The neutralization and recycling of red mud – a review

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Abstract. Red mud (bauxite residue) is a highly alkaline solid waste of alumina production. The amount of red mud is huge while its alkalinity hinders the application in construction industries. Up to now, the most common treatment of red mud is disposed in man-made dams. However, the long-term storage of large-amount red mud pollutes environment and underground water, threatens human life and leads to a huge waste of valuable metals resources. Recently, researchers have proposed various strategies to solve the red mud problems. In this review, the neutralization and recycling status of red mud have been systematically illustrated, their advantages and disadvantages are also discussed in detail in terms of environmental and economic benefits. Moreover, the recommendation of red mud treatment in the future is suggested, the application in road base material is highly valued because of the huge consumption.

1. Introduction

Aluminum is an abundant metal element in the earth’s crust (accounts for 8.3% of total elements). As a kind of easy processing and recycling light metal, aluminum is widely used in chemical industries, and is of great importance for the development of automobile, aviation, construction and other important industries. The industrial production of aluminum mainly consists of two steps, the production of alumina and alumina electrolysis. Alumina production process mainly includes acid process, electric heating process and alkaline process, among which the alkaline process is the mainstream process of alumina production. Depending on different processing methods, the alkaline process can be further divided into Bayer process, sintering process and Bayer-sintering process, among which the Bayer process is adopted by most factories. In recent years, the global alumina production is rising continuously owing to the rapid economic development, especially China, whose alumina production accounts for more than 50% of global alumina production. However, the development of alumina industry also leads to a series of environmental and safety problems, especially red mud, the solid waste generated in the alumina production process, has brought serious environmental pollution to the soil and underground water. Moreover, the existence of red mud also threatens the safety of human.

Red mud is so named because of its red color owing to the high iron oxide content, and its similar appearance with soil. Generally, depending on the alumina production process and the bauxite grade used, there will be 0.5-2.0 tons of red mud produced for each ton of alumina. In 2020, the global stock of red mud has exceeded 5 billion tons, of which China’s red mud stockpile exceeds 800 million tons.

The chemical composition of red mud is relatively complex, the physical and chemical properties vary from each other depending on the mining areas and production process. However, there are some common characteristics of red mud from different factories, such as small particle size, complex phase composition and high alkalinity. For example, Yu and Samal et al. found that the particle sizes of more than 40% red mud are less than 10 um, which makes it difficult to stay on the ground and very easy to
cause serious dust. The main chemical components of red mud generally include CaO, SiO$_2$, Al$_2$O$_3$, Fe$_2$O$_3$, TiO$_2$, Na$_2$O, K$_2$O, etc., it consists of many different types of minerals, such as aragonite and calcite etc. The red mud contains a certain percentage of soluble alkali and insoluble alkali, although the alkali content varies, it is basically between 2%-6%, but the percentage can exceed 10% in some specific factories. The existence of alkali makes the red mud appear strongly alkalinity, the pH is between 9.0-13.2 with the average value around 11.3±1.0. The soluble alkali seeps from red mud, pollutes the soil and the groundwater, brings serious environmental and safety problems. Also, the strong alkalinity of red mud causes strong corrosion to organisms, siliceous materials and metals, severely restricting its recycling. In addition, due to the incomplete reaction of bauxite during the dissolution process and the silicon impurities contained in the system, a large amount of aluminum and sodium contained components will be generated during the production process, which is a waste of valuable metal sources.

