–100 million tons of RM are produced annually in whole world [2]. So far, RM reserves have exceeded 2.7 billion tons [3]. How to utilize the RM has become an urgent problem to be solved.

Many scholars reviewed the utilization methods of RM, including utilization in road base and subgrade material [4], environmental remediation material [5], catalyst [6] and element recovery [7], etc. In these processes, due to the high iron content in RM, the iron recovery process can not only realize the decrement, innocuity and resource recovery but also create good economic benefits. Therefore, the iron recovery process from RM is becoming a hot topic [8]. The paper reviews the iron recovery process and gives the conclusions and prospects of the iron recovery process by comparing the advantages and disadvantages of each process.

## 2. Research Methodology

The purpose of this article is to review the progress of research on recovered iron processes and to show as many different processes as possible. By searching from Web of Science, ScienceDirect and CNKI based on the term “red mud”, several published articles about iron recovery from RM have been selected in the article. The authors hope that this review article inspire scholars studying related fields.

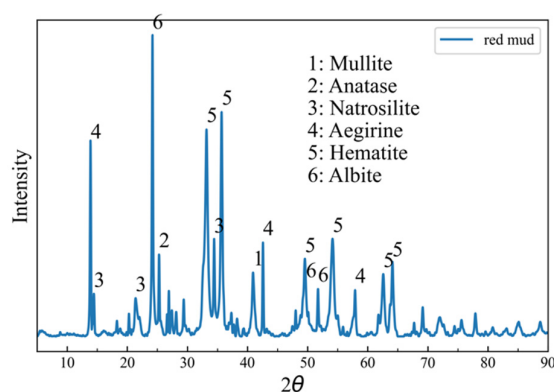
## 3. The Production, Properties and Harm of Red Mud

There are three main processes for producing aluminum hydroxide, including the sintering method, the Bayer method, and the combined method. Among them, the aluminum hydroxide produced by the Bayer method accounts for about 95% of the total due to its simple process and low energy consumption [9]. In the article, the utilization of Bayer RM is discussed.

The most important ore used in the Bayer method is bauxite. The bauxite mainly consists of alumina trihydrate ( $\text{Al}(\text{OH})_3$ ), bomite ( $\gamma\text{-AlO}(\text{OH})$ ) and alumina ( $\alpha\text{-AlO}(\text{OH})$ ), and it also contains kaolinite ( $\text{Al}_4[\text{Si}_4\text{O}_{10}](\text{OH})_8$ ), goethite ( $\text{FeO}(\text{OH})$ ), hematite ( $\text{Fe}_2\text{O}_3$ ), anatase ( $\text{TiO}_2$ ) and ilmenite ( $\text{FeTiO}_3$  or  $\text{FeO}\cdot\text{TiO}_2$ ) [9,10]. The steps of the Bayer process are as follows [11]: the bauxite is pulverized and mixed with lime and sodium hydroxide, and the aluminum hydroxide slurry and solid residue are obtained after sedimentation and separation. The solid residue is red due to a large amount of iron oxide, so it is called red mud. Some scholars comprehensively summarized the chemical compound of RM [3,9,12], which varies widely with raw materials and processes. The chemical compound and mineral composition of RM used in a previous experiment [13] are shown in Table 1 and Figure 1, respectively. The main elements in RM include iron, silicon, titanium from bauxite, residual aluminum, sodium from sodium hydroxide and calcium from lime. Trace elements include rare earth elements, U, Th and other radioactive elements. The iron content of RM reaches more than 20% and even more than 40% in high iron RM.

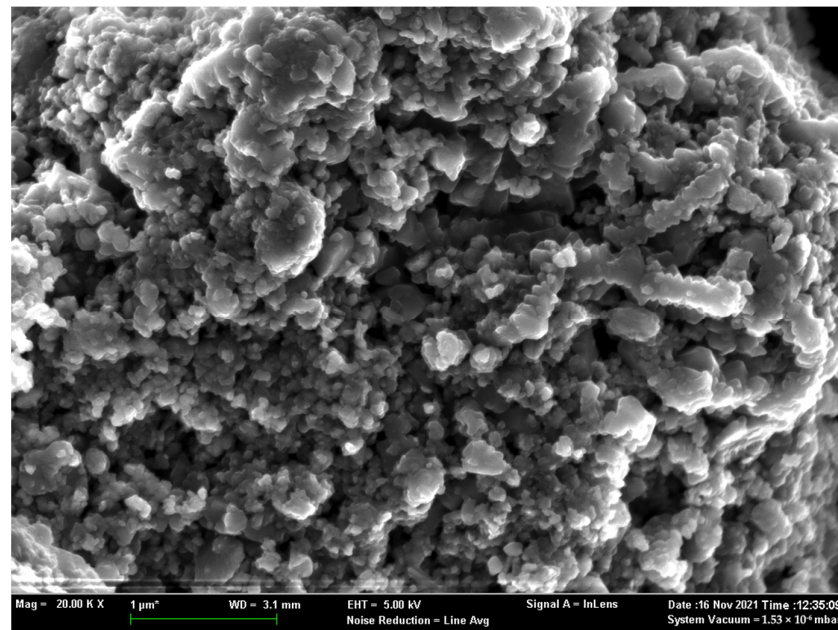
**Table 1.** Chemical compound of RM used in previous experiment [13].

| Compound                | Wt %  |
|-------------------------|-------|
| $\text{Fe}_2\text{O}_3$ | 37.68 |
| $\text{Al}_2\text{O}_3$ | 22.69 |
| $\text{SiO}_2$          | 18.22 |
| $\text{Na}_2\text{O}$   | 12.37 |
| $\text{TiO}_2$          | 6.2   |
| $\text{CaO}$            | 0.947 |
| $\text{ZrO}_2$          | 0.543 |
| $\text{NiO}$            | 0.238 |

