Environ Sci Pollut Res (2016) 23:12822–12834
DOI 10.1007/s11356-016-6478-7

REVIEW ARTICLE

Proposal for management and alkalinity transformation of bauxite residue in China

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Received: 15 October 2015 / Accepted: 14 March 2016 / Published online: 29 March 2016

Abstract Bauxite residue is a hazardous solid waste produced during the production of alumina. Its high alkalinity is a potential threat to the environment which may disrupt the surrounding ecological balance of its disposal areas. China is one of the major global producers of alumina and bauxite residue, but differences in alkalinity and associated chemistry exist between residues from China and those from other countries. A detailed understanding of the chemistry of bauxite residue remains the key to improving its management, both in terms of minimizing environmental impacts and reducing its alkaline properties. The nature of bauxite residue and the chemistry required for its transformation are still poorly understood. This review focuses on various transformation processes generated from the Bayer process, sintering process, and combined Bayer-sintering process in China. Problems associated with transformation mechanisms, technical methods, and relative merits of these technologies are reviewed, while current knowledge gaps and research priorities are recommended. Future research should focus on transformation chemistry and its associated mechanisms and for the development of a clear and economic process to reduce alkalinity and soda in bauxite residue.

Keywords Bauxite residue · Waste management · Alkalinity transformation · Neutralization · Amelioration · Chemical mechanism

Introduction

The nonferrous metals industry of China has been developing rapidly under the guidance of the scientific development concept policy (Xu et al. 2012). However, these rapid developments have caused numerous environment issues (Clark et al. 2015; Wu et al. 2015) especially as China is the largest producer of alumina in the world (Si et al. 2013; Liu et al. 2014b). Bauxite residue (red mud) is an alkaline solid generated during the process of alumina production. The accumulative inventory of bauxite residue reached an estimated 0.6 billion tons with an annual growth of more than 70 million tons in China, and currently, almost all bauxite residue is stored on land (Xue et al. 2016). Bauxites alkaline nature is detrimental to the environment, and the dust formed from the surface of disposal areas is harmful to the surrounding biology (Zhang et al. 2008; Johnston et al. 2010; Banning et al. 2014; Lockwood et al. 2015). Furthermore, due to breaching of the dams containing the residue, leaching of alkaline waste from the slurry is a potential problem (Zhou and Song 2004; Pulford et al. 2012; Samal et al. 2013; Zhu et al. 2016a). Nevertheless, some innovative technical options have been developed for the utilization of bauxite residue, but have yet to be applied to industrial processes (Tuazon and Corder 2008; Gräfe and Klauber 2011; Klauber et al. 2011; Kirwan et al. 2013; Pontikes and Angelopoulos 2013). Additionally, with increasing alumina production and the gradually decreasing grade of bauxite, annual production of bauxite residue appears to have increased, which makes its management more prominent in China.

Responsible editor: Philippe Garrigues

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These reasons highlight the need for effective alkalinity transformation of bauxite residue and to manage the environmental impacts of alumina production and contribute to industry sustainability. In recent decades, significant developments on alkalinity transformation of bauxite residue produced from the Bayer, sintering, and Bayer-sintering processes have been made in China (Liu et al. 2014a, b; Zhang 2014b; Pan et al. 2015; Zhu et al. 2016a, b). These processes can be classified in accordance with the conversion medium including alkaline solid transformation, waste acid synergy, acid gas sequestration, chloride salt neutralization, and bio-driven amelioration. Their current status, transformation mechanisms, and potential issues are presented in this review. Furthermore, existing knowledge gaps and research priorities are discussed and recommended.

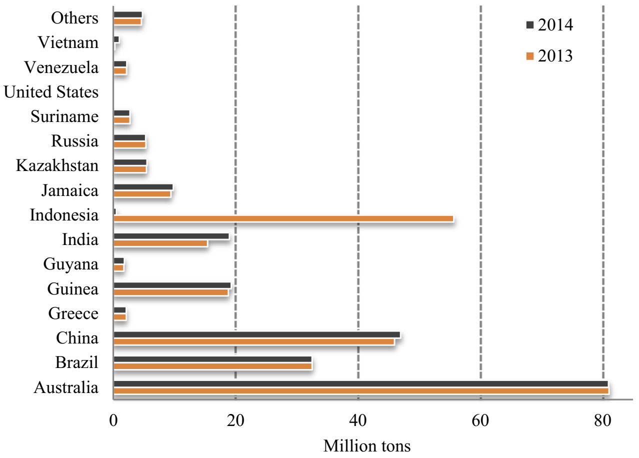

Fig. 2 Bauxite mine production in 2013 and 2014 in China and other producing countries (USGS 2015). Date for bauxite production from USA in 2013 and 2014 was not available

Resources and distribution of bauxite

Reserves and distribution of bauxite

Bauxite is the main mineral resource for alumina production, and 98 % of alumina is obtained from bauxite in the world. It is composed of one or more aluminum hydroxide minerals including gibbsite ($Al(OH)_3$)), boehmite (γ-AlO(OH)), and diaspore (α-AlO(OH)), major impurities such as quartz (SiO_2), hematite (α-Fe_2O_3) and rutile (TiO_2), and other impurities in minor or trace amounts (Bi 2006; MEC 2013).

Bauxite resources are estimated to be as follows: Africa (32 %), Oceania (23 %), South America and the Caribbean (21 %), Asia (18 %), and elsewhere (6 %). Figures 1 and 2 show the reserves and bauxite production in China and other producing countries (USGS 2015). Mine reserves in China occupy sixth place with 830 million tons, while mine production reached 47 million tons during 2014; China is fast becoming the second largest producer in the world (USGS 2015). Almost all of China's reserves are distributed between 7 provinces; Shanxi, Henan, Guangxi, Guizhou, Yunnan, Chongqing, and Shandong, the latter holding a 99 % share of the domestic reserves. Diaspore contains the high content of Al_2O_3 and SiO_2 and is the major type of bauxite in China; therefore, the cost of alumina production is much higher than using imported bauxite. Additionally, many mines are still at the early stages of exploitation due to the limitation of mining costs, traffic conditions, and environmental protection issues. Hence, the struggle between the expansion of alumina production and the supply of domestic bauxite will become more apparent.

