Sustainable utilization of bauxite residue (Red Mud) as a road material in pavements: A critical review

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Abstract: Sustainable utilization of the bauxite residue (red mud) generated from alumina refining has recently increased due to increased environmental concerns because of its high alkalinity and problematic pollutants when placed in landfills. This paper attempts to review recent research findings of utilizing red mud as a road material in pavement structures, including road bases and asphalt mixtures. Previous laboratory investigations indicated that red mud is feasible to be used as a raw material in road bases with satisfactory unconfined compressive strength (UCS), frost resistance and durability performance, but their durability during the service life of the pavement is still unclear. The incorporation of red mud in asphalt mastics was found to be able to improve its stiffness, such as softening point, complex modulus and viscosity. In terms of asphalt mixtures, replacing limestone filler in mixtures by red mud resulted in some positive effects on mechanical behaviors, high bulk density and good rutting resistance. However, the moisture susceptibility and raveling resistance of asphalt mixtures became relatively worse because of the addition of red mud. Referring to this overview, it is necessary to qualify the long-term service performance of road base materials prepared with red mud by using laboratory accelerating evaluation or road trials. For asphalt mixtures prepared with red mud, how to improve durability, in particular moisture resistance, will be a future research focus. In addition, the influence of red mud on cracking resistance at low temperature, fatigue properties and long-term durability of asphalt mixtures is still needed to be further explored.

Keywords: Red mud; Pavements; Road base; Asphalt mixture; Asphalt mastic

1. Introduction

The worldwide industrialization and urbanization have promoted a high demand of aluminum, which has in turn accelerated the emission of by-products during the extraction and refining processes of bauxite [1]. Red mud (Bauxite residue) is a type of industrial waste discharged by alkali treatment of bauxite in order to extract aluminum. Due to its high content of iron oxide, this type of solid waste appears red or brown and is commonly known as red mud. Generally, depending on the raw material quality and processes, generating one ton of alumina will discharge around 0.6-2.5 tons of red mud, and this results in a massive amount of red mud generated in the industry [2]. Because red mud has high alkalinity and potential environment pollution, it cannot be disposed directly and pumping into holding ponds is a commonly used approach [3][4][5]. Even if it is dry, the landfills used for disposal of red mud are still limited due to its considerable amount [6]. Up to now, the cumulative stock of red mud is around 600 million tons in the world, but the comprehensive utilization rate is less than 5% [7]. Therefore, with the promotion of environmental protection, it is highly desirable to minimize red mud waste disposal by adopting effective and efficient methods to utilize it as a secondary resource.

In the literature, enormous attempts have been made to find suitable approaches to realize the innocuous utilization of red mud. Because red mud contains a high amount of metallic oxides and alkalis, there are numerous research studies seeking to further recover alumina, soda, ferric and titanium oxide [8][9]. So far a main application of red mud is to reuse it for producing construction/building materials such as light weight aggregates [10], bricks [11], low density foamed products [12], and traditional ceramics [13]. In addition, red mud can also be widely used in the cement industry, where it can be considered as a raw material or additive for cement production and can act as a replacement of cement in concrete production [14][15]. Due to its alkalis and aluminates presence, as well as high alkalinity, the utilization of red mud to produce geo-polymer has also attracted attention in recent years [16][17]. The developed geo-polymer materials can be applied in different practical fields, such as paving blocks [18], road base materials [19], and phase change materials [20]. Because red mud is the mixture of oxides and hydroxides with fine particles, the purpose of using red mud as absorbents has been motivated, and several studies have reported utilizing red mud for adsorbing pollutants, such as phosphate [21], congo red [22], fluoride [23], nitrate [24], heavy metals [25][26], etc., from water or wastewater. Although there are several attempts to utilize red mud, the consumption of this solid waste is still relatively low.

In recent years, utilization of red mud has been extended to the field of pavements as a road material to replace natural materials used in road bases and asphalt mixtures. A brief overview of the literature reveals that the application of red mud in asphalt mixtures is limited due to a lack of experimental investigations in the laboratory. One research direction is to focus on the mechanical performance development of road base materials after adding red mud. The other one is to study the mechanical behavior of asphalt mixtures by using red mud as the replacement of mineral fillers. This review thus gives a synthetical overview of the development of red mud utilization in pavements during the past decades. First, characteristics of red mud including physical, chemical, as well as environmental impacts, were reviewed and its potential in road pavements was evaluated. Following this, utilizing red mud in road bases and asphalt mixtures were individually summarized by considering the achievements and shortcomings. Finally, conclusions and

recommendations were presented and new potential research areas were presented.

2. Generation and waste management strategies of red mud

2.1 Generation of alumina and bauxite residues

Bauxite is one of the most important naturally occurring resources for producing aluminum, with 98% of aluminum estimated to be extracted from bauxite throughout the world [27]. Bauxite is a natural ore which normally composing aluminum hydroxide minerals such as gibbsite (Al(OH)₃), boehmite (γ -AlO(OH)), and diaspore (α -AlO(OH)) [28]. In addition, bauxite contains other impurities, which include quartz (SiO₂), hematite (α -Fe₂O₃), rutile/anatase (TiO₂), kaolinite (Al₂Si₂O₅(OH)₄), and other minor minerals [29].

Because of the difference in terms of the mineral content of bauxite resources, there are different aluminum production methods including the Bayer process, Sintering process, and Combination Bayer-sintering process. The Bayer process developed by Karl Josef Bayer in 1888 is the most commonly used process for commercial aluminum production [2]. This method is suitable for refining bauxite ores with a high Al₂O₃/SiO₂ ratio between 7-10 [30]. To produce aluminum by the Bayer process, the bauxite is first milled and then mixed with water, lime and caustic soda to digest the bauxite components at high temperature and pressure, as shown in Figure 1. It should be noted that the optimal processing conditions, which include the pressure, temperature and the caustic concentration, are significantly affected by the characteristic of bauxite resources. During the digestion stage, the useful minerals in bauxite (gibbsite, boehmite, and diaspore) are digested by the sodium hydroxide liquid and then the aluminum hydroxide (Al(OH)₃) is selectively precipitated, as shown in Equations 1 and 2. These two chemical equations increase their reactions with increasing temperature and caustic soda concentration [31]. Gibbsite:

$$Al_2O_3 \cdot 3H_2O + 2OH^- \rightarrow 2[Al(OH)_4]^-$$
Boehmite and diaspore: (1)

$$Al_2O_3 \cdot H_2O + 2OH^- + 2H_2O \to 2[Al(OH)_4]^-$$
⁽²⁾



Figure 1. A schematic representation of the Bayer process [32].