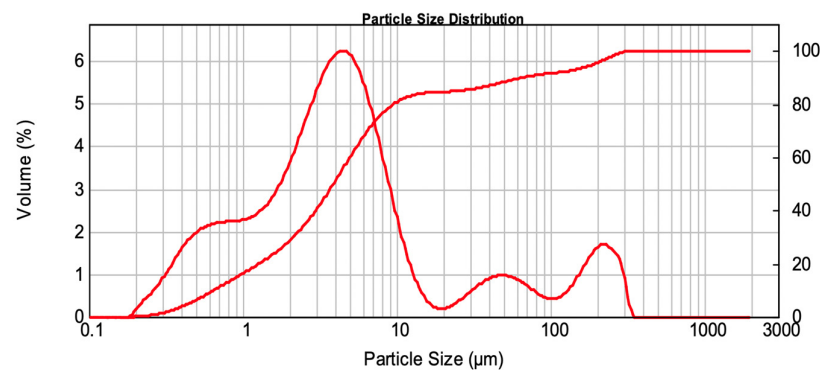


**Figure 1.** Mineral composition of RM used in previous experiment [13].

RM has the characteristics of fine particles, porosity, strong alkalinity and radiation [14]. As shown in Figures 2 and 3, RM have many pores inside, and most of the RM has a particle size of less than 10  $\mu\text{m}$ . At present, most of the RM is disposed by stacking, which will not only increase the cost but also cause serious pollution to the environment. Due to the fine particle size of RM, it will be entrained by wind and pollute the air when it is stacked in the open air. It will leak into the ground when it rains and will cause water pollution and corrosion of buildings. The radioactivity of RM will also cause damage to the environment. On 4 October 2010, the dam wall of the Ajka oil refinery collapsed and the surrounding area was flooded with RM, causing serious pollution to the Danube River [15–18].



**Figure 2.** SEM image of RM used in previous experiment [13].



**Figure 3.** The particle size distribution of RM used in previous experiment [13].

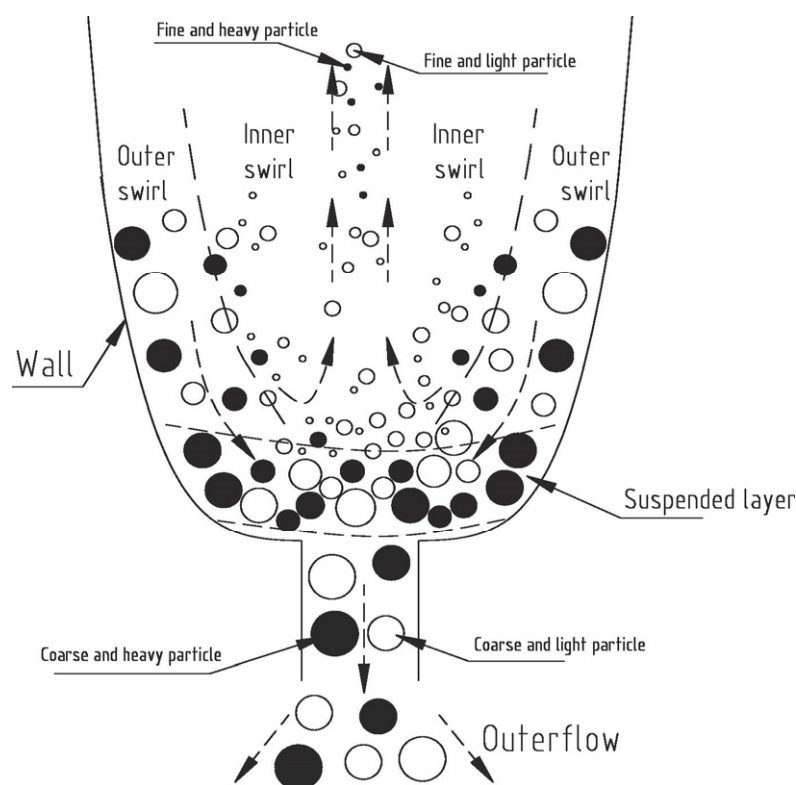
#### 4. Iron Recovery Process from Red Mud

Since both RM and iron ore contain a variety of metal elements, most of the iron recovery processes from RM reference the experience of mature processes in the metallurgical industry, such as the physical, hydro and pyrometallurgy method. There are also emerging processes such as the biological method, electrochemical method, etc. The research progress of these processes is introduced in the following sections.

##### 4.1. Physical Recovery Method

The physical recovery method is to directly separate iron-bearing minerals based on the different physical properties and includes gravity separation, magnetic separation and flotation.

Gravity separation refers to the process of separating the mineral particles by the difference in relative density, particle size, shape and movement speed and direction in the medium. Liu et al. [19] adopted a combination of a two-stage cyclone and suspension cone concentrator for separation, and they obtained concentrate with the TFe of 48.83%. Jiang et al. [20] replaced the traditional straight cones with the cubic parabolic cones in the hydrocyclone, as shown in Figure 4, which effectively reduces the fine particle content in the underflow. However, the iron in the RM is concentrated in the fine particles, which account for a large proportion of RM and have a low iron grade, resulting in low TFe of the concentrate obtained by gravity separation.



**Figure 4.** Particles in the parabolic cone [20].

Some scholars also use the magnetic separation process to separate the iron-containing part according to the magnetic difference. Jamieson et al. [21] combined low-intensity magnetic separation and wet high-intensity magnetic separation and obtained the concentrate with a  $\text{Fe}_2\text{O}_3$  mass fraction of 56%. In order to increase the TFe of the concentrate, Li et al. [22] adopted a two-stage magnetic separation process; each stage includes weak magnetic separation and vertical ring strong magnetic separation. The TFe and recovery rate of the concentrate were 24.63% and 43.29%, respectively. The reason for the lower TFe may lie in that the iron in RM mainly exists in the form of hematite and goethite, which are magnetic or weak magnetic materials, so a higher magnetic field is required. Li et al. [23] used high gradient superconducting magnetic separation to improve the TFe of the concentrate. The mass fraction of the  $\text{Fe}_2\text{O}_3$  was about 67%. The separation effect is not good and the cost is high. This may be due to the intergrowth of various elements in RM, including Al, Si, Na, Ti and Fe, such as aluminum in the form of high-alumina goethite and high-alumina hematite [21]. Magnetic separation cannot separate aluminum and iron, resulting in lower TFe in the concentrate.

Some scholars have used the selective functional groups to enrich the iron oxide in RM by the flotation process. Song et al. [24] studied the dealing effect of different collectors. When the methylenebisacrylamide ( $\text{C}_6\text{H}_{12}\text{N}_4$ ) and starch was mixed as the collector, the TFe and iron recovery of the concentrate were 49.81% and 74.64%, respectively. When the sodium oleate ( $\text{C}_{18}\text{H}_{33}\text{NaO}_2$ ) was used as the reverse collector, the TFe and iron recovery of the concentrate were 51.54% and 78.84%, respectively. Huang et al. [25] found that the humic substances have acidic functional groups and are more selective in interacting with iron oxides, the TFe and iron recovery and of the obtained concentrate were 61.12% and 86.25%, respectively.