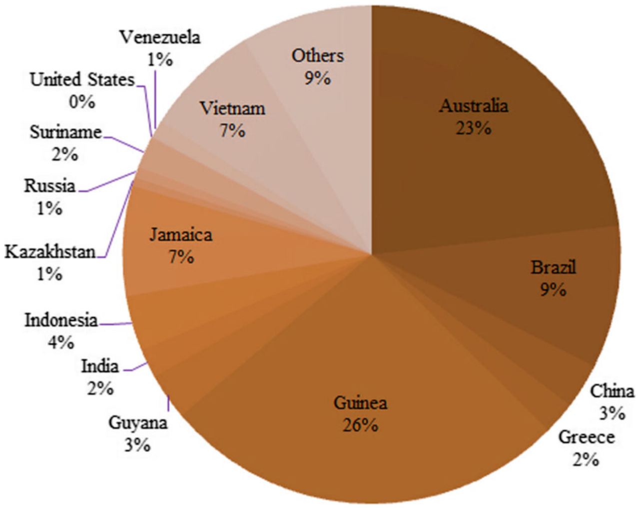

Fig. 1 Reserves distribution of bauxite in China and other countries in 2014 (USGS 2015)

Production processes and distribution of alumina

The production process of alumina, depending on the resource characteristics of bauxite ores in China, is the modified Bayer process for diaspore. Various modifications to the traditional Bayer process have been conducted in an attempt to resolve the problem of high silica concentrations in some ores. The modified processes including Bayer process with dressing, lime Bayer process, and Bayer-sintering combined process have been applied successfully in dealing with Chinese middle/low grade bauxite widely used in Shanxi, Henan, Shandong, and Guizhou (Table 1). The Bayer process with dressing is dedicated to resolve the problem of high silica or high sulfur levels in some bauxite ores. Firstly, bauxite is dressed for pre-desilication and desulfurization, then processed by the Bayer process. Lime Bayer process converts sodium aluminosilicate hydrate to hydrated calcium aluminosilicate by excessive addition of lime in the digestion step of the Bayer process. This reduces the consumption of chemical

Table 1 The production processes of the selected typical companies of alumina plant in China

Refinery	Type of ore	Process	Production time
Zhongzhou Alumina Plant	Diaspore	Bayer process with dressing	2004–Present
		Intensified sintering process	1999–Present
Zhengzhou Alumina Co.	Diaspore	Bayer-sintering combined process	2002–Present
Shandong Alumina Co.	Diaspore	Sintering	1949–Present
Shanxi Alumina Co.	Diaspore	Bayer-sintering combined process	1987–Present
Shanxi Alumina Ltd, CPI	Diaspore	Lime Bayer process	2006–Present
		Bayer-sintering combined process	2010–Present
Pingguo Alumina Co.	Diaspore	Bayer process	1995–Present
Guizhou Alumina Plant	Diaspore	Sintering	1958–Present
		Bayer-sintering combined process	1978–Present
Wenshan Alumina Plant	Diaspore	Bayer process	2012–Present

Note: summarized from the references Liu et al. (2000), Li et al. (2004), Zhao et al. (2004), Fu et al. (2005), Liu et al. (2006)

alkali and achieves improved economic efficiency. During the combined Bayer-sintering process (Fig. 3), high silica bauxite is firstly subjected to a traditional Bayer caustic leach with the resulting bauxite residue containing sodium aluminum silicates which is then sintered with limestone and soda ash. The sintered mass is then leached with water to recover alumina and soda.

The production of alumina in China has been steadily increasing in recent years and approximately half of the alumina produced is distributed in Henan and Shandong; the others are located in Shanxi, Guangxi, Guizhou, Chongqing, Yunnan, and Neimenggu. The distribution of alumina production is presented in Fig. 4.

Management of bauxite residue

Generation of bauxite residue

The alumina extraction process requires a caustic liquor of Na and Ca hydroxides (pH >13) and produces two streams, a green liquor for alumina precipitation and an insoluble solid waste extract, the bauxite residue, for disposal. For each ton of alumina, 0.5–2.0 tons of bauxite residue are produced depending on the bauxite grade and the efficiency of alumina extraction process (Liu et al. 2014a, b; Borra et al. 2015; Zhu et al. 2015). In China, annually, 70 million tons of bauxite residue is produced from the combined extraction processes (Gu 2014;

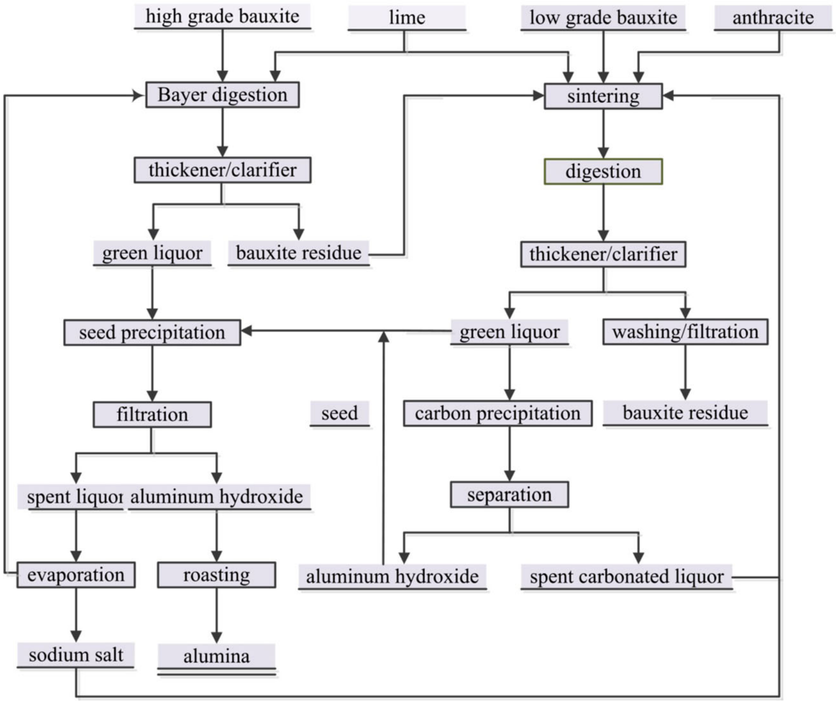

Fig. 3 Schematic of Bayer-sintering combined process (mixed combination process) (modified from Zhang 2014b)