The Sintering process is unique in China and Russia, which is used to deal with relatively low-grade bauxite ores with Al_2O_3/SiO_2 ratios ranging between 3 and 6 [30]. To produce aluminum, bauxite was first sintered in rotary sintering kilns by adding limestone and soda ash at a temperature above $1200^{\circ}C$ with sodium aluminate (soluble) and calcium silicate (insoluble) being generated, as shown in Equations 3 and 4. The detailed schematic process for the Sintering method is shown in Figure 2.

$$Na_2CO_3 + Al_2O_3 \rightarrow Na_2O \cdot Al_2O_3 + CO_2 \uparrow$$
(3)

(4)

$$2CaO + SiO_2 \rightarrow 2CaO \cdot SiO_2$$

During the sintering process, ferric oxide is combined with soda ash to form sodium ferrite, as shown in Equation 5.

$$Na_2CO_3 + Fe_2O_3 \rightarrow Na_2O \cdot Fe_2O_3 + CO_2 \uparrow$$
(5)

The sintered bauxite clinker is then dissolved by water and the soluble Na₂O·Al₂O₃ enters the solution. The sodium aluminate is finally treated by carbon dioxide to obtain aluminum hydroxide. The retained insoluble iron oxide hydrate and calcium orthosilicate are discharged as the bauxite residue (red mud) [33][34]. It should be mentioned that the Sintering process consumes significantly higher energy than that of the Bayer process and the combined Bayer-sintering process [7].



Figure 2. A schematic representation of the Sintering process. 1-crushing; 2-wet grinding; 3correction of pulp; 4-sintering; 5-leaching; 6-first desilication; 7-thickening; 8-filtration; 9-second desilicatin; 10-carbonization; 11-thickening; 12-filtration; 13-precipitation; 14-thickening; 15-

evaporation; 16-filtration; 17-evaporation; 18-calcination; 19-silo for alumina [2]. The Combination Bayer-sintering process is a developed approach based on the standard Bayer process with the view of resolving the problem of high silica content in some bauxite ores. During this process, as shown in Figure 3, the high-silica bauxite was first subjected to a traditional Bayer caustic leach process. The generated bauxite residue with sodium aluminum silicates was then subjected to the sintered process, and followed by a leaching process with water to reclaim alumina and soda [35][36].



Figure 3. A schematic representation of the Combination Bayer-sintering process [37] Depending on the bauxite grade and the extraction process, there are around 0.6-2.5 tons of red mud generated when producing 1 ton of alumina [38]. It has been estimated that the annual red mud emission in 2018 was over 160 tons worldwide with about 105 million tons generated in China [39].

2.2 Traditional waste management strategy of red mud

The normally adopted waste management method of the processed red mud is by landfilling. However, with the increasing awareness of environmental protection and land conservation, how to dispose of bauxite residue properly is a great challenge to the aluminum industry throughout the world. Because of this, considerable research has been conducted to change present disposal waste management approaches [40][41]. Normally, there are three main technologies for bauxite residue disposal to landfills, including wet processing, dry processing and semi-dry processing [42]. In the wet processing, the bauxite residue is first washed in a thickener to promote the sedimentation of the solids and obtain residue slurry with the solids content ranging between 15%-30%. The residue slurry is then pumped into a storage yard created by dams, tailing ponds and other earthworks to allow the solids to deposit and the supernatant liquid in the storage yard is then collected as recovered caustic soda [43]. As the dilute residue slurry has a high alkalinity with a pH>12, its disposal after wet processing has a risk of leaking caustic and alkaline liquid into subsoil water and triggering environmental hazards to the surrounding area/communities [44].

As landfills are now becoming scarce and limited, the dry stacking method has been adopted by many plants because of its advantages such as reduced space occupation of the bauxite residue and less leakage potential [7][45]. In this process, the bauxite residue is pre-thickening to obtain residue slurry with a solid content over 50% and then deposited in layers with a thickness of 0.4-0.7 m. The high solid content of the residue slurry can be realized through filtration by using drum filters or a plate and frame filter press, as shown in Figure 4. As the top layer of slurry is dry, bulldozers are used to turn up the bottom layer in order to accelerate the drying process and reduce dust emissions. Then, consecutive residue slurry is layered on top to allow the increase in the spatial storage yard, which subsequently reduces the landfill occupied [46].



Figure 4. Plate and frame filter press, Gardanne, France [44].

The semi-dry processing is the result of a co-disposal of bauxite residue slurry generated from the Bayer process and the sintering process. The residue from the sintering process normally has a low permeability coefficient and a high shear yield strength because of its hydration and hardening ability. So, the dried residue from the sintering process can be applied to construct a dam as a direct replacement of clay. Meanwhile, the residue slurry from the Bayer process can be pumped into the dam directly without dewatering [47].

Among these three disposal processes to landfills, some efforts have been done to partially neutralize high alkalinity to minimize its environmental impacts, including seawater neutralization in plants close to the sea, carbonation using waste carbon dioxide obtained during production, as well as accelerated carbonation using intensive farming methods.

Up to now, collaborative attempts are still ongoing to seek reasonable solutions for bauxite residue disposal, such as improving storage, monitoring and safety standards to reduce environment impacts, developing remediation techniques to improve the reclamation potential of storage yard, and promoting reuse approaches to reduce the volume of stocked bauxite residue [48][49]. Even though such measures are being developed, red mud still occupies considerably landfills and sustainable waste management strategies are needed to use red mud as a secondary resource material.