Multiple physical methods were combined by some scholars. Rai et al. [26] obtained iron oxide and titanium oxide from RM by using the hydrocyclone due to their high density, and then, the iron is recovered by the two-stage magnetic separation. The mass fraction of  $\text{Fe}_2\text{O}_3$  in the concentrate was 70% and the iron recovery was 75%.

The physical recovery method has the advantages of simple process and low energy consumption, but the iron recovery and TFe of the concentrate are not high enough to meet the need of industrial production. Because of the similar physical properties of iron minerals and veinlets, it is difficult to separate them by the traditional physical methods. On the one hand, some scholars adopted more advanced techniques such as high-gradient superconducting magnetic separation to separate them, but the cost is high. In the future, a physical method will be used mainly in conjunction with chemical methods, and scholars should develop corresponding physical methods based on chemical methods with simple steps, low energy consumption and low cost.

#### 4.2. Chemical Recovery Method

The chemical recovery method refers to the process of converting iron in RM to other forms through chemical reactions and then extracting iron, which is divided into the hydrometallurgy method and pyrometallurgy method according to the type of chemical reaction.

##### 4.2.1. Hydrometallurgy Method

The hydrometallurgy method mainly refers to the process of dissolving iron by acid leaching and then enriching iron. Some scholars studied the influence of acid leaching conditions on the leaching rate. Xie et al. [27] leached iron with hydrochloric acid (HCl) and studied the effects of temperature, acid concentration, leaching time, liquid–solid ratio and particle size on the leaching rate. They found that temperature and acid concentration have the greatest effect, while leaching time and liquid–solid ratio have the second effect. Reducing particle size within a certain size range can increase the leaching rate, but when the particle size was less than 150  $\mu\text{m}$ , there was no significant effect on the leaching rate by reducing the particle size. The optimum conditions for the acid leaching were: red mud size 150  $\mu\text{m}$ , acid leaching temperature 80  $^{\circ}\text{C}$ , acid concentration 10 mol/L, liquid–solid ratio 8:1, and acid leaching time 150 min, and the leaching rates of Fe was 95.1%. In addition to HCl, Chen et al. [28] used concentrated sulfuric acid, glacial acetic acid and oxalic acid ( $\text{C}_2\text{H}_2\text{O}_4$ ) to leach RM and studied the effects of acid concentration, leaching time and temperature on iron leaching. They found that sulfuric acid leaching was more effective and that increasing the sulfuric acid concentration and temperature can increase the iron leaching rate. The leaching mechanism was consistent with the core shrinking model controlled by internal diffusion. The maximum Fe leaching rate of 67.93% was achieved at a temperature of 50  $^{\circ}\text{C}$ , a sulfuric acid concentration of 2.8 mol/L, and a leaching time of 45 min.

By exploring the complete hydrometallurgy process, Yu et al. [29] leached RM with 1 mol/L  $\text{C}_2\text{H}_2\text{O}_4$  at 75  $^{\circ}\text{C}$  for 2 h and then irradiated the solution with UV light for 1 h to reduce  $\text{Fe}^{3+}$  to  $\text{Fe}^{2+}$  by oxalate and formed a precipitate ( $\beta\text{-FeC}_2\text{O}_4 \cdot 2\text{H}_2\text{O}$ ) by combining with the remaining oxalate. Although the oxalate in the filtrate can be reused, the oxalate reacting with  $\text{Fe}^{3+}$  cannot be reused, which greatly increases the process cost. In order to reduce the amount of oxalate used in the process, Yang et al. [30] used dilute HCl for washing, sulfuric acid for acidity adjustment and 3 times stoichiometry of iron scraps instead of oxalate for the reduction in  $\text{Fe}^{3+}$  to reduce the use of oxalate to reduce cost. In addition, in order to reduce cost, Yang et al. [31] proposed a process shown in Figure 5, in which they leached RM with  $\text{C}_2\text{H}_2\text{O}_4$ , adjusted the pH with calcium carbonate to form a co-precipitation of calcium oxalate and iron hydroxide, and then used a mixture of hydrochloric acid and calcium chloride to selectively dissolve the precipitate and recover the oxalate and iron.

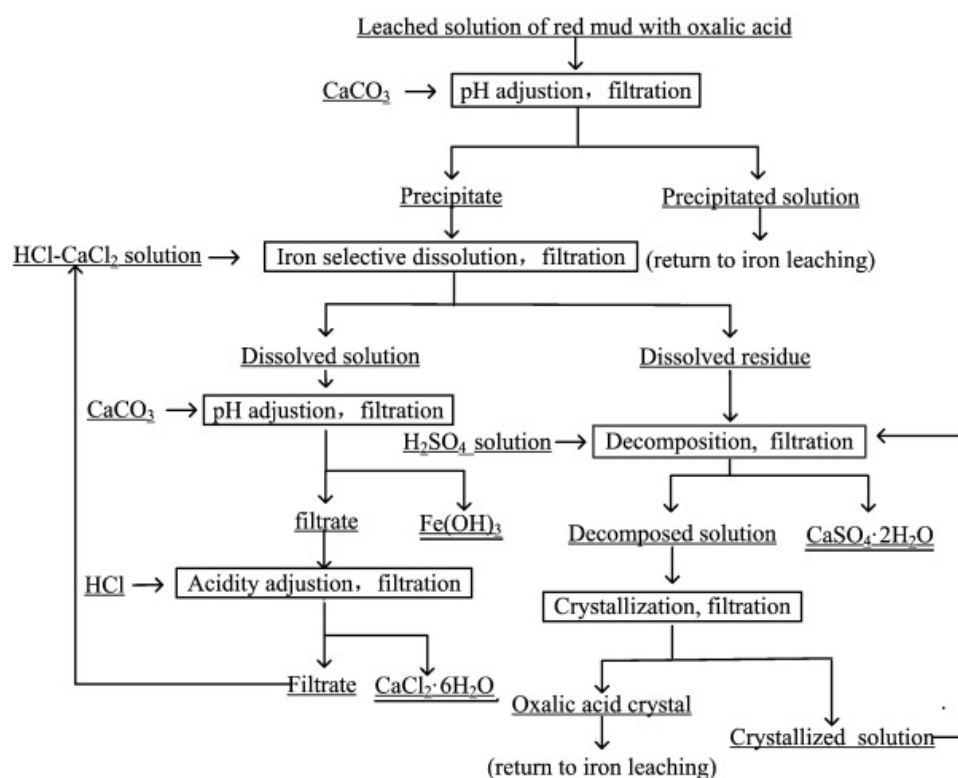


Figure 5. Hydrometallurgy process flow of Yang et al. [31].