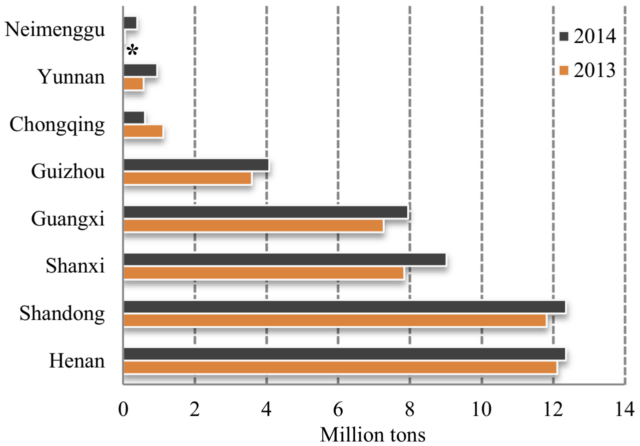

Fig. 4 The distribution of alumina production province in 2013 and 2014 in China (NBSC 2013, 2014). Date for alumina production from Neimenggu in 2013 was available; however, the amount being little was replaced by *

Xu et al. 2015), and the accumulative inventory of bauxite residue has reached an estimated 0.6 billion tons based on its current rate of production.

Disposal of bauxite residue

Globally, the disposal of bauxite residue is a serious concern and considerable research is being directed towards managing this environmental problem; management, environmental legacy, and land conservation are just some of the issues (Klauber et al. 2011; Liu et al. 2014a, b). Almost all bauxite residue generated in China is disposed on land and include methods such as wet processing, dry processing, and semi-dry processing.

Wet processing is the conventional disposal method where residue slurry containing 15–30 % solids is deposited on the land surface in depressions contained by dams. This presents advantages such as low cost land-based disposal, potentially minimizing re-entrainment of wind-blown dust particulates (Zhu and Qi 2009; Rao 2010). Conversely, dry processing is a developing method where residue slurry contains 50–65 % solids; this provides advantages such as reduced surface area, less potential for leakage to groundwater, easy to manage deliquoring, and the recovery of solids for alternative use (Wang and Xiu 2011; Liu et al. 2014a, b; Zhang 2014a, b). Semi-dry processing on the other hand results from the joint disposal of bauxite residue from the Bayer and sintering processes; the requirement of clay is reduced, and bauxite residue from the Bayer process can be pumped to the dam directly without dewatering, which takes full advantage of the reservoir capacity (Wang 2005; Sun 2009).

Utilization of bauxite residue

Bauxite residue has been used for building materials, application in environment materials, and recovery of valuable metals. However, due to its high alkalinity, high water content, and complex composition, few processes have been applied commercially. For example, its use in building materials is limited due to excessive Na and demands are small for its use in environment materials. Any recovery of valuable metals would generate secondary residues, which would then require further disposal (Klauber et al. 2011; Samal et al. 2013; Liu et al. 2014a, b). With respect to these challenges, the alkalinity and complex composition of bauxite residue requires further investigation. Currently bauxite residue is stored in bauxite residue disposal areas (BRDAs), and a focus on the stabilization of their surfaces to prevent wind and water erosion, minimize environmental pollution, and promote organic matter accumulation should remain a focus (Gherardi and Rengel 2003; Mendez and Maier 2008; Courtney et al. 2013).

Alkalinity and associated chemistry

Chemical and mineral compositions of bauxite residue

The main chemical components of bauxite residue are Na_2O, Al_2O_3, SiO_2, Fe_2O_3, CaO, and TiO_2, their percentage distribution depending on the bauxite source and extraction process (Table 1; Fig. 3). The main minerals include sodalite, cancrinite, hydrogarnet, tri-calcium aluminate (TCA), calcite, perovskite, hematite, goethite, and diaspore, their concentrations depending on ore source and digestion process. The chemical and mineral characteristics of bauxite residue are fundamental to its alkalinity and are presented in Tables 2 and 3, respectively.

Alkalinity of bauxite residue

The alkaline mineral compositions of bauxite residue from Bayer process, sintering process, and Bayer-sintering combined process consist of sodalite ($[Na_6Al_6Si_6O_{24}]\cdot[2NaX$ or $Na_2X]$), cancrinite ($[Na_6Al_6Si_6O_{24}]\cdot 2[CaCO_3]$), hydrogarnet ($Ca_3Al_2(SiO_4)_x(OH)_{12-4x}$) and tricalcium aluminate (TCA, $Ca_3Al_2(OH)_{12}$) (Table 3). The following dissolution reactions result in bauxite residue becoming extremely alkaline: sodalite ($[Na_6Al_6Si_6O_{24}]\cdot[2NaX$ or $Na_2X] \rightarrow 8Na^+ + 6Al(OH)_3 + 6H_4SiO_4 + 8X$, X can be OH^-, Cl^-, CO_3^{2-}, or SO_4^{2-}) (Whittington 1996; Whittington et al. 1998), cancrinite ($[Na_6Al_6Si_6O_{24}]\cdot 2[CaCO_3] + 26H_2O \rightarrow 6Na^+ + 2Ca^{2+} + 6Al(OH)_3 + 6H_4SiO_4 + 8OH^- + 2HCO_3^-$), hydro-garnet ($Ca_3Al_2(SiO_4)_x(OH)_{12\text{-}4x} \rightarrow 3Ca^{2+} + 2Al(OH)_3 + xH_4SiO_4 + (6{-}4x)OH^-$) (Paradis et al. 2007) and tri-calcium aluminate ($Ca_3Al_2(OH)_{12} \rightarrow 3Ca^{2+} + 2Al(OH)_3 + 6OH^-$) (Suryavanshi et al. 1999).