2.3 Sustainable management strategies for utilizing red mud

In the recent two decades, alumina oxide production has experienced a rapid growth all over the world, as shown in Figure 5. The annual alumina oxide output increased from 52.583 million tons in 2000 to 125.999 million tons in 2017, while the annual emission of red mud reached to over 70 million tons [2]. Researchers have been thus working on developing sustainable and economical approaches for utilizing red mud. Table 1 summarizes possible techniques adopted for utilizing red mud in a variety of applications. It can be seen that there are numerous potential approaches for utilizing red mud with relatively established techniques. However, with respect to commercial/industrial implementation, the barriers for red mud utilization should not be neglected. In addition, the materials that red mud would be replacing are very common and easy to get. Because of this, any negative factor or minor impediment features may affect the large-scale application of red mud.





Categories	Final products		
	or detailed	Practicality and Techniques	References
	utilization		
Recovery of valuable elements	Sodium	Mixed red mud with hydrated lime and treated in hydrothermal process; Reduce the red mud alkalinity; Solid residue could be used to produce cement	[50]
	Aluminum	10%-12% sodium ferrite dosage; 1.0-1.2 M ration of calcium oxide to ferrite dosage; 1000-1050 $^\circ C$ sintering temperature with 30-40 min	[51]
	Iron	Iron recovered through a reductive sintering- magnetic separation-digesting process. NaOH solution with 3% concentration was used in the digest process.	[52]
	Sc ₂ O ₃ , TiO ₂	The Sc_2O_3 was leached by the HCl solution, while the TiO_2 was leached by H_2SO_4 solution. Finally recovered by concentrating, hydrolyzing, washing and calcination.	[53]
	Scandium	Extracted from red mud by activated carbon is modified by tri-butyl phosphate (TBP). Not economic.	[51]
Environmental applications	Water purification	Because of its high specific adsorption area, red mud can remove metal ions, cadmium ions and arsenate from water.	[54][55][56]
	Desulfurizer	Absorbent SO ₂ from acid waste gases using Bayer bauxite residue because of its high alkali, large absorption capacity and simple production process.	[57]
	Soil remediation	Using bauxite residue as additives to stabilize polluted soil for minimizing the leaching of heavy metals	[58]
Construction materials	Cement	With high content of $\beta - 2CaO \cdot SiO_2$, red mud can be incorporated as the raw meal to produce Portland cement clinker at high temperature.	[59]
	Glass- ceramics	Sintering red mud contains suitable chemical components (CaO, Al2O ₃ , SiO ₂) for producing glass-ceramics; CaO-SiO ₂ -Al ₂ O ₃ glass-ceramic has been made from red mud and fly ash.	[60]
	Fired building materials	Producing fired bricks with sintering temperature of 1000 °C and a holding time of 120 min; Using red mud and barium carbonate to produce acid proof fracturing proppants through sintering process and the formation of monoclinic celsian (BaAl ₂ Si ₂ O ₈) reduced the acid solubility.	[61][62]

Non-fired building materials	Producing composite baking-free brick with cement and lime-fly ash depending on the combined solidifying performance and reached the 1st-grade of Chinese standard for the brick.	[63]
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3. Characteristics and potential environmental impacts of red mud

3.1 Chemical composition of red mud

In order to make a reasonable use of the bauxite residue, it is of great important to obtain knowledge of its chemical and mineralogical composition so as to understand its unique physicochemical behavior for utilization [64][65]. The types and concentrations of chemical components in the bauxite residue vary widely based on the characteristics of the bauxite ore and the aluminum production process [66][67]. Regardless of extraction process, the dominant chemical oxide components in the bauxite residue are Al₂O₃, Fe₂O₃, SiO₂, TiO₂, Na₂O and CaO, with small quantities of Ce, Cl, Cr, K, Mg, Mn, Nb, P, S, Sr, Th, Y, Zn, Zr, and others [68]. Table 2 shows the main chemical oxide compositions of the bauxite residue generated from different alumina plants worldwide. As shown in this table, the concentrations of these six main components are Al₂O₃ (8-22%), Fe₂O₃ (5-46%), SiO₂ (5-25%), TiO₂ (2.5-17%), Na₂O (3-12%) and CaO (1-45%). It should be noticed that the residues produced from the sintering or combined Bayer-sintering process exhibit higher CaO (around 40%) content than that from the Bayer process (less than 20%). The higher CaO content endows the residue more cementitious behavior that has the potential to be a supplementary cementitious material [7].

Country	Plant	Extraction process	Major composition (wt.%)					Reference	
			AI_2O_3	Fe_2O_3	SiO ₂	TiO ₂	Na ₂ O	CaO	
China	Shanxi	Bayer-sintering	10.05	6.75	22.2	2.55	3.00	42.25	(Liu et al., 2014)
China	Pingguo Alumina Co. LTD	Bayer	18.49	31.26	8.35	6.18	3.23	18.05	(Rao., 2010)
China	Shandong alumina Co.LTD	Sintering	8.03	17.54	18.19	4.81	3.21	44.64	(Zhang et al., 2018)
China	Shandong alumina Co.LTD	Bayer	25.11	36.43	16.93	6.02	12.27	1.54	(Zhang et al., 2018)
China	Zhengzhou Alumina Co. LTD	Bayer-sintering	7.60	8.20	20.40	7.30	3.00	44.70	(Jiang et al., 2007)
China	Shanxi Alumina Co. LTD	Bayer-sintering	9.18	6.66	18.10	6.72	4.00	38.09	(Nan et al., 2009)
Spain	San Ciprián	Bayer	19.8	39.23	8.77	10.09	5.02	4.54	(Pérez-Villarejo et al., 2012)
Brazil	Alunorte company	-	19.0	38.0	19.9	3.83	8.58	0.87	(Hildebrando et al., 2013)
India	Bharat Aluminum Co. Ltd.	Bayer	22.36	34.27	8.31	17.13	6.12	1.73	(Samal et al., 2015)
UK	ALCAN Chemicals plant	-	20	46	5	6	8	1	(Srikanth et al., 2005)
Grece	Aluminum de Grece	-	19.95	40.8	6.80	5.80	2.70	12.60	(Tsakiridis et al., 2004)