In addition to  $C_2H_2O_4$ , Zhang et al. [32] leached the RM with hydrochloric acid for 4 h, added calcium chloride to form iron and scandium complexes and used D201 resin to adsorb iron complexes to achieve the separation of iron and scandium. The adsorption efficiency of iron exceeded 96%, while the adsorption of scandium was negligible. Calcium chloride could be recycled to reduce the cost. Zhang et al. [33] also used hydrochloric acid leaching and extracted iron with a mixture of 30% Aliquat 336, 15% 2-Octanol and 55% kerosene for 20 min. The iron leaching and extraction rates reached 95.9% and 95.7%, respectively. Sokolov et al. [34] also used a mixture of 1-octanol ( $C_8H_{18}O$ ) and 1-decanol ( $C_{10}H_{22}O$ ) (70%) with 2-undecanone ( $C_{11}H_{22}O$ ) (30%) to extract iron from HCl solution. They obtained an  $FeCl_3$  solution with the iron(III) content of 90.5 g/L and total impurities less than 50 mg/L.

Oxalate can form the precipitate by combining with  $Fe^{2+}$ , so  $C_2H_2O_4$  becomes the main raw material for hydrometallurgy method. Because of the high cost of  $C_2H_2O_4$ , many scholars have optimized the process. The hydrometallurgy method can achieve both a high recovery rate and TFe in laboratory-scale experiments. The chemical reaction can be carried out at a lower temperature, so hydrometallurgy also has the advantage of low energy consumption. However, the hydrometallurgy method has complex steps. Due to the strong alkalinity of the RM, a large amount of acid is consumed during acid leaching, which makes the cost significantly high, making it difficult to promote in the industry. The method will also produce an acidic waste stream, which contains many metallic elements. Future research on the hydrometallurgy method could focus on how to reduce acid consumption during leaching such as treating RM with acidic solid waste or using the alkaline leaching process to extract iron, and how to recover other metallic elements as well as raw materials from the acidic waste stream after iron extraction. Industrial tests on the process should also be carried out.

#### 4.2.2. Pyrometallurgical Method

The pyrometallurgy method is the process of changing the existing form of iron through a high-temperature chemical reaction and then separating. According to the form

of iron after reduction, the pyrometallurgy method can be classified into the reduction to Fe process and reduction to  $\text{Fe}_3\text{O}_4$  process.

Many scholars have studied the pyrometallurgical process of reduction to Fe. Mishra et al. [35] used 200% excess petroleum coke to reduce iron at 1050 °C for 2 h. After reduction, most of the iron was recovered by magnetic separation, and the remaining iron was recovered by smelting. Raspopov et al. [36] have obtained cast iron by reacting with carbon in a resistance furnace at 1200–1500 °C. There are many factors affecting the reduction reaction including temperature, reaction time, etc. In order to improve the physical characteristics of obtained pig iron, Valeev et al. [37] roasted RM at 1750 °C and found the titanium carbide, and phosphide eutectic in pig iron as shown in Figure 6 gives it a high hardness, high strength and good fluidity. In order to effectively find the optimal working conditions, Guo et al. [38] used metallization rate as an indicator, which means the percentage of metallic iron to total iron, and they designed orthogonal experiments to find the best working condition. Under the best working condition, the metallization rate was 98.16% and the TFe of concentrate was 96.25%. Lazou et al. [39] conducted smelting reduction experiments at 1650 °C for 90 min in an induction furnace as shown in Figure 7, and they obtained pig iron with TFe of 92.74%. The backscattered electron (BSE) images of slag are shown as Figure 8, and the chemical composition of these phases is given in combination with the XRD results. They also used FactSage to predict elemental concentrations in the metal phase and found that the predicted Si and Cr concentrations were much higher than experimental values. Ning et al. [40] added CaO and  $\text{Al}_2\text{O}_3$  to the reduced RM for composition adjustment, and they separated slag and iron at 1550 °C. The quality of pig iron obtained meets the requirements of steelmaking, and the slag-melting point is lower than 1400 °C with a strong desulfurization capacity.