The dissoluble alkalinity substances in bauxite residue are NaOH, Na_2CO_3, $NaHCO_3$, $NaAl(OH)_4$, KOH, K_2CO_3. These dissoluble alkalinities can be separated by water in the washing process but still remain in part in the residue. Therefore, the alkaline anions in bauxite residue solution are OH^-,

Table 2 Chemical compositions of bauxite residue generated by different processes in China

Refinery	Extraction process	Na_2O	Al_2O_3	SiO_2	Fe_2O_3	CaO	TiO_2	LOI	Ref.
Zhongzhou Alumina Plant	Intensified sintering	2.54	8.06	17.15	9.82	37.02	4.65	16.98	(Liu 2007)
Zhengzhou Alumina Co.	Bayer-sintering	3.00	7.60	20.40	8.20	44.70	7.30	11.00	(Jiang et al. 2007)
Shandong Alumina Co.	Sintering	3.91	8.80	19.11	12.21	35.52	2.38	11.77	(Wang 2005)
Shanxi Alumina Co.	Bayer-sintering	4.00	9.18	18.10	6.66	38.09	6.72	12.22	(Nan et al. 2009)
Shanxi Alumina Ltd, CPI	Lime bayer	3.57	23.60	17.44	5.97	20.24	6.94	10.62	(Yu et al. 2009)
Pingguo Alumina Co.	Bayer	3.23	18.49	8.35	31.26	18.05	6.18	13.88	(Rao 2010)
Guizhou Alumina Plant	Bayer-sintering	3.1	8.5	25.9	5.0	38.4	4.4	11.1	(Gu et al. 2012)

CO_3^{2-}, HCO_3^-, $Al(OH)_4^-$, $H_2SiO_4^{2-}$ and $H_3SiO_4^-$. Part of OH^-, CO_3^{2-}, HCO_3^-, and $Al(OH)_4^-$ in the residue solution are derived from the dissoluble alkalinity substances. Most of OH^-, CO_3^{2-}, HCO_3^- and $Al(OH)_4^-$, and all of $H_2SiO_4^{2-}$ and $H_3SiO_4^-$ are the dissolution products of sodalite, cancrinite, hydrogarnet, and tri-calcium aluminate. The solution pH is buffered by these alkaline anions, ranging from 9.2 to 12.8 with an average value of 11.3 ± 1.0 (Thornber and Binet 1999). Residue pH will therefore not change until these solids are completely dissolved and their reaction products removed.

Alkalinity transformation of bauxite residue

Alkaline solid transformation

Alkaline solids (lime and hydrated lime) transform alkaline substances by replacing Na_2O of sodalite and cancrinite with CaO or $Ca(OH)_2$ (Eqs. (1) and (2)), precipitating CO_3^{2-} and $Al(OH)_4^-$ as $CaCO_3$ and hydrocalumite at the same time. When the additive CaO reaches saturated state in the low concentration liquid phase, the equilibrium system of CaO–

Table 3 Mineral compositions of bauxite residue generated by different processes in China

Refinery	Extraction process	Sodalite $[Na_6Al_6Si_6O_{24}]\cdot[2NaX\ or\ Na_2X]$[a]	Cancrinite $[Na_6Al_6Si_6O_{24}]\cdot 2[CaCO_3]$[b]	Hydrogarnet $Ca_3Al_2(SiO_4)_x\cdot(OH)_{12\text{-}4x}$[c]	TCA $Ca_3Al_2(OH)_{12}$	Calcite $CaCO_3$	Dicalcium silicate Ca_2SiO_4
Zhongzhou Alumina Plant	Intensified sintering	○	○	20.50	○	1.60	46.40
Zhengzhou Alumina Co.	Bayer-sintering	○	●	○	○	46.8	●
Shandong Alumina Co.	Sintering	●	○	●	○	14	45
Shanxi Alumina Co.	Bayer-sintering	5.8	○	○	○	36.2	35.8
Shanxi Alumina Ltd, CPI	Lime Bayer	○	●	●	●	●	○
Pingguo Alumina Co.	Bayer	○	8.9	33.2	○	4.2	12.5
Guizhou Alumina Plant	Bayer-sintering	●	○	○	●	●	●

Refinery	Perovskite $CaTiO_3$	Hematite α-Fe_2O_3	Goethite α-FeOOH	Diaspore α-AlOOH	Quartz SiO_2	Ref.
Zhongzhou Alumina Plant	8.00	10.20	○	○	●	(Liu 2007)
Zhengzhou Alumina Co.	10.2	7.4	○	○	○	(Jiang et al. 2007)
Shandong Alumina Co.	4.00	7.00	○	○	○	(Wang 2005)
Shanxi Alumina Co.	9.4	6.7	○	3.2	○	(Wang 2012)
Shanxi Alumina Ltd, CPI	●	●	○	●	○	(Li 2002)
Pingguo Alumina Co.	12.5	28.3	●	7.8	○	(Liao et al. 2015)
Guizhou Alumina Plant	●	●	●	○	●	(Gu et al. 2012)

○ not found

● Present, but the content is not clear

[a] X can be OH^-, Cl^-, CO_3^{2-} or SO_4^{2-}; sodalite has between 0 and 6 water of hydration (Whittington 1996; Whittington et al. 1998)

[b] Cancrinite has 0 to 2 waters of hydration depending on the ions in the cage: 0 for $2\cdot CaCO_3$, 1 for $2\cdot NaOH$, and 2 for Na_2SO4 (Whittington and Cardile 1996)

[c] Hydrogarnet ($Ca_3Al_2(SiO_4)_x(OH)_{12\text{-}4x}$) can be expressed as $3CaO\cdot Al_2O_3\cdot xSiO_2\cdot yH_2O$, and x depends on the concentration of SiO_2, Al_2O_3 and Na_2O in aluminate solution and the addition of CaO in digestion process

Notes to Tables 2 and 3

$Na_2O–Al_2O_3–SiO_2–H_2O$ is reached, and Ca^{2+} of CaO or $Ca(OH)_2$ partly replaces Na^+ of sodalite and cancrinite under the conditions of appropriate temperature, the mass ratio of CaO/bauxite residue and appropriately stirring strength. Moreover, $\beta\text{-}CaO \cdot SiO_2$ and $Na_2O \cdot Al_2O_3$ can emerge into a steady solid solution containing some portion of Na_2O, and this solid solution alkali cannot be precipitated by the common processes. With hydration of CaO, Na_2O can be precipitated effectively as Eq. (3). The soluble product of the reactions (Eqs. (1), (2), and (4)) is NaOH which increases the pH of the residue solution comparatively, which can then be leached with water and be recovered to alumina extraction process.