Due to the special characteristics of small particle size, high alkalinity, complex composition and large annual output, the recovery of valuable metals and recycling of red mud has many limitations. So far, most red mud is stored in man-made dams. However, this kind of treatment cannot solve the environmental and safety problems caused by red mud thoroughly. On one hand, the accumulation of red mud not only causes a series of pollution to the nearby soil and groundwater resources, but also harms the local economic development, ecological environment and the safety of human living environment. On the other hand, the use of red mud dams will not only occupy a lot of land resources, but also the fund required for dam maintenance and the risk of dam failure continue to increase with the increasing amount of red mud (the management and storage cost per ton of red mud is around 50-100 yuan). In 2010, 1 million cubic meters of red mud leaked from the Ajkai alumina red mud storage dam in southwest Hungary. In 2014, the pipe leakage of the dam (Chinalco Henan Branch) caused partial collapse and seriously polluted the surrounding environment and groundwater; in 2016, a red mud landslide occurred in the red mud dam of Henan Xiangjiang Wanji Aluminum Company, which caused the bury of a large number of houses and other properties in a downstream village, leading to huge property losses.

Researchers have been investigating the harmlessness of red mud in many different ways, mainly include the neutralization of red mud, the recovery of valuable metals, the application of red mud for building materials and other functional materials. In this review, we systematically introduced the current status of harmless treatment and recycling of red mud so far. The advantages and disadvantages of all treatment methods are discussed in terms of the simplicity of treatment process, the processing cost and the economic benefit etc. This review can provide a good reference for harmless treatment and recycling of red mud.
Figure 1 The schematic of red mud neutralization and recycling

2. Neutralization of red mud

2.1. Acid neutralization

The acid neutralization is a very simple method that based on the principle of acid-base neutralization, the general idea of acid neutralization is to take use of the waste acid/acid gases to neutralize red mud. Zeng et al. investigated the dealkalization performance by using different acids that of same concentration, they found that the sulfuric acid showed the best dealkalization rate of 91.06%. Li et al. also used oxalic acid to neutralize red mud, and the result showed that dealkalization rate is higher than 95% when the amount of oxalic acid is 15%, the temperature is 80℃, the temperature is 40 min and the liquid/solid ratio (L/S) equals to 4 mL/g. Moreover, it is demonstrated that the oxalic acid can effectively dissolve the cancrinite and calcite in the red mud, which is helpful for the recycling of metal elements. The main advantage of using acids to neutralize red mud is the high efficiency, also, taking use of waste acids makes the cost low enough to be accepted by industry. However, on the other hand, it should be noted that the consumption of acids is huge, while the amount of waste acid is far from enough; in addition, the factories that produce waste acids maybe far away from the factories that produce red mud, and it costs a lot to transport the acids or red mud in that case, let alone the high requirements of transporting acids.

In addition to acids, acid gases, including CO2, SO2 et al., also have been investigated to neutralize red mud. Li et al. used CO2 to neutralize red mud, the dealkalization rate was 49.3% with the L/S of 7:1, the temperature of 50 ℃ and the pressure of CO2 of 4MPa. Tang et al. proposed the idea of using lime kiln exhaust gas to neutralize red mud, the principle is to neutralize red mud with the large amount of acid gases. The dealkalization rate was 31.10% with the L/S of 8:1, the temperature of 60 ℃, the volume ratio of CO2 of 20% and the rotating speed of 400 r/min.

Overall, using waste acids and acid gases to neutralize red mud itself is cost efficient, in terms of efficiency, it is much higher using acids compared that with using acid gases. Principally, the neutralization process using waste acids/acid gases can achieve the purpose of “using waste to cure waste”, while the application of this method is strongly dependent on the location, the cost would be dramatically high if long-distance transportation is needed. Besides, a large amount of water will be needed considering the high L/S in the literature, and the neutralization process will produce a large
amount of dilute salt solution that would cause secondary-pollution to the environment and remaining to be dealt with, meaning that the method can not completely solve the red mud problem but causes new environmental problems.