Table 2. Chemical compositions of bauxite residue generated from different aluminum plants worldwide [7][43][59][69][70][71][72][73][74][75]

Because of their different chemical oxide components, the mineralogical of the bauxite residue is even more complicated. It has been reported that there are numerus mineral compositions in the bauxite residue, including calcite (CaCO₃), diaspore AlO(OH), calcium aluminium hydrate (x.CaO.yAl₂O₃.zH₂O), quartz (SiO₂), hematite (Fe₂O₃), gibbsite Al(OH)₃, boehmite AlO(OH), rutile (TiO₂), anatase (TiO₂), CaTiO₃, Na₂TiO₃, kaolinite Al₂O₃.2SiO₂.2H₂O, sodalites, aluminium silicates, cancrinite (NaAlSiO₄)6CaCO₃, hydroxycancrinite (NaAlSiO₄)6.NaOH.H2O, chantalite CaO.Al₂O₃.SiO₂.2H₂O and hydrogarnet Ca₃Al₂(SiO₄)n(OH)_{12–4n} [76]. By comparing different production processes, the red mud obtained from the sintering process contains more Calcite (CaCO₃), Dicalcium Silicate (Ca₂SiO₄) and Perovskite (CaTiO₃), while those from the Bayer process contains more Hematite (Fe₂O₃), gibbsite-Al(OH)₃ and goethite-FeO(OH), as shown in Figures 6 and 7.



Figure 6. Mineral compositions of bauxite residue from sintering process: a [77], b [78].



Figure 7. Mineral components of bauxite residues from the Bayer process: a [79], b [18].

3.2 Physical properties of red mud

For practical applications, most research concerns the fineness and grain size of red mud to replace natural powder materials used in products. The particle size of red mud is normally in the range of 0.1 µm-100 µm, which can be detected by different methods. Kumar et al. investigated the grain size distribution of red mud with the help of a laser particle size analyzer and found that the particle diameters X_{10} , X_{50} and X_{90} of red mud were 0.37, 1.57, 56.88 μ m and its particle size was significantly lower and distribution narrower than that of the fly ash used in concrete, as shown in Figure 8(a) [18]. He et al. determined the particle size distribution of red mud based on the Standard Test Method for Particle-Size Analysis of Soils (ASTM D422-63) with the mean particle size (D₅₀) located at 6.3 μ m, while the coarse particles (>75 μ m) content accounted for 24%, as shown in Figure 8(b) [80]. Zhang et al. compared the grain size distribution of red mud from the Bayer and sintering processes by using a laser particle size analyzer, as shown in Figure 9 [69]. The particle size distribution of the Bayer red mud was wider than the sintering red mud and the Bayer red mud containing more fine particles with a grain size under 10 μ m. It is noticed that the red mud tends to aggregation once exposed to air as the particle becomes coarser even after sieving through 0.075 mm. The natural fine grain behavior of red mud makes it easier to be mixed with other powder materials for various applications.



Figure 8. Particle size distribution curves of red mud obtained from different methods: (a) laser particle size analyzer [18]; (b) ASTM D422-63 [80].



Figure 9. Particle size distribution of red mud from the sintering and Bayer processes [69] Moreover, as a fundamental property, the morphology of red mud has been widely characterized by many researchers with the application of scanning electron microscopy (SEM), as presented in Figure 10. The microscopy reveals that red mud is composed of fine particles with the particle size ranging from a few microns to dozens of microns. The red mud is characterized as irregular shapes including flaky and spherical particles [81]. Normally, big aggregates consist of much smaller particles due to agglomeration. Particles are identified as amorphous substances (poorlycrystallized) and relatively dispersed and disordered [82]. Because of the particle agglomeration, there are spaces between fine particles. For comparison to mineral filler of limestone powder in asphalt, the pore size distribution and the specific surface area of red mud were also investigated, as shown in Figure 11. It is indicated that, for similar particle size level, the specific surface area of red mud was obviously larger than that of the limestone powder. The porous nature as well as the high specific surface area of red mud would contribute to its adsorption capacity as well as interfacial bonding behavior.



Figure 10. Scanning electron micrograph of red mud: (a) [82]; (b) [80].



Figure 11. The derivative pore size distribution curves of mineral filler and red mud [69]

3.3 Environmental impacts of red mud

Based on the literature, the environmental influence of red mud normally focuses on its relatively high pH values, heavy metals leaching and radiation safety.

Because of the high alkalinity, heavy metals presence and trace radioactive elements of red mud, it is normally realized as a hazardous material and poses serious threats to the environment. In accordance with Basel Convention on the Control of Trans boundary Movements of Hazardous Wastes and Their Disposal, "Hazardous waste" is defined as a category of a material which possesses one or more hazardous behaviors, such as corrosivity, toxicity, ignitability, reactivity and/or infectivity. With respect to red mud, its ignitability, reactivity as well as infectivity are relatively lower than that of normal hazardous materials. So, the toxicity and corrosivity are the two main parameters to appraise the environmental impacts of red mud. The Toxic Characteristic Leachability Procedure (TCLP) is commonly used to characterize the toxicity of red mud by measuring the leaching concentration of heavy metal elements. Table 3 presents the heavy metal contents of red mud from different aluminum plants compared with the Chinese Identification standard for hazardous wastes (GB5085.1-7, 2007). It is clear that the toxic element contents of all red muds are lower than the Standard level. So, these red muds cannot be defined as a hazardous waste, but a general industrial solid waste. Consequently, in the Chinese National Catalogue of Hazardous Wastes issued by the Chinese government in 2008, red mud is not categorized as a hazardous material.