$$[Na_6Al_6Si_6O_{24}]\cdot[2NaOH] + 6CaO + 6H_2O \rightarrow [Ca_3Al_6Si_6O_{24}]\cdot[Ca(OH)_2] + 8NaOH + 2Ca(OH)_2 \tag{1}$$

$$[Na_6Al_6Si_6O_{24}]\cdot 2[CaCO_3] + 6CaO + 6H_2O \rightarrow [Ca_3Al_6Si_6O_{24}]\cdot 2[CaCO_3] + 6NaOH + 3Ca(OH) \tag{2}$$

$$\beta\text{-}CaO\cdot SiO_2(Na_2O \cdot Al_2O_3) + (CaO)_{hydration} + nH_2O \rightarrow CaO\cdot SiO_2\cdot nH_2O + Na_2O\cdot Al_2O_3 \tag{3}$$

$$CaO\cdot SiO_2\cdot nH_2O + Na_2O\cdot Al_2O_3 + 3CaO \rightarrow 3CaO\cdot Al_2O_3\cdot(n\text{-}2)H_2O + CaO\cdot SiO_2\cdot H_2O + 2NaOH \tag{4}$$

The dealkalization of bauxite residue by alkaline solids was developed rapidly and formed the hydrothermal process (hydrothermal alkali process) (Mei et al. 1997; Wang et al. 2010a; Zheng et al. 2010), atmospheric lime process, and lime-soda sintering process (Jiang 2009; Li et al. 2009; Liu et al. 2012) in China. The lime-soda sintering process is different from the other processes as CaO, Na_2CO_3, and bauxite residue are mixed and sintered at a temperature above 1273 K. The oxides in the residue are transformed into calcium silicate, sodium aluminate, calcium ferrite, and calcium titanate. After leaching with water, calcium silicate, calcium ferrite, and calcium titanate remain in the residue and sodium aluminate enters into the leached liquor and is then recovered. The hydrothermal lime process and atmospheric lime process have both been used to dealkalize bauxite residue generated from the sintering process and Bayer-sintering combined process, achieving favorable results.

The processes of alkalinity transformation of bauxite residue by alkaline solids, lime, and hydrated lime are suitable additives, but Al_2O_3 still remains in bauxite residue. In order to achieve dealkalization of the residue and recover Al_2O_3, some researchers have proposed a new approach, the combined process of calcification-carbonization (Fig. 5) (Zhang 2011). During this process (Fig. 5), the equilibrium phase of bauxite residue (sodium aluminosilicate) was firstly converted to hydrogarnet ($3CaO \cdot Al_2O_3 \cdot xSiO_2 \cdot (6{-}2x)H_2O$, $x < 1$). Obtained intermediates which can react with CO_2 are changed into calcium silicate, calcium carbonate and aluminum hydroxide. Calcium silicate and calcium carbonate remain in the residues. These generated residues can then be used directly in the production of cement. Aluminum hydroxide can also be leached with green liquor from the Bayer process and be recycled.

Waste acid synergy

Hydroxides and oxides in bauxite residue can be easily leached with waste acids (H_2SO_4, HCl, HNO_3 and H_3PO_4) (Zhu 2012; Sushil and Batra 2012). Alkali of bauxite residue including attached alkali, free alkali, and chemistry alkali react with these waste acids. The mechanisms of these reactions (taking H_2SO_4 as an example) are summarized as Eqs. (5)–(8) (Lu et al. 2010; Zhu 2012). Insoluble sodalite is leached and the product of sodium sulfate can be crystallized and recycled.

$$OH^- + H^+ \rightarrow H_2O \tag{5}$$

$$2NaAlO_2 + H_2SO_4 + 2H_2O \rightarrow Na_2SO_4 + 2Al(OH)_3 \tag{6}$$

$$Na_2SiO_3 + H_2SO_4 \rightarrow Na_2SO_4 + H_2SiO_3 \tag{7}$$

$$[Na_6Al_6Si_6O_{24}]\cdot[2NaOH] + 4H_2SO_4 + 10H_2O \rightarrow 4Na_2SO_4 + 6H_2SiO_3 + 6Al(OH)_3 \tag{8}$$

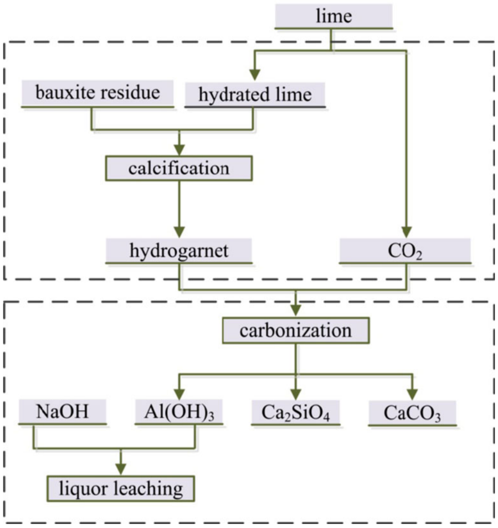

Fig. 5 Schematic of the combined process of calcification-carbonization for dealing with bauxite residue (modified from Zhang 2014b)

However, the leached liquor and residue has always been a complex problem to deal with. This process will produce large amounts of waste liquor and bring about additional environment pollution. Research on alkalinity transformation of bauxite residue by waste acid is limited in China. Unless it is to recover rare and precious metals, bauxite residue will be processed by waste acid (Wang and Li 2007; Gao 2014). If the leachate and leaching residue can be resolved, the alkalinity transformation of bauxite residue by waste acid warrants further research.