2.2. CaO neutralization

CaO neutralization is an effective hydrothermal method that has been widely investigated, the principle is to replace Na+ with Ca2+ and react into more stable and less soluble cancrinite. Yang et al. investigated the dealkalization with CaO at atmosphere and found the alkali dissolution increases with the increase of temperature, 7 L/S ratio, lime ratio and reaction time. Shu et al. studied the dealkalization of red mud by using lime under atmosphere, they demonstrated that the order of effect significance factors are the ratio of added lime, the reaction time, the temperature and the L/S. The N/S of red mud was lower than 0.2 under optimized experimental condition. Zheng et al. obtained the dealkalization rate of 80% under the temperature of 70~90 ℃ and L/S of 3.9 The CaO is a relative cheap and widely distributed material all around the world, which brings great convenience to the CaO neutralization process. However, the dealkalization efficiency under room temperature and atmosphere is not high enough and requires harsher experimental conditions, which implies higher cost. In addition, this process generally uses a large amount of water and the dilute solution that contains Na+ is impossible to be fully recycled in industrial process, which causes water waste and leads to secondary pollution to the environment.

![Figure 2: General flowsheet of red mud treatment with CaO](image)

2.3. Seawater neutralization

Seawater neutralization is the simplest neutralization method which is widely used in offshore factories. The Ca2+ and Mg2+ in seawater can replace the Na+ in the red mud and reduce the alkalinity. The seawater neutralization method does not diminish the hydroxide from the red mud, instead, it converts the soluble waste into less soluble solids. Hanahan et al. investigated the geochemical results of red mud neutralization by seawater. They proved that the neutralization can greatly reduce the pH of red mud, and it is easier to handle the processed residue due to the agglomerate consolidation. Besides, the seawater neutralization is also helpful for the acid neutralization because the formation of aluminium hydrotalcite-like compounds needs to take carbonate from seawater. In contrast, Menzies et al. proposed that the residue sand needs a lot of water before revegetation because of the high salinity. They
demonstrated that the in-suit neutralization technique may not be a viable technique for routine revegetation, but within a dedicated structure may be promising. Rai et al. studied the seawater neutralization method using Taguchi’s design, they tested the experimental parameters such as the weight of red mud, the experimental time, the temperature and the seawater volume etc. By detecting the pH changes before and after the experiments, they showed the parameter contribution of red mud and seawater, respectively, and found that the pH was rarely influenced by experimental time and temperature.

Although with the advantages of high efficiency and low cost, the seawater neutralization method is not a universally applicable method due to the location limitation, but it is the most appliable treatment method for the seashore plants.

2.4. Biological neutralization

Biological neutralization, including the bacterial neutralization and revegetation-driven amelioration is highly valued owing to the environmental-friendly character. Bacterial neutralization, namely adding bacterial cells which can actively grew in the red mud and form organic acids to neutralize red mud eventually. The revegetation-driven amelioration generally takes use of alkali tolerant plants and microorganisms to neutralize red mud. Hamdy et al. used more than 150 bacterial cultures to neutralize the red mud and the result showed that the Lactobacillus, Leuconostoc and Bacillus were predominant species and played the critical role in acid production. As a result, the pH of the red mud was lowered from 13 to 7, the treated red mud supported the growth of plants survived for more than 300 days, while the plants survived for only 6-24 h in untreated red mud. M. S. Reddy et al. identified several species that can tolerate high alkalinity environment and produce organic acids by 16S rDNA sequence analyses. In another study, they investigated the influence of Aspergillus tubingensis to vegetation establishment. The result indicated that the addition of A. tubingensis, gypsum and sludge has positive influence on the growth of bermudagrass and showed the potential to help with the establishment of vegetation and neutralization. Yang et al. investigated the performance of Chloris virgata Swartz under alkali environment and found the relationship among relative growth rate, photosynthetic activity and pH.

In a word, the biological neutralization of red mud shows very promising results and the methods are environmental-friendly. However, this kind of method generally needs a lot of time to achieve the desired pH, which is hard to match the production speed of red mud. Moreover, the dissolution mechanisms and ion exchange processes still remain unknown, and there is no assess standard the survivability of microbial inoculants in the long term, which needs more further investigation to get deep understandings.