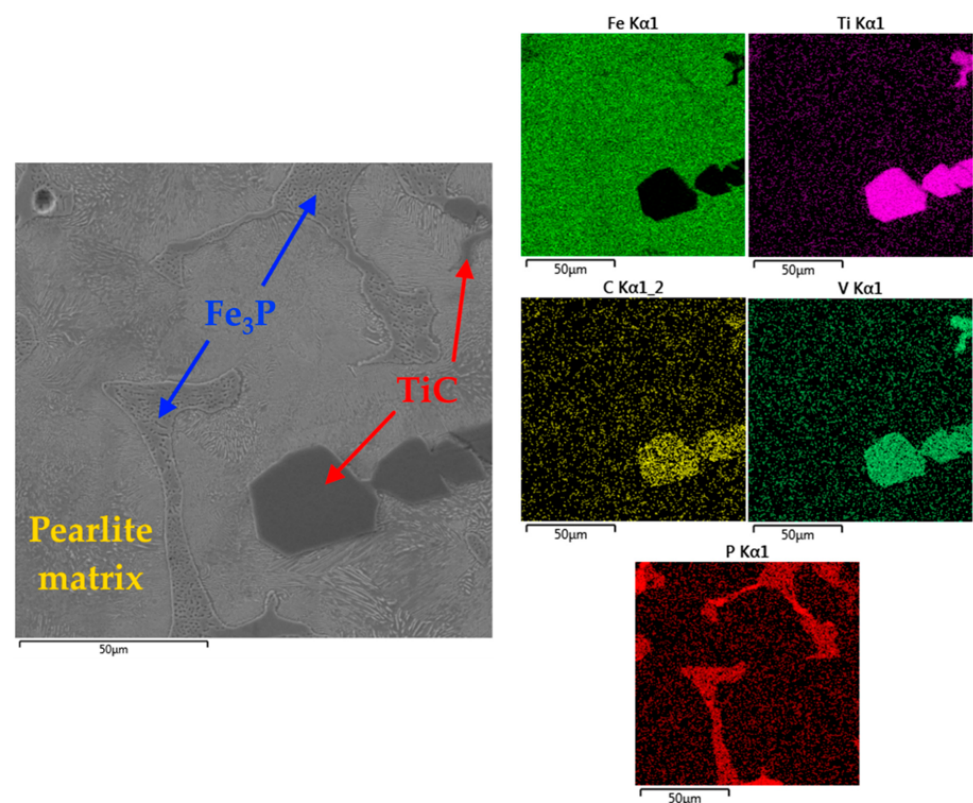
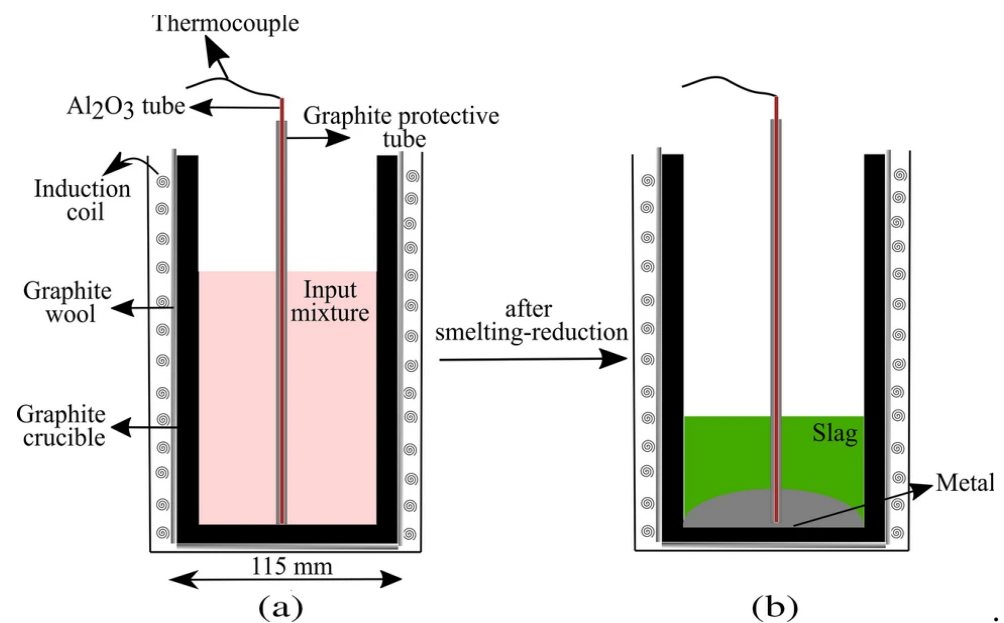
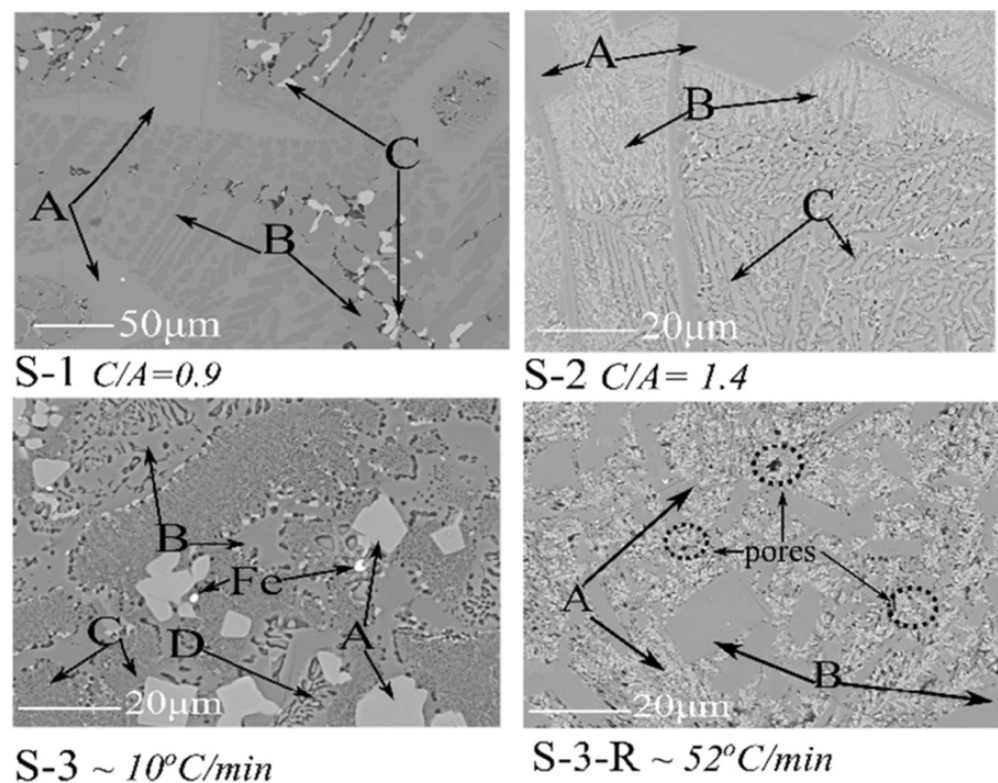


Figure 6. SEM images of the pig iron sample obtained at 1750 °C [37].



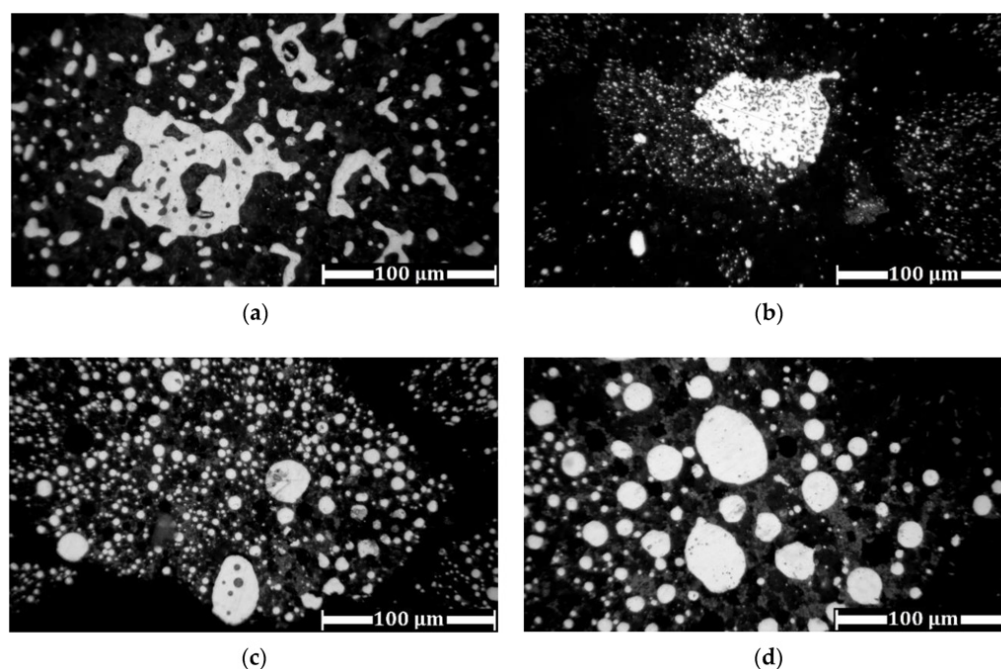
**Figure 7.** Schematic diagram of the smelting reduction process and input mixture (a) before and (b) after smelting [39].