Acid gas sequestration

Acid gasses (CO_2 and SO_2) transform the alkaline substances of bauxite residue by the reaction of CO_2 and SO_2 with hydroxide to form bicarbonate and sulfite which can then be oxidized to sulfate under atmospheric conditions. Alkaline transformation with CO_2 also capitalizes on the reversibility of key alkalinity reactions between hydroxide, bicarbonate, and carbonate (Eqs. (9)–(11)) (Wang et al. 2013). Then the hydroxide component of bauxite residue may be converted to carbonate and bicarbonate. Sodium metaaluminate and sodium silicate can also be precipitated by CO_2 as Eqs. (12) and (13) if they are present. Additionally, much of the hydroxide is involved in the solubilization of residual aluminate ($Al(OH)_4^-$). With consumption of free hydroxides from the aluminate anion by CO_2, this will lead to alumina precipitation (Eqs. (14) and (15)) (Johnston et al. 2010). As a result, bicarbonate and carbonate should be the dominant products in this process. However, some work on transformation by CO_2 suggests that the precipitated product is dawsonite by either Eq. (16) or Eq. (17) (Cooling et al. 2002; Guilfoyle et al. 2005). This requires further research.

$$OH^- + CO_2 \rightarrow HCO_3^- \tag{9}$$

$$OH^- + HCO_3^- \rightarrow CO_3^{2-} + H_2O \tag{10}$$

$$H_2O + CO_2 \rightarrow HCO_3^- + H^+ \tag{11}$$

$$NaAlO_2 + CO_2 + 2H_2O \rightarrow NaHCO_3 + Al(OH)_3 \tag{12}$$

$$Na_2SiO_3 + CO_2 + H_2O \rightarrow Na_2CO_3 + H_2SiO_3 \tag{13}$$

$$NaAl(OH)_4 + CO_2 \rightarrow NaHCO_3 + Al(OH)_3 \tag{14}$$

$$2NaAl(OH)_4 + CO_2 \rightarrow Na_2CO_3 + 2Al(OH)_3 + H_2O \tag{15}$$

$$NaAl(OH)_4 + H_2CO_3 \rightarrow NaAlCO_3(OH)_2 + 2H_2O \tag{16}$$

$$NaAl(OH)_4 + CO_2 \rightarrow NaAlCO_3(OH)_2 + H_2O \tag{17}$$

Transformation reactions with hydroxide, sodium metaaluminate, and sodium silicate by SO_2 are somewhat distinct with transformation by CO_2 (Eqs. (18)–(21)) (Chen and Nie 2007; Liu and Li 2015). Furthermore, prolonged treatment of bauxite residue with SO_2 exhausts the free Na^+ and increases the H^+ concentration and, thereby, accelerates the dissolution of sodalite ($[Na_6Al_6Si_6O_{24}] \cdot [2NaOH]$) (Eq. (22)) (Smith 2009). Generated sulfite oxidizes further to sulfate in the presence of O_2 (Eq. (23)).

$$2OH^- + SO_2 \rightarrow SO_3^{2-} + H_2O \tag{18}$$

$$H_2O + SO_2 \rightarrow H_2SO_3 \tag{19}$$

$$2NaAlO_2 + SO_2 + 3H_2O \rightarrow Na_2SO_3 + 2Al(OH)_3 \tag{20}$$

$$Na_2SiO_3 + SO_2 + 2H_2O \rightarrow Na_2SO_3 + H_4SiO_4 \tag{21}$$

$$\begin{aligned}&[Na_6Al_6Si_6O_{24}] \cdot [2NaOH] + 4SO_2 \\ &\quad + 10H_2O \rightarrow 4Na_2SO_3 + 6H_4SiO_4 + 6Al(OH)_3\end{aligned} \tag{22}$$

$$2Na_2SiO_3 + O_2 \rightarrow 2Na_2SO_4 \tag{23}$$

Alkaline transformation of bauxite residue by acid gas has been studied particularly by thermodynamics and kinetics (Nan et al. 2010; Pang et al. 2012; Ji et al. 2014). Usually, bauxite residue can be roasted at different temperatures due to its characteristics prior to transformation with acid gas. Almost all of the attached alkali and/or free alkali in bauxite residue can be neutralized by CO_2 and SO_2 (Wang et al. 2009). It is, however, impossible to change the alkali by acid gas excluding the dissolution of sodalite by SO_2 according to previous literature.

Though the effect of acid gas is not clear, the innovative nature of this transformation is the stable mineral sequestration of CO_2 and SO_2 to reduce atmospheric and industrial carbon dioxide and sulfur dioxide emissions. Meanwhile, it may provide additional benefits such as trapping metal and metalloid contaminants (Yang et al. 2010; Yi and Han 2012; Lin et al. 2013; Liu and Naidu 2014). This process achieves the synergistic action of acid gas and bauxite residue, which has a strong positive role in environmental protection.

Chloride salt neutralization

Calcium chloride and magnesium chloride can also be used to precipitate soluble hydroxides, aluminates and carbonates as insoluble solids. By addition of $CaCl_2$ and/or $MgCl_2$, soluble alkaline substances can be converted to calcite ($CaCO_3$), hydrocalumite ($Ca_4Al_2(OH)_{12} \cdot CO_3$), aluminohydrocalcite ($CaAl_2(CO_3)_2(OH)_4 \cdot 3H_2O$), brucite ($Mg_3(OH)_6$) and hydrotalcite ($Mg_6Al_2(CO_3)(OH)_{16} \cdot 4H_2O$) (Eqs. (24)–(27)) (Hanahan et al. 2004; Menzies et al. 2004; Palmer et al. 2012). The insoluble TCA existing in bauxite residue will participate in forming soluble $Al(OH)_4^-$ ions (Alekseev et al. 1984; Alekseev 1985; Blenkinsop et al. 1985; Whittington et al. 1997; Palmer et al.

2011). It is thought that a combination of two predominant reactions (Eqs. (28) and (29)) involving Na_2CO_3 and NaOH take place. The initial dissolution step of TCA involves a combination of these reactions occurring simultaneously. Equation (28) releases NaOH into bauxite reside until the Na_2CO_3 concentration is depleted, and the pH increases as 6 mol of NaOH are released. Nevertheless, the consumption of 2 mol of NaOH in Eq. (29) reduces the rate at which the pH increases. It is also believed that the dissolution of $Ca(OH)_2$ formed from Eq. (29) causes an increase in OH^- ions. The dissolution of $Ca(OH)_2$ occurs once the majority of NaOH has been neutralized by the dissolution of TCA. The increased concentration of OH^- ions in bauxite residue is then believed to cause further dissolution of TCA, removing OH^- ions by the formation $Ca(OH)_2$. It is not until the majority of TCA is dissolved that the dissolution of $Ca(OH)_2$ becomes dominant. The presence of carbonate also promotes $Ca(OH)_2$ dissolution through the precipitation of calcite, resulting in the additional release of OH^- ions into bauxite residue. The presence of $Ca(OH)_2$ in the liquor promotes the formation of hydrocalumite during the calcium chloride and magnesium chloride neutralization processes (Palmer et al. 2012). Therefore, the concentration of $Ca(OH)_2$ affects the efficiency of the calcium chloride and magnesium chloride neutralization process for bauxite residue both directly and indirectly.