Due to the presence of heavy metal elements, the radiation safety of red mud is also evaluated. The safety thresholds of red mud, internal exposure index (I_{Ra}) and external exposure index (I_r) , are all under the threshold limits of the Chinese standard Limits of Radionuclides in Building Materials

(GB 6566-2010). It is therefore indicated that utilizing red mud as a building material could meet the national radioactive requirements [69].

The environmental impacts of red mud are mainly because of its high pH value. The pH value of red mud is normally from 9 to 12.5 and can reach the standard threshold limit level (12.5) [18]. The leaching and precipitating of highly alkaline liquid from red mud is a risk to contaminating surface and underground water resources [83]. A massive discharge of red mud as a result of storage structural failure and other accidents leads to environmental disasters because of its high alkaline nature. In 2010, a red mud leakage accident in Hungary led to the death of 10 people and hundreds seriously burned [84]. After this tragedy, the Hungarian government declared that the red mud is poisonous and its corrosivity is attributed to its strong alkalinity [85].

Due to its small size, red mud is able to easily cause dust under the action of wind and thus affect air quality [86]. Moreover, evaporation of high alkali components from red mud could accelerate the formation of high alkaline rainfall, which eventually threatens soil and water [87].

Table 3. Leaching concentrations of toxic metal elements in the leachates of red mud and the standard thresholds [7][69].

Toxic heavy metals	Unit		Taviaita						
		Henan	Guangxi	Shanxi	Indonesia	Shandong	Shandong	thresholds	
		Bayer	Bayer	Combined	Bayer	Sintering	Bayer	thesholds	
Cd	mg/L	<0.003	<0.003	<0.003	<0.003	Ν	Ν	1	
Pb	mg/L	<0.05	<0.05	<0.05	<0.05	0.046	0.813	5	
Cr	mg/L	0.16	0.12	<0.01	<0.01	0.457	8.624	15	
As	mg/L	<0.1	<0.1	<0.1	<0.1	0.682	0.180	5	
Cu	mg/L	<0.01	<0.01	0.027	0.015	0.643	2.776	100	
Zn	mg/L	<0.006	<0.006	<0.006	<0.006	1.203	3.303	100	
Ni	mg/L	<0.01	<0.01	<0.01	<0.01	1.546	1.062	5	

4. Utilization of red mud in road bases

A road pavement is a type of multiple-layer structure, which is normally made up of asphalt mixture surface layers or a concrete surface layer, road base layers and road sub-base layers. As a layered system, each layer bears and carries the load from the above layer and transfers the load to the next layer below [88][89]. As semi-rigid material has high strength, good moisture stability and low cost, it is a commonly used as a road base material for asphalt pavements in China [90]. The semi-rigid base material, as a hydraulic material, is produced by homogenously blending predetermined ratios of water, aggregates (coarse and fine) and hydraulic materials, followed by paving and compaction stages to construct a semi-rigid road base [91][92]. In a semi-rigid road base materials are lime-fly ash stabilized aggregate (LFSG) and cement stabilized aggregate (CSG). With the view to seeking a feasible potential for red mud utilization, there are several investigations that have been carried out to employ red mud as a secondary material instead of the hydraulic material for road base construction.

Liu et al. investigated the strength development of red mud stabilized by lime and fly ash in order to demonstrate the feasibility of using red mud for road base construction [93]. An orthogonal experimental method was employed to analysis the influence of red mud types and dosages on the unconfined compressive strength (UCS) of the stabilized materials. The experimental findings indicated that the stabilized material with red mud dosage of over 50% can meet the Chinese standards strength requirement for road base materials [94]. This is because the boehmite (γ -AlO(OH)) and goethite (FeO(OH)) in red mud participated in a reaction with active materials in fly ash and lime, such as Ca(OH)₂ and tricalcium aluminate (Ca₃Al₂O₆), to produce a short-term reaction with hydrated silica aluminates like gismondine (CaAl₂Si₂O₈ • 4(H₂O)) and CSH being generated after a long-term reaction. The continuous hydration process with curing generated more hydration products, which consequently increases UCS [95].

Mukiza et al. investigated the strength development of a type of eco-friendly material used for road base prepared with 30% of red mud [19]. By optimizing the mix ratios, the stabilized material obtained UCS of 5.92 MPa and 6.66 MPa after 7 days and 28 days curing, respectively. Long-term soaking curing also contributes to the growth of UCS because the alkaline environment promotes the formation of hydration products. The microstructure analysis revealed that the formation of C-S-H, C-A-S-H, N-A-S-H gels, and ettringite with prolonged curing time is responsible for the strength development, as shown in Figure 12. In addition, the Ca_2SiO_4 , Ca_3SiO_5 and AlO_2^- present in red mud are fully involved in hydration reactions.



Figure 12. Micrographs of stabilized material prepared with red mud cured for 3d (a), 7d (b) and 28d (c and d)

Zhang et al. [96] utilized red mud, electrolytic manganese (EMR) and carbide slag (CS) as main raw components to produce a road base material. The developed road base material with the optimized material ratios obtained 7-day UCS of 5.6 MPa, which is higher than the strength threshold (3–5 MPa) of the Chinese standards for highway road base. This is because the incorporation of red mud supplied a high alkalinity environment and promoted the formation of hydraulic substances (C-A-S-H gel and AFt), which in turn contributed to the internal bond strength of the road base materials. In addition, the freezing-thawing test and wetting-drying cycle test revealed that this road base materials had qualified frost resistance and durability. After laboratory testing, the developed road base material was used for road base construction, as shown in Figure 13. The field test indicated that the cored samples after 7 days curing obtained even higher

strength than that in the laboratory. Using red mud to produce road base material not only consumes a significant amount of industrial solid waste, but also delivers a new idea for synergistic utilization of related solid wastes.