**Figure 8.** BSE images of slags obtained with different  $\text{CaO}/\text{Al}_2\text{O}_3$  ratios (S-1 and S-2) and different cooling rates (S-3 and S-3-R) [39]. A, B, C, and D represent four different phases.

Some scholars have found that better results are achieved by adding alkaline additives to the roasting. Zinoveev et al. [41] investigated the effect of the addition of  $\text{Na}_2\text{CO}_3$  and  $\text{K}_2\text{CO}_3$  on the reaction. The experimental results showed that iron can be almost completely reduced after roasting at 1000–1200 °C for 20 min. As shown in Figure 9, they found that iron grains in the sample with additive reduction were significantly larger. Under the best working condition, the recovery rate and TFe were 77.27% and 72.05% with the addition of

$\text{Na}_2\text{CO}_3$  and 92.39% and 70.77% with the addition of  $\text{K}_2\text{CO}_3$ . Liu et al. [42] used carbon powder as a reducing agent and  $\text{CaCO}_3$  and  $\text{MgCO}_3$  as additives, which can react with  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$ . The TFe, metallization rate and iron recovery of obtained concentrate were 88.77%, 96.98% and 81.40%, respectively. They added 13% slaked lime to the remaining silica–aluminate residue. The compressive strength of residue reached 24.10 MPa, which can be used as construction material. Huang et al. [43] investigated the mechanism of the reduction reaction with different additives. The experimental results indicated that  $\text{Na}_2\text{CO}_3$  dissociates the basic oxide  $\text{Na}_2\text{O}$ , which can displace FeO from  $2\text{FeO}\cdot\text{SiO}_2$  and  $\text{FeO}\cdot\text{Al}_2\text{O}_3$ . The generated  $\text{Na}_2\text{O}\cdot 2\text{SiO}_2$  with a low melting point produces a liquid phase which can promote the diffusion of crystalline masses, accelerate the growth of Fe nuclei, as well as catalyze the carbon gasification reaction and increase the carbon activity and the rate of CO reduction of iron oxides. Adding  $\text{CaF}_2$  can lower the melting point and viscosity of solid-phase products, optimize the heat and mass transfer conditions, and promote the mutual diffusion between ions and the growth and enrichment of iron grains. Finally, the metallization rate, TFe and recovery rate of obtained iron powder were 92.79%, 89.57% and 91.15%, respectively. Liu et al. [9] believed that an excessive liquid phase can also encapsulate the particles, leading to an incomplete reduction of iron oxides and partial dissolution of ferrous compounds in the RM, and some metallic iron particles encapsulated by the liquid phase cannot be recovered by magnetic separation, so the amount of the additive should be strictly controlled.



**Figure 9.** Microstructure of reduced samples after 3 h of roasting at 1200 °C with 17.1%  $\text{Na}_2\text{CO}_3$  (a) and 22.01%  $\text{K}_2\text{CO}_3$  (b), without additions at 1300 °C (c) and 1350 °C (d) [41].

Other heating methods of the process were also developed. Jayasankar et al. [44] applied thermal plasma technology in the process. The characteristics of high temperature, high energy fluxes, and plasma state of the plasma arc greatly reduce the reaction time. Agrawal et al. [45] demonstrated that microwave heating allows for rapid and uniform heating of the sample. They experimentally compared the carbothermal reduction reactions using muffle furnace and microwave heating, and they found that microwave heating significantly improves the TFe and recovery rate of iron at a lower time and with less reducing agent. They also observed ferrite spheres of appreciable size and purity in microwave-heated reduced samples, accounting for 8–10% of the total iron in the feed. In

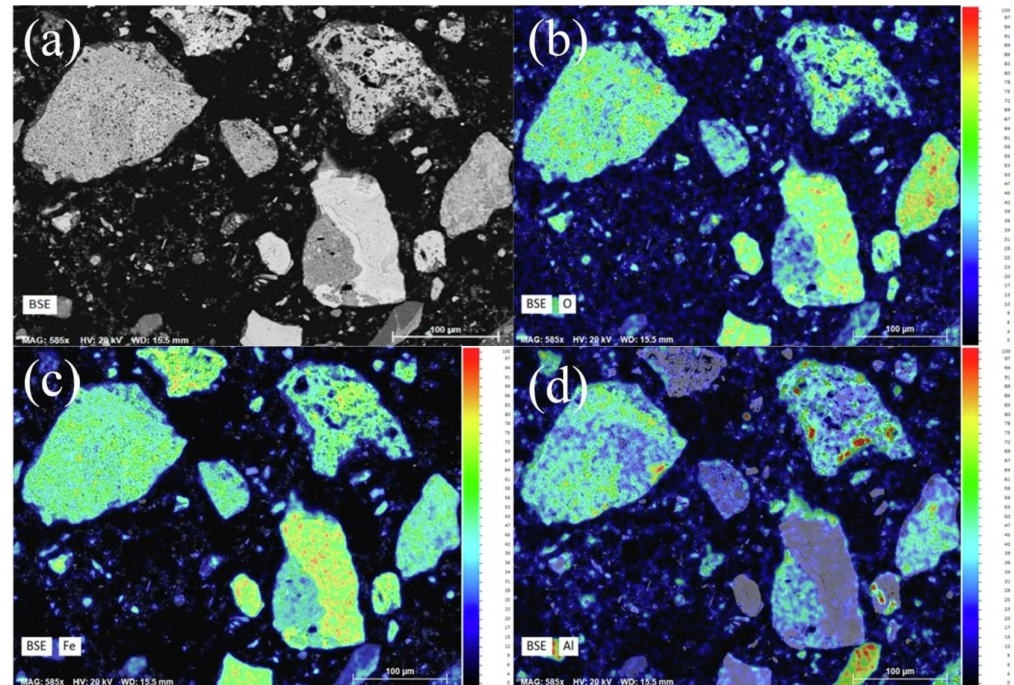
addition, microwave heating provides faster reduction, cleaner process, and less energy and reductant consumption.