$$4CaCl_2 + 2NaAl(OH)_4 + Na_2CO_3 + 4NaOH \rightarrow Ca_4Al_2(OH)_{12} \cdot CO_3 + 8NaCl \tag{24}$$

$$CaCl_2 + 2NaAl(OH)_4 + 2Na_2CO_3 + xH_2O \rightarrow CaAl_2(CO_3)_2(OH)_4 \cdot xH_2O + 2NaCl + 4NaOH \tag{25}$$

$$CaCl_2 + 3MgCl_2 + NaAl(OH)_4 + Na_2CO_3 + 5NaOH \rightarrow CaCO_3 + Mg_3(OH)_6 + Al(OH)_3 + 8NaCl \tag{26}$$

$$CaCl_2 + 3MgCl_2 + NaAl(OH)_4 + Na_2CO_3 + 5NaOH \rightarrow CaCO_3 + Mg_3(OH)_6 + Al(OH)_3 + 8NaCl \tag{27}$$

$$Ca_3Al_2(OH)_{12} + 3Na_2CO_3 \rightarrow 2NaAl(OH)_4 + 3CaCO_3 + 6NaOH \tag{28}$$

$$Ca_3Al_2(OH)_{12} + 2NaOH \rightarrow 2NaAl(OH)_4 + 3Ca(OH)_2 \tag{29}$$

Ammonium chloride transforms alkaline substances in bauxite residue into chloride, in which free NH_4^+ bonds with metal ions as Eqs. (30), (31), (32), and (33). Products of NH_3 and CO_2 are collected and returned to the cyclic utilization process of NH_4Cl, achieving reuse of ammonium chloride (Eqs. (34) and (35)). This novel technology for sodium elimination was studied by the Chinese Academy of Sciences (Wang et al. 2010b). Bauxite residue leached (hot pressure) with NH_4Cl produces residues that contain insoluble hydroxides formed from hydrolysis, and these residues can be used in the production of cement (Wang et al. 2010c).

$$NH_4Cl + NaOH \rightarrow NaCl + NH_3 + H_2O \tag{30}$$

$$2NH_4Cl + Na_2CO_3 \rightarrow 2NaCl + 2NH_3 + CO_2 + H_2O \tag{31}$$

$$2NH_4Cl + CaO \rightarrow CaCl_2 + 2NH_3 + H_2O \tag{32}$$

$$6NH_4Cl + Al_2O_3 \rightarrow 2AlCl_3 + 6NH_3 + 3H_2O \tag{33}$$

$$NaCl + NH_3 + CO_2 + H_2O \rightarrow NH_4Cl + NaHCO_3 \tag{34}$$

$$CaCl_2 + 2NH_3 + CO_2 + H_2O \rightarrow 2NH_4Cl + CaCO_3 \tag{35}$$

Chloride salt including calcium chloride, magnesium chloride, and ammonium chloride precipitate soluble hydroxides, aluminates, and carbonates as insoluble substances, lowering liquor pH and alkalinity and allowing discharge. The residue can be disposed of more safely or be reused in other applications. Calcium chloride and magnesium chloride neutralization (seawater neutralization) of bauxite residue is used by some coastal refineries in Australia (Menzies et al. 2009; Santini et al. 2011; Couperthwaite et al. 2014). Though an excess of NH_4Cl is required by ammonium chloride neutralization, this technology produces no secondary pollutants. Successful implementation of this technology would be a significant milestone in the development of the chloride salt transformation technology.

Revegetation-driven amelioration

Revegetation-driven amelioration is regarded as a promising way forward in an attempt to ameliorate bauxite residue (Wong and Ho 1994; Jones et al. 2011; Courtney et al. 2014; Santini et al. 2015). The focus is on the potential application of tolerant plants and suitable microorganisms as further amendments to ameliorate the alkalinity and salinity of the restored bauxite residue. For in situ revegetation of the amended bauxite residue disposal areas (BRDAs), it is necessary to understand the application of plants and establishment of microbes and investigate the transfer of sodium and mitigation of pH (Banning et al. 2011; Schmalenberger et al. 2013; Krishna et al. 2014).

Using salt-resistant halophytes may be a potential means of ameliorating the high saline and sodic condition on bauxite residue surface, according to the similar and

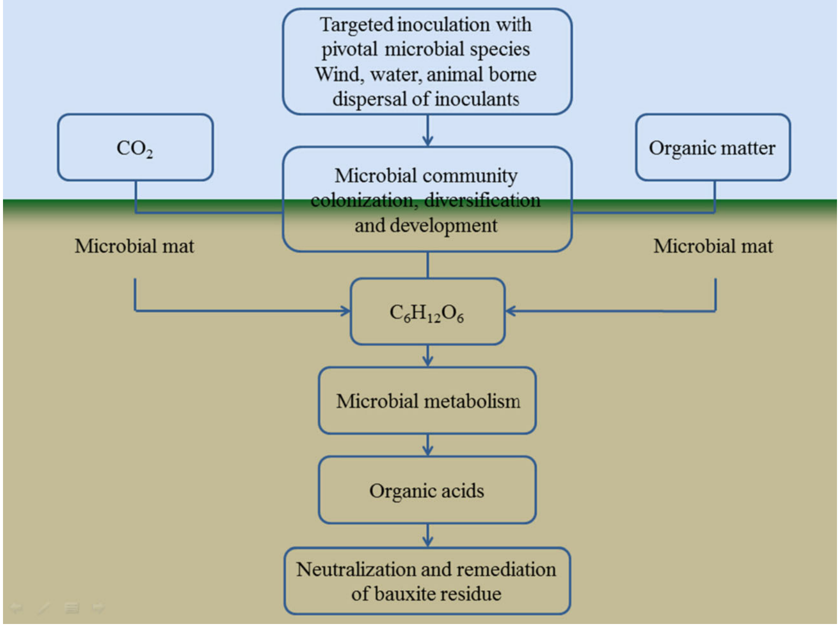

Fig. 6 Mechanisms by which organic acid production possibly contribute to pH neutralization in bauxite residue (modified from Santini et al. 2015)

parallel works in saline-sodic soils (Gräfe and Klauber 2011). Yang et al. (2009) evaluated the effects of alkaline buffering capacity on the performance (relative growth rates, photosynthesis, and Na uptake) of *Chloris virgata* Swartz. in solutions ranging from pH 9.5 to 9.7 with an acid neutralizing capacity (ANC) between 0 and 316 mmol H^+L^{-1}. Yang et al. (2009) revealed that relative growth rate and photosynthetic activity declined with increasing buffering capacity, while the accumulation of Na^+ in tissues was the highest (4.6 wt.%) from solutions buffered at 316 mmol H^+L^{-1} ANC. Li et al. (2010) reported that the effects of various alkali and salt stresses on growth, organic solutes, and cation accumulation in *Spartina alterniflora* Loisel. and demonstrated that the plant was capable of surviving in all treatments under pH ≥8.3 regardless of salinity level.