Figure 13. Field-test for road base material prepared with red mud [96]

Hu et al. [97] studied the feasibility of utilizing red mud as an original composition to prepare geopolymer-stabilized base material in pavements. Geopolymers are normally defined as inorganic binders prepared by alkali activation of aluminosilicates by means of complex dissolution–reorientation–solidification reactions [98]. The final products of geopolymer reaction are some amorphous alkali aluminosilicate gels which contribute to strength development [99]. Because red mud possesses a certain amount of Al₂O₃ and SiO₂, it has the potential to be applied as a raw material for geopolymer preparation. Red mud and class F fly ash can be selected to produce geopolymer stabilized aggregate base and the influence of curing conditions (temperature and moisture) on final material strength was also studied. The results showed that the 7-day curing UCS of geopolymer-stabilized aggregate base was 10.5 MPa and this value kept growing with the increase of curing temperature. Based on the strength threshold (3–5 MPa) of Chinese standards for highway road base, the geopolymer-stabilized base material prepared with the incorporation of red mud easily reached the strength requirement. In terms of the strength formation mechanism, the incorporation of the high alkalinity red mud could contribute to the geopolymerization process, which in turn forms a dense structure with a high strength.

The research mentioned above indicated that the utilization of red mud as a raw material in road base construction is feasible and able to obtain qualified UCS, frost resistance and durability performance in the laboratory. However, during the long-term service life, the road base is subjected to repeated traffic loading and serious environmental attacks, which might lead to various defects such as micro-cracks and loosened structured layer. It is thus especially important to characterize the long-term performance of road base materials prepared with red mud, including long-term laboratory tests and field observation. Although the leaching behavior and radionuclide content of road base materials with red mud have proved to be lower than that of the related standard thresholds [100][101], it is still necessary to minimize the long-term impact on the environment by various measures in practice.

5. Utilization of red mud in asphalt mixtures

Asphalt mixture used in asphalt pavements is a composite material prepared by using aggregates, mineral filler and bitumen at predetermined ratios in terms of blending and then laid and

compacted into a pavement layer. The mineral filler is an important component in asphalt mixtures, which extends the effective adhesive binder required to bind the aggregates, and increases the stiffness of asphalt mastic and asphalt mixtures [102]. Generally, the mineral filler used for asphalt mixture production is obtained from crushing and ball-milling of limestone boulders. With the increasing awareness of environmental protection and shortage of natural resources, several alternative powder materials have been incorporated into asphalt mixtures as replacement mineral fillers, such as fly ash, diatomite, cement and steel slag filler [103]. In recent years, using red mud in asphalt mixtures to replace mineral filler has also attracted attention. These investigations can be briefly separated into two categories: red mud in asphalt mixtures.

5.1 Red mud in asphalt mastic

In asphalt mixtures, mineral filler and bitumen are assembled to form asphalt mastics and fill the interstices between aggregates [104][105]. The presence of mineral filler stiffens the bitumen which in turn improves the mechanical performance of asphalt mixtures [106]. Due to its specific physical, chemical and geometrical behavior, the mineral filler has an effect on the mechanical and rheological properties of asphalt mastics [107]. Hence, in order to utilize red mud in asphalt mixtures, its influence on properties of asphalt mastic was first investigated by several researchers. The influence of red mud dosages on conventional properties (penetration, softening point and ductility) of asphalt mastic was investigated by Fu et al. [108]. The results demonstrated that the incorporation of red mud from 3% to 15% improved deformation resistance and reduced temperature sensitivity. However, the incorporation of red mud reduced the asphalt ductility which in turn had a detrimental effect on its cracking resistance at low temperature. Based on the microtopography analysis, the small red mud particle with a large surface area absorbed part of the oil in the bitumen and improved the adhesion between them, which in turn enhanced the consistency of asphalt mastics. Zhang et al. [69] evaluated the replacement of mineral filler with red mud on the penetration and softening point of asphalt mastics, as shown in Figure 14. Due to the replacement of mineral filler by red mud, the softening point increased but the penetration decreased with increasing red mud percentage. It demonstrated that the incorporation of red mud may has positive effects on the high-temperature performance improvement of asphalt mixtures. With respect to the same red mud dosage, the mastics with the sintering red mud presented higher softening point and lower penetration results in comparison with the Bayer red mud. The temperature susceptibility of asphalt mastic was characterized by using the penetration index (PI), which was calculated using Equation 1. The PI results indicated that the incorporation of red mud improved the elasticity of asphalt mastic which in turn had a positive effect on the rutting resistance of asphalt mixture.

$$PI = \frac{1952 - 500 \log(Pen_{25}) - 20SP}{50 \log(Pen_{25}) - SP - 120} \tag{1}$$

where, Pen_{25} is the penetration at 25°C (0.1 mm) and SP is the softening point (°C)



Figure 14. Softening point (a) and Penetration results of asphalt mastic prepared with the red mud replacing mineral filler [69]

Zhang et al. [109] characterized the influence of red mud with different FBv (filler-bitumen ratio by volume) on the viscosity of asphalt mastics at high temperature when compared with conventional asphalt mastic with limestone filler, by using the Brookfield Viscometer. The results indicated that the asphalt mastics prepared with red mud exhibited higher viscosity and worse workability than those with limestone filler and fly ash. This is because the porous structure and large Rigden's voids of red mud can absorb and fix more free asphalt which in turn leads to high viscosity and worse workability [110]. Taking the workability in consideration, the optimum FBv of asphalt mastic with the addition of red mud was determined.

The influence of red mud on rheological properties of asphalt mastic was evaluated by using the dynamic shear rheometer (DSR). Zhang et al. [111] evaluated the complex modulus and phase angle of asphalt mastics composed of the same dosage of limestone filler, red mud and three other composite fillers by using frequency sweep tests, as shown in Figure 15. The results indicated that the complex modulus of asphalt mastic with red mud was higher than that of the asphalt mastic with limestone filler. Furthermore, the asphalt mastics prepared with red mud presented slightly lower phase angles than those prepared with limestone filler. It is suggested that red mud has the potential to improve the permanent deformation and elastic behavior of asphalt mastics. Apart from that, the rutting parameter (G*/sin δ) was investigated by different researchers to evaluate the high-temperature performance of asphalt mastics [111][112]. The multiple stress creep and recovery (MSCR) test revealed that the asphalt mastics with limestone and fly ash [109]. The researchers obtained consistent results, indicating that the high temperature performance of the asphalt mastics with limestone and fly ash [109]. The researchers obtained consistent results, indicating that the high temperature performance of the asphalt mastics prepared with the conventional mineral filler.