3. Recovery and recycling of red mud

3.1. Recovery of valuable elements

There are some valuable elements in the red mud, including Al, Na, Fe, Ti and rare earth elements, and researchers have been investigating how to recover the valuable elements and improve the economic value of red mud. It should be noted that the extraction and recovery of elements is generally a synchronous process, meaning that several elements can be extracted at the same time.

Zhong et al. extracted alumina and sodium oxide from red mud through a hydro-chemical process and produced sodium aluminate hydrate, the Na₂O in the final residue was less than 1%, which is environmental-friendly and could be used as construction materials. Kinnarinen et al. investigated the dissolution behavior of Na, Al and caustic compounds from red mud and found that the dissolution of sodium from the solids was independent on the grinding of particles, while it was strongly dependant on the L/S ratio. Agrawal et al. studied the extraction of Fe from carbonated red mud by magnetic separation and muffle furnace roasting, they got a 50.5% iron-rich concentrate and recovered 70% of the total iron from red mud. Zhu et al. recovered aluminum, iron and other metal elements from red mud with a series of process. As a result, they recovered 96% of scandium and vanadium by acid leaching.
and high grade TiO₂ was obtained. Although this process was very efficient in extracting elements, the process is quite complicated and consumes a lot of acid alkali, which is not good for the environment and leads to high cost.

Although the recovery of valuable elements has been widely investigated, the content of these elements can not always satisfy the requirements of industrial production. For example, the Fe content in red mud depends on the origin area of bauxite, some red mud contains a large percentage of Fe while some does not. Therefore, the extraction of valuable elements should be considered thoroughly, such as the cost, the environment and the safety issues.

3.2. Synthesis of catalyst with red mud

Red mud is a fine material with small particle sizes and owes high surface area and sintering resistance. Due to the very special properties of red mud, it has been employed as catalyst or catalyst support in several kinds of reactions, like exhaust gas cleaning and hydrogenation etc.22 Cao et al. investigated the application of modified red mud in the ammonia decomposition by supporting the Ni catalysts.22,23 Firstly, they modified the surface and crystal structure by acid digestion and alkali reprecipitation, and the NH₃ conversion efficiency of 97.9% was reached at 700 °C by combing with Ni catalyst, and the efficiency is comparable to the Ni-based catalysts systems that reported before. It was demonstrated that the high surface area and narrow pore size distribution were helpful for improving the catalytic activity and hydrogen production. Cao Wei et al. used the red mud as main material and prepared a Fenton-like catalyst by acidification and calcination.24 The molasses wastewater was used to dealkalize the red mud and the as-prepared catalyst was nearly neutral. The excellent stability and settleability of the catalyst have been proved by recycling the catalyst for several times, which implied the application of red mud in Fenton-like oxidation in wastewater treatment. Hu et al. investigated the catalytic oxidation of CO with the red mud supported CoO₄ catalysts.25 Similar with the NH₃ conversion process, the red mud was modified and used as catalyst support. The as-prepared catalyst was proved to has high oxygen content, high surface area and high porosity, the Co₃O₄ nanoparticles were highly dispersed and these features enabled the high catalytic reactivity and excellent stability.

In a word, the catalyst preparation processes are generally low cost and has promising catalytic reactivities, the application of red mud in catalyst industry can help with the recycle of red mud. However, the large amount of red mud produced cannot be fully digested by catalyst industry, the acid digestion and alkali reprecipitation also causes new environmental issues.

3.3. Fabrication of building materials with red mud

The red mud has outstanding hydraulic activity because of the large amount of silica, calcium oxide and aluminosilicate etc. Therefore, researchers have conducted many studies to use red mud as construction raw materials, including the preparation of cement, permeable bricks, glass-ceramics, ceramic foam (insulation materials) and road base materials etc.26