High-temperature reduction and melt separation make the process energy consumption too high; scholars found that the iron in the RM can be reduced to  $\text{Fe}_3\text{O}_4$  at a lower temperature, and magnetic separation will greatly reduce the energy required for the process. Liu et al. [46] used coal coke as the reducing agent and soda lime as the additive, roasted RM at 1000 °C for 3 h, and followed that with water leaching to extract Al and Na and magnetic separation to extract Fe. The recovery rate and the TFe of the concentrate were 51.2% and 52.8%, respectively. Sadangi et al. [47] roasted RM at a temperature of 1150 °C for 60 min and obtained the concentrate with the TFe of 65.93% and the recovery rate of 61.85% by magnetic separation. They also demonstrated that the reduction reaction occurred from the outer surface of the particles to the inner core. Agrawal et al. [48] proposed the process of carbonation, pre-magnetic separation, reduction roasting and two-stage magnetic separation, and they chose charcoal as the reducing agent. The obtained concentrate had the TFe of 50.5% and the recovery rate of 70%.

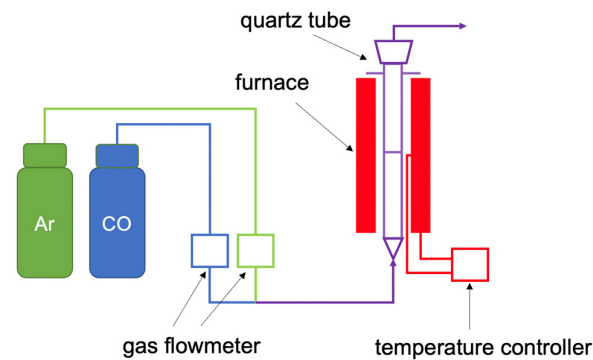
When a solid reductant is used, the reaction temperature is still high, and the reaction needs to take a long time. If the gas reductant is used, the reaction temperature and time can be greatly reduced. Samouhos et al. [49] passed  $\text{H}_2$  through the fixed bed reactor at 480 °C for 30 min and obtained the concentrate with the TFe of 54% after magnetic separation. They performed a Vibrating Sample Magnetometer (VSM) test on the concentrate and found that greater magnetism causes better enrichment. Tang et al. [50] pointed out that fluidized roasting in traditional pyrometallurgy has the advantages of fast reaction speed, rapid mass transfer, low energy consumption and uniform temperature. If the reaction is carried out in a fluidized bed with a gas reductant, the reaction temperature and reaction time can be further reduced. Some scholars applied fluidized roasting to the iron recovery process from RM; Liu et al. [51] chose CO as a reducing agent, reduced the RM in a fluidized bed at 540 °C for 15 min and obtained the iron concentrate with 56.41% TFe after magnetic separation. They also found that the magnetic force of the reduced RM was positively correlated with the dealing effect in the experiment. Yuan et al. [52] passed a mixture of  $\text{H}_2$  and CO in the fluidized bed reactor to reduce RM at 520 °C, and this semi-industrial experiment was conducted continuously and steadily for 45 h. The recovery rate and TFe of the concentrate were 95.22% and 55.54%, respectively. Yu et al. [53] roasted the RM with a concentration of 30% CO in the fluidized bed at 560 °C for 15 min; after magnetic separation, the recovery rate and TFe of the obtained concentrate were 89.34% and 55.44%, respectively. According to the BSE image shown in Figure 10, they found that some fine silicon slag was embedded in the magnetite. In the magnetic separation process, the Fe-Al hercynite ( $\text{Fe}(\text{Al},\text{Fe})_2\text{O}_4$ ) transformed from aluminogothite would mix into the concentrate, leading to a decrease in the TFe of the concentrate. Li et al. [54] found that in the conventional pyrometallurgy process, the iron-bearing minerals in the fluidized roasted ore are closely embedded with the veinlets, reducing the TFe of the concentrate obtained by magnetic separation. This problem also occurs and is urgent to be solved in the fluidized reduction process of the RM.

In previous work, Kong et al. [13] conducted the experiment at the test rig shown in Figure 11, adjusted the gas velocity to enter the turbulent bed and observed a significant discoloration of the RM due to reduction within 5 min. The maximum moment in the VSM test of the reduced RM was used as a characterization of the dealing effect, and the effects of CO concentration, reaction temperature and reaction time were studied by using response surface analysis. The maximum moments of reduced RM under different working conditions are listed in Table 2. The variance distribution of RSM is listed in Table 3. The *p* value represents the strength of significance and the larger mean square value represents the greater the influence on the dealing effect. From Table 3, it can be found that the *p*-value of the model is 0.0032, and the underfitting phase is not significant, indicating that the model is reasonable. The order of influence of these three factors on the dealing effect is CO concentration, reaction time, and temperature. The response surface method (RSM) also

predicts the results accurately at the condition of longer reaction time as listed in Table 4. In the future, more attention will be paid on expanding the experimental condition design by evaluating the dealing effect by the TFe and recovery rate.



**Figure 10.** BSE image of roasted product and its map scanning: (a) BSE Image; (b) O; (c) Fe; (d) Al [53].



**Figure 11.** Schematic diagram of the experiment system [13].

**Table 2.** The maximum moments of reduced RM under different working conditions [13].

| Temperature (°C) | CO Concentration (%) | Reaction Time (min) | Maximum Moment ( $A \cdot m^2/kg$ ) |
|------------------|----------------------|---------------------|-------------------------------------|
| 600              | 5                    | 8                   | 15.7401                             |
| 550              | 8                    | 2                   | 15.4587                             |
| 600              | 2                    | 5                   | 15.1976                             |
| 500              | 5                    | 2                   | 13.9256                             |
| 550              | 2                    | 2                   | 9.6766                              |
| 600              | 8                    | 5                   | 15.2625                             |
| 550              | 5                    | 5                   | 15.2115                             |
| 550              | 5                    | 5                   | 15.2115                             |
| 500              | 5                    | 8                   | 15.3054                             |
| 500              | 2                    | 5                   | 10.2996                             |
| 550              | 5                    | 5                   | 15.2115                             |
| 600              | 5                    | 2                   | 14.1055                             |
| 550              | 5                    | 5                   | 15.2115                             |
| 550              | 2                    | 8                   | 14.6415                             |
| 550              | 5                    | 5                   | 15.2115                             |
| 500              | 8                    | 5                   | 14.7424                             |
| 550              | 8                    | 8                   | 16.6117                             |