Figure 15. Master curves of complex modulus and phase angle of asphalt mastics containing different types of mineral fillers [111]

With respect to characterizing the behavior of asphalt mastics with red mud at low temperature, the Bending Beam Rheometer (BBR) and Direct Tension (DT) tests were employed. Based on the BBR test, the asphalt mastic prepared with red mud exhibited larger creep stiffness modulus (S) and smaller creep slope (m value) in comparison with that prepared with limestone filler at the filler-bitumen ratio of 0.9, as shown in Figure 16 [112]. This indicated that the addition of red mud had detrimental effects on the low temperature cracking resistance of asphalt mastic. The DT test obtained a similar trend with the asphalt mastic prepared with red mud having a smaller tensile elongation at break than that of the mastic prepared with limestone filler, as shown in Figure 17 [78]. In order to improve the cracking resistance of asphalt mastics prepared with red mud, hydrated lime and white mud were incorporated together. The white mud is a type of solid waste generated from paper manufacturing, which is mainly composed of CaCO₃ and possesses some impurities such as Na₂O, K₂O, and Fe₂O₃. The results indicated that the incorporation of hydrated lime and white mud can both contribute to the low temperature performance of asphalt mastics prepared with red mud.



Figure 16. BBR test results of asphalt and asphalt mortar prepared with red mud [112]



Figure 17. Tensile elongation at break of different asphalt mastics at -12°C [78]. The bond strength between aggregate and asphalt mastic is a crucial indicator to characterize the capability of the related asphalt mixtures to resist moisture damage [113]. After incorporating red mud, the bonding behavior of the aggregate-mastic interface in both dry and wet conditions were characterized by different methods. The Pull-Off Tensile Strength (POTS) test was applied to investigate the adhesive ability of the asphalt mastics-aggregate interface, as shown in Figure 18 [110]. The results indicated that the adhesion of asphalt mastic was enhanced with the increase of filler-bitumen ratio in the dry condition, while a higher filler-bitumen ratio had detrimental effects on the adhesion behavior under moisture conditions. Under the same filler-bitumen ratio, the asphalt mastic with red mud showed higher adhesion strength than that of the limestone filler, regardless of dry or wet condition. This research also demonstrated that the adhesive property of the asphalt mastic was dominated by the chemical composition of the filler, but not the physical features. The bonding behavior of asphalt mastics investigated by Zhang et al. [78] obtained different results with the asphalt mastic containing red mud showing lower bonding strength than that prepared with limestone powder subjected to moisture attack. It is suggested that the incorporation of red mud has a detrimental effect on the service durability of the corresponding asphalt mixtures, especially in the pluvial region. However, the mixed use of red mud with white mud can improve the moisture resistance of aggregate-asphalt mastic bonding under both shortterm and long-term moisture conditioning, as shown in Figure 19.



Figure 18. BBS test results of the asphalt and mortar in dry condition (a) and moist condition (b) [110].



Figure 19. Bonding strength of different asphalt mastic-aggregate substrate interfaces in dry and wet conditions [78]

Based on the results mentioned above, the addition of red mud resulted in improved asphalt mastic stiffness, such as higher softening point, higher complex modulus and higher viscosity, because of the porous nature of red mud. The high stiffness of asphalt mastic with red mud indicates its potential of rutting resistance of related asphalt mixtures, but the workability of asphalt mixtures during mixing processes then becomes a big concern. With respect to the influence of red mud on the low-temperature cracking resistance and the moisture resistance of asphalt mastics, they can be improved by adding a certain amount of active filler, such as hydrated lime or white mud.

5.2 Red mud in asphalt mixture

As the asphalt mixture is the product for road surfacing in the pavement, its mechanical performance is the final criterion to characterize the probability of utilizing red mud as a replacement of mineral fillers. The influence of red mud on the mechanical properties of asphalt mixtures was investigated by many researchers.

Choudhary et al. [114] prepared Marshall compacted specimens by using red mud and found that the bulk specific gravities of mixtures prepared with red mud (2.505 g/cc) was higher than that of the normal mixture (2.469 g/cc). The increase of red mud content also resulted in an increase in the bulk density of the asphalt mixture [115]. This is because the fine particle size of red mud acts as a binder extender which provides more workability to obtain a higher density. Moreover, the higher specific gravity of red mud plays a role in contributing to a higher density of asphalt mixture. Because of the higher porosity of red mud, red mud mixtures always obtain a higher air void content than that of conventional mixtures at similar binder contents. In addition, the OAC (the binder content corresponding to 4% air voids of compacted Marshall specimens) of red mud mixture (5.66%) was higher than that of the conventional mixture (5.34%) [116]. This indicates that using red mud to prepare asphalt mixtures could consume a higher amount of bitumen, which in turn has the potential to increase the overall cost of the mixture. The lower voids filled with bitumen (VFB) of the red mud mixture demonstrates its potential to be used in hot climate regions due to its lower possibility of bleeding.

Lima et al. [117] investigated the influence of red mud on the rutting resistance of asphalt mixtures

based on the French standard NFP 98-253-1. The results indicated that substituting the mineral powder for 3% of red mud reduced the permanent deformation from 6.10% to 5.31%, as shown in Figure 20. The superior rutting resistance of asphalt mixtures prepared with red mud was also verified based on the Marshall Stability test [116]. The parameter of Marshall Quotient (MQ), which is defined as the ratio of Marshall Stability (kN) to flow (mm) was employed to characterize the asphalt mixture's resistance to permanent deformation. The results indicated that the MQ value of asphalt mixtures prepared with red mud was higher than that of the conventional mixture, which in turn has better rutting resistance. This is the fact that the finer nature of red mud enables it to distribute uniformly in asphalt mixtures and form an integrated structure [118]. In addition, the chemical composition of red mud may also contribute to the superior high-temperature performance of related asphalt mixtures [112].