Subgrade soils is used as the base for the pavement which swells and shrinks when get contact with water. Generally, strong crushed rocks or soil stabilizers are used to fix this problem while the cost are ineffective. Sridevi et al. used the mixture of red mud, fly ash and lime as the stabilizers and investigated the influence of factors such as the mixture ratios and curing time on the stabilization properties.26 The result showed that the hydration product forms with time and increases the unconfined compressive strength. In addition to subgrade materials for pavement, red mud also has been used as the road subgrade material, the specific gravity, maximum dry density and liquid limit and other properties have been tested systematically to see whether it can satisfy the government standards. The red mud is also stabilized by hydrated lime and gypsum to reduce the plastic index. Liu et al. conducted studies on the application of red mud on base course construction,27 the red mud was mixed with cement and the optimized mixture totally satisfies the Chineses standards for road base materials, Indian standards and US Bureau of transportation requirements.

Pan et al. prepared a novel kind of cementitious material with red mud and Portland cements,28 the hydration process of hardened cement paste samples was stopped by alcohol drenching, the properties
of as-prepared samples are tested and compared with commercial cement. The results showed that the hardened cement has high strength without strength rebounding with curing ages, it also has outstanding chemical and freezing resistance, as well as more compact and integrated microstructures compared with commercial cement. Hou et al. prepared ceramic foams with red mud, fly ash and foaming agents. They investigated the influence of foaming agent on the crystal phase and structure. It was demonstrated that the main phases in the foams are different when adding different foaming agents, the morphology and physical properties of the foams are mainly dominated by foaming agent contents.

On the whole, it will undoubtedly be a very promising recycling method of red mud if it is actually used in the construction industry because the large consumption in this area. Also, the synergistic results of red mud and other solid wastes also improves the properties and the economic and environmental benefits. However, the treatment standards for red mud varies because of the different standards for construction materials in different countries, so the amount of red mud that can be added needs to be determined according to the practical situation. It should be noted that alkali in red mud will lead to “salinization” if being directly used in construction materials. Thus, the fixation of alkali in red mud requires in-depth and systematic research. In addition, the influence of heavy metals contained in red mud on personal safety in the long run should be carefully and thoroughly studied before practical application in the building materials.

3.4. Wastewater treatment
Red mud has been widely investigated for wastewater treatment especially in heavy metal adsorption. For example, the acid-treated and centrifuged red mud was used as for coagulant production. The addition of waste base in red mud causes the flocculation and sorption by surface enlargement. It was found the treated red mud has very stable mineral phase. Ma et al. used the red mud to treat heavy metal contaminated water and soil and proved the red mud has good adsorption abilities. The final goal is to control waste by waste and achieve zero pollution.

4. Conclusion and prospects
In conclusion, the methods about neutralization, recovery and recycling of red mud are described and discussed systematically in this review. In terms of the neutralization of red mud, the seawater neutralization is the most cost-effective method while its application has area limitation, and the long-term influence on the marine ecosystem still needs to be investigated. The neutralization by acid/acid waste/acid gas waste is not likely to be applied in large-scale because of the huge production of red mud, the area limitation, the cost and the secondary pollution caused by salts. The CaO neutralization is an effective method that can be applied in different areas because CaO is widely distributed and easy to get, but the conditions required for the process to obtain desired efficiency is harsh and improves the cost. On the other hand, the recovery and recycling of red mud is highly valued not only because of the environmental-harmless of red mud but also because of the potential economic value that it can produce. The recovery of valuable elements from red mud has been widely investigated but there is no quantitative calculation of cost and benefit, respectively. Besides, the chemical positions and properties of red mud from different regions vary, so the applicability should be considered. The application of red mud for building material can consume a large quantity of red mud, but the researchers should make sure it will not lead to “salinization”.

There are some factors that should be considered carefully during the processing of red mud: 1) the processing approaches should be based on the huge production of red mud and the cost should be considered. 2) the properties of red mud from different areas varies, implying that the selection of approaches should be combined with the practical condition of local area. 3) the environment influence should be considered and the secondary pollution should be avoided. Taking all factors into consideration, the utilization of red mud in construction industry is a promising method due to the low cost and huge consumption.
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