**Table 3.** The variance distribution of RSM [13].

| Source           | Sum of Squares | df | Mean Square | F-Value | p-Value |
|------------------|----------------|----|-------------|---------|---------|
| Model            | 33.76          | 3  | 11.25       | 7.76    | 0.0032  |
| Temperature      | 4.55           | 1  | 4.55        | 3.14    | 0.1     |
| CO concentration | 18.79          | 1  | 18.79       | 12.95   | 0.0032  |
| Reaction time    | 10.42          | 1  | 10.42       | 7.19    | 0.0189  |
| Lack of fit      | 18.86          | 9  | 2.1         |         |         |
| Pure error       | 0              | 4  | 0           |         |         |

**Table 4.** Predicted moment and experimental moment [13].

| Temperature (°C) | CO Concentration (%) | Reaction Time (min) | Experimental Moment (A·m <sup>2</sup> /kg) | Predicted Moment (A·m <sup>2</sup> /kg) |
|------------------|----------------------|---------------------|--------------------------------------------|-----------------------------------------|
| 525              | 3.5                  | 3.5                 | 11.4235                                    | 12.8168                                 |
| 525              | 6.5                  | 6.5                 | 15.2387                                    | 15.4908                                 |
| 575              | 3.5                  | 6.5                 | 14.8505                                    | 14.7124                                 |
| 575              | 6.5                  | 3.5                 | 14.4225                                    | 15.1034                                 |

In the early studies, scholars mostly used the high-temperature roasting process to reduce Fe<sub>2</sub>O<sub>3</sub> of RM to Fe and the smelting process to separate them. However, the high-temperature roasting process requires a temperature of more than 1200 °C and a time of more than 2 h, while the smelting process requires more than 1300 °C, which will result in a large amount of energy consumption. Some scholars improved the process based on the high-temperature roasting process, using a lower temperature of roasting to reduce Fe<sub>2</sub>O<sub>3</sub> to Fe<sub>3</sub>O<sub>4</sub> and using magnetic separation. The temperature required for roasting is 1000 °C, and magnetic separation can be carried out at room temperature, reducing the energy required for the process. Some scholars also found that the use of a fluidized reduction process can reduce the reaction temperature to 500 °C, and the reaction time is reduced to 30 min. It has also been found that the reaction time can be further reduced if the gas velocity is increased to reduce the RM in a turbulent bed.

The fluidization reduction process has a lower reaction temperature and shorter reaction time than other pyrometallurgical processes, which greatly reduces the energy consumption of the reaction, and simpler steps, which makes it easier to be promoted in the industrial applications. So, the fluidization reduction is a promising process. In future research, the iron grade and recovery rate of the concentrate should be improved. For the secondary solid waste generated after the recovery of iron, corresponding processes should also be developed to utilize or extract the elements.

#### 4.3. Emerging Recovery Method

In addition to the above methods, the bioleaching method, which is considered as a green technology with low energy and cost, was also provided. Some scholars have studied the recovery of iron from kaolin [55], quartz sands [56], bauxite [57] and iron oxides ores [58]. Qu et al. [59] used *Aspergillus niger* to leach heavy metals from RM. When bioleaching was carried out at 1% pulp density, the leaching rate of iron reached a maximum 87.5%, which is significantly lower than hydrometallurgy method. Other metals will enter the leaching solution in large quantities. Eisele and Gabby [60] pointed out that anaerobic organisms are widely used in bioleaching iron processes, but anaerobic organisms require neutral or weakly acidic pH conditions, which leads to a large amount of acid neutralization during RM leaching, limiting the development of RM bioleaching. In the future, it is necessary to find microorganisms that can adapt to alkaline environments and selectively leach iron in the bioleaching process and implement larger scale experiments to verify the feasibility of the process.

In addition to bioleaching, Maihatchi Ahamed et al. [61] used the electrochemical method to recover iron, which can reduce energy consumption and CO<sub>2</sub> emissions compared to the pyrometallurgical method. The alkalinity of RM can reduce the amount of alkali added to the media. They found that the efficiency of the electrochemical method is low, which is probably due to the lower conductivity caused by the formation of triple-phase bubbles with the suspension. They also found that there was 3% Al and Na in the obtained concentrate and that the current yield of the RM was about a quarter of the current yield of the hematite at high current densities. Further research should be carried out to increase the current yield, improve the conductivity of the suspension and reduce impurities in the concentrate.

In the long term, the emerging method will become the mainstream method in the future due to its low energy consumption and environmental friendliness, but it will not be able to treat the urgent RM in a short time. The emerging method still needs a lot of work, such as improving the recovery rate and iron grade, larger scale experiments, reducing process costs, developing better processes, etc.

## 5. Conclusions

This paper reviewed three kinds of methods on recovering iron from RM, including physical, chemical and emerging methods. The following conclusions are obtained:

1. The physical method is to separate the iron-bearing part and the other parts based on different physical properties, including gravity separation, magnetic separation and flotation. These processes have the characteristics of simple process and low energy consumption, but the iron recovery rate and the TFe of the concentrate are low;
2. The hydrometallurgy method is the chemical method with acid leaching. The RM leached with acid can dissolve the metallic elements, and then the iron in the solution is extracted by precipitation formation, extraction or adsorption. The hydrometallurgy method has a high recovery rate and TFe of the concentrate, but the steps are complicated and require a large amount of acid to neutralize the alkalinity of the RM, resulting in high costs;
3. The pyrometallurgy method is the chemical method in which iron is reduced by thermochemical reaction and then separated by smelting or magnetic separation. The pyrometallurgy method can be divided into a reduction process into Fe and Fe<sub>3</sub>O<sub>4</sub> according to the form of iron after reduction. The reduction process into Fe requires high temperature, high energy consumption and long reaction time, but the TFe of the concentrate is high. Comparingly, the reduction process into Fe<sub>3</sub>O<sub>4</sub> in a fluidized bed will reduce the reaction temperature and reaction time, and the reduction in the turbulent bed will further reduce the reaction temperature and reaction time. However, the concentrate has a lower TFe, which should be the focus in future research. The fluidization reduction process is currently the most promising process and should be promoted on an industrial scale
4. The emerging method is a green and clean technology, but the current research is in the laboratory scale. In the future, process efficiency improvements, costing, and larger scale experiments are required. In the long term, the emerging method is a very promising process but requires a lot of research.

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