Figure 20. Permanent deformation performance of compacted asphalt mixtures prepared with red mud [117]

The influence of red mud on moisture susceptibility of asphalt mixtures was characterized by using the Retained Marshall Stability (RMS) based on the ASTM D1075-11 specification [119]. Choudhary et al. [116] found that the RMS value of the mineral filler mixture (88.57%) was higher than that of the red mud mixture (82.28%), but both of them satisfied the criterion of the minimum requirement (75%) [120]. In another publication, Choudhary et al. [114] employed different waste materials as fillers and compared their influences on moisture susceptibility of asphalt mixtures by using the RMS parameter. The results indicated that the RMS of red mud mixture (89.21%) was lower than that of the conventional mixture (93.56%), but the result was higher than that of mixture prepared with glass powder (82.71%), copper tailings (88.56%) and rice straw ash (80.21%). This correlated well with the bonding behavior of asphalt mastics that the asphalt mastic with red mud showed a lower bonding strength than that of the mixture with limestone powder after moisture conditioning [78]. In order to evaluate the raveling resistance of asphalt mixture with red mud as the filler, the Cantabro durability test was performed by Choudhary et al. [116], as shown in Figure 21. The weight loss of mixtures indicates the loss of adhesion after different curing conditions. It can be seen that both mixtures displayed good raveling resistance in the dry condition with a weight loss less than 5%. However, the red mud mixture experienced a greater weight loss (16.07%) than that of the mineral filler mixture (4.74%) in the wet condition, indicating its worse raveling resistance. Zhang et al. [112] performed the Hamburg Wheel Tracking test to evaluate the moisture sensitivity and the rutting resistance of porous asphalt mixtures (air void content 18%) prepared with red mud, as shown in Figure 22. The mixture prepared with red mud showed a lower

rutting depth in comparison with that of the mixture prepared with limestone powder. This suggests that the porous asphalt mixture prepared with the red mud resulted in better moisture resistance.







Figure 22. Rutting depth development of porous asphalt mixtures prepared with limestone filler and red mud [112]

The review on asphalt mixture performance demonstrated that the incorporation of red mud as the replacement of limestone filler has a positive effect on the mechanical behavior of the corresponding asphalt mixtures, such as high bulk density and good rutting resistance. With respect to moisture susceptibility and raveling resistance, many researchers found that the addition of red mud has a detrimental effect on these two parameters. So, how to improve the moisture resistance of red mud asphalt mixtures will be a future research focus. In addition, there is still a lack of research on mixture performance such as cracking resistance at low temperature, fatigue properties, as well as long-term durability.

6. Conclusions and recommendations

This paper focuses on a critical review on the utilization of red mud as a road material in pavements, including using red mud in road bases and asphalt mixtures. In terms of chemical oxide compositions, red mud has a high content of CaO, Fe₂O₃, and Al₂O₃, which endows its more cementitious behavior and considers it as a construction material. With respect to physical features, red mud particles are porous and have a higher specific surface area than that of limestone powder, which contributes to its adsorption capacity and interfacial bonding properties. Referring to its environmental impacts, its toxic trace elements and radiation level are below the standard levels, which demonstrates that red mud is not categorized as a hazardous material.

The utilization of red mud as a road base material in literature can be classified in two aspects consisting of the production of hydraulic-stabilized road base materials and geopolymer-stabilized materials. For hydraulic-stabilized materials in road bases, red mud is normally mixed with fly ash, hydrated lime and cement to form hydrated products (C-S-H, C-A-S-H, N-A-S-H gels). Referring to geopolymer-stabilized materials, red mud is participated in the dissolution–reorientation– solidification reactions and generates amorphous alkali aluminosilicate gels. Laboratory investigations indicate that it is feasible to use red mud as a raw material in road base construction with the required UCS, frost resistance and durability.

In asphalt mixtures, red mud is incorporated as a replacement of natural mineral fillers. In order to characterize the feasibility of utilizing red mud in asphalt mixtures, the influence of red mud on asphalt mastics was investigated. The addition of red mud was found to be able to improve the stiffness of asphalt mastics, such as improved softening point, complex modulus and viscosity. The high stiffness of asphalt mastics with red mud indicates its potential for rutting resistance of asphalt mixtures. With respect to the influence of red mud on low-temperature cracking resistance and moisture resistance of asphalt mastics, adding a certain amount of active filler, such as hydrated lime or white mud, can help to improve these two properties. The incorporation of red mud as the replacement of limestone filler has some positive effects on mechanical properties of asphalt mixtures such as good rutting resistance. With respect to moisture susceptibility and raveling resistance, the addition of red mud might have detrimental effects on these two parameters.

Based on this overview, the authors believe that further research is needed in certain directions so as to promote the application of red mud as a road material in pavements:

- In terms of red mud as road base materials, although UCS, frost resistance and durability were proved to meet the requirement in the laboratory, the long-term performance during the service lifetime is still not clear. It is necessary to qualify the long-term service performance of road base materials prepared with red mud by using accelerated laboratory evaluation or trial testing and road observation.
- 2) With respect to asphalt mastics and mixtures, the incorporation of red mud had detrimental effects on their bonding strength, moisture susceptibility and raveling resistance. How to improve these properties of red mud asphalt mixtures, in terms of surface modification or using other active powders, will be a future research focus. In addition, the adhesion mechanism between bitumen and red mud in both dry and wet conditions still needs further clarification.
- 3) Moreover, previous research mainly focused on the influence of red mud on volume performance, rutting resistance and moisture susceptibility of asphalt mixtures. How the red mud influences the low-temperature cracking resistance, fatigue properties and long-term durability of asphalt mixtures should also be a future research direction.
- 4) The high alkalinity of red mud is still a big challenge for safe utilization as a road material in pavements. Further studies are needed on investigating applicable and economic techniques to neutralize or solidify alkaline compositions prior to or during utilization.

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