Microorganisms have beneficial effects towards improving the chemical properties of bauxite residue amended with gypsum, organic wastes, and fertilizers. For example, in restored bauxite residues (amended with compost and gypsum) with different restoration histories, Schmalenberger et al. (2013) established bacterial communities and investigated their successional development and community structure. In a rehabilitated bauxite residue disposal area (amended with gypsum and organic matter), Courtney et al. (2013) and (2014) demonstrated that microbes decreased pH, reduced Na^+ concentrations, and enhanced conditions for the further establishment of flora and fauna. Hamdy and Williams (2001) demonstrated that damaged bacterial cells actively grew following amelioration using nutrients or hay. The microorganisms grew from less than 10 to more than 109 cells per gram of bauxite residue and formed organic acids that lowered the pH from 13 to approximately 7.0.

Production of organic acids by fermentative metabolism is also of interest to bioremediation of bauxite residue because they may contribute to reducing pH by reaction of H^+ with various sources of alkalinity in both pore water and solids (Santini et al. 2015). The mechanisms of organic acid generation may contribute to pH neutralization (Fig. 6). In addition, production of carbon dioxide, extracellular polymeric substances (EPS) that bind soil particles into stable aggregates, altering the ionic balance through solubilization of minerals, and selective uptake of ions from solution (Santini et al. 2015), may all contribute to in situ remediation of bauxite residue.

Nevertheless, the lack of the knowledge of solution composition, dissolution mechanisms, ion exchange processes, chemical speciation, the presence and form of macro and micronutrients, and a model for salt transportation and deposition present future challenges. Insufficient knowledge of the stress and tolerance levels of halophytes, glycophytes, and microbes (including resident, injured, and alkaliphilic microbes) to salinity and alkalinity are further problems. Furthermore, it is unclear how to assess the survivability of microbial inoculants and to understand the factors which influence survival and the extent to which microbial inoculants may influence amelioration of bauxite residue in the long term.

Conclusion

Bauxite residue is dominated by the presence of multiple alkaline solids and dissoluble alkaline substances. A diverse range of methods have been highlighted regarding alkalinity transformations from the view point of mechanisms, processes and deficiencies, all being highly complex and considerably different. The transformation processes being selected based on end-application requirement. Alkaline solid transformation, chloride salt neutralization and bio-driven amelioration primarily focus on the storage of bauxite residue in BRDAs while waste acid synergy and acid gas sequestration focus on the application of bauxite residue following the process. These methods can not completely remove alkalinity, but it must be alleviated for successful disposal, metal recovery, and bioremediation. Nevertheless, these transformation technologies do have drawbacks which limit their commercial application. Key knowledge gaps that require further advancement of understanding include the following:

- The behavior of insoluble alkalinity of bauxite residue in transformation processes is poorly understood.
- Dissolution behaviors of hydrogarnet, tri-calcium aluminate and sodalite are poorly understood.
- Limited knowledge of dissolved salts and liquor dynamic transport within bauxite residue profiles.
- Clear and low-cost technologies to reduce alkalinity and soda.
- The synergistic application of waste acid and bauxite residue.
- Lack of the knowledge of the stress and tolerance levels of halophytes, glycophytes, and microbes to high alkalinity.

Future progress on sustainable management and alkalinity transformation of bauxite residue will be dependent on the development of an improved understanding of the complex neutralization and transformation chemistry of bauxite residue. Continuing research into the chemistry and technology of alkalinity transformation is required. Current research priorities are therefore recommended as follows:

- Development of reaction thermodynamics of acid gas-alkaline substances in bauxite residue and optimize thermodynamics conditions.
- Development of the effect of concentration gradient of acid gas and kinetics of sequestration process.
- Development of an understandable transformation theory for insoluble alkalinity of bauxite residue based on mineralogy.
- Establishment of a comprehensive data set in relation to the dissolution behavior of alumina extraction processes, specifically solids (in particular hydrogarnet, tri-calcium aluminate and sodalite).
- Development of a combined reaction transport and hydrological model for solution flow and reaction processes within bauxite residue profiles.
- Development of a clear and economic process for the reduction of alkalinity and soda in bauxite residue, e.g., direct carbonation technology of residue by stack gas carbon dioxide.
- Achievement of the synergistic action of waste and bauxite residue and development of a critical technology applicable to the hazardous wastes.
- Development of a thorough understanding of the stresses and tolerance levels of halophytes, glycophytes, and microbes to the alkalinity of bauxite residue.

Much of the current and traditional research approaches have focused on removal and separation of alkaline substances. The conventional ideas of alkalinity transformation may require change, and some novel research directions are worth further exploration.

- Stabilization and/or immobilization of alkaline substances in bauxite residue.
- Soil formation process of in situ bioremediation in the amended bauxite residue disposal areas.
- Remediation of bauxite residue disposal area through natural weathering processes.
- Alkalinity transformation of bauxite residue by biomass based on microbiology.

The management of bauxite residue still remains a challenge for the alumina industry and innovative technologies for the development of emission reductions from bauxite residue may be developed further both in China and in the international community.

Acknowledgments Financial supports from the Environmental Protection's Special Scientific Research for Chinese Public Welfare Industry (No. 201509048), the National Natural Science Foundation of China (No. 41371475) and the Open-End Fund for the Valuable and Precision Instruments of Central South University (No. CSUZC201610) are gratefully acknowledged.

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