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# Advances In Bioremediation of Extremely Alkaline Bauxite Residue: A Review

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**Abstract.** Bioremediation of alkaline materials using alkaliphilic and other varieties of microorganisms is one of its own kinds of ecofriendly, sustainable, cost- and energy efficient, clean and green technology. Until recently, the extremely alkaline bauxite residue (pH >11) was treated using various physio-chemical techniques to render it suitable for utilization. However, the present study discusses possible bioremediation techniques to reduce the exorbitant pH and sustain it for a practical duration. It is observed here that there are microbial species, which can proliferate in such extreme chemico-minerological conditions. This study reviews various bioremedation technologies which have successfully been employed by previous studies on soils and wastes. Microbial species identified and their working mechanisms, which have potential to treat alkaline wastes and soils is discussed. This review is an attempt at introducing new avenues and prospects for research into the field of amendment and utilization of bauxite residue. To increase the utilization of bauxite residue, microbial enzyme might constitute a novel technique for waste management.

**Keywords:** Bauxite residue, Biotreatment, Alkalophilic bacteria, physical-chemical methods

## 1 Introduction

Insoluble residue of Bayers process of alumina extraction from bauxite is known as red mud or bauxite residue (BR). It is reported to be rich in iron oxide with a plethora of other elements (such as aluminium, sodium, silicon, calcium, titanium, etc) available in different forms [1]. BR has been observed to demonstrate extortionate alkalinity (pH > 11.5) with high salinity and sodicity [2]. It is also reported that BR exhibits several geotechnical issues such as unreliable compaction characteristics, high collapse potential, and small shear strength [3]. Moreover, this industrial waste has been associated with several geo environmental impediments counting the abundance of metal oxides, erratic and high sedimentation properties, and leaching characteristics in addition to dusting and dispersion features [1, 4].

Moreover, construction and maintenance of impoundment for BR needs skilled manpower along with machineries incurring extravagant expenses. Needless to say, improper impoundment facility creates the risk of caustic exposure to ecosystem, due to leakage of alkaline leachate into groundwater, overflow of materials during inundations and storms, and dusting (mostly $Na_2CO_3$) from pulverized dry surfaces [2]. Such catastrophes have been witnessed in the past during dike failure in Hungary (2010), China (2016) and India (2019), which led to massive loss of human and animal life, vegetation along with disruption of ecological balance for substantial time post-incident. Thus, it is safe to deduce that aggressive and beneficial utilization minimizes the environmental risk and pollution footprint on the eco-system. However, the associated complications of BR restrict its utilization to a bare 2-3% [3].

Available literature demonstrates variety of conventional and novel amelioration techniques being employed on BR, such as use of carbon dioxide, sea-water, organic and mineral acids, industrial and other wastes (fly ash, gypsum, blast furnace slag, copper slag), lime, cement, chemical reagents, physical and mechanical treatments (heating, compaction, consolidation) [5-8]. However, a rebound of pH post-treatment has been observed for these treatments, which poses difficulty in utilizing BR in practical applications [7].

Soil microbial bio-diversity is known to alleviate salinity and pH associated problems in extremely alkaline soils when provided with a supportive environment [9]. Such microbial communities further proliferate by synthesizing organic and inorganic acids, carbon dioxide, extracellular polymeric substances (EPS) and modifying ionic balance of the extreme prevailing conditions. However, the exact mechanism by which microorganisms induce pH reduction in alkaline soils and wastes like BR has not been fully understood.

Furthermore, it is observed that organic acids such as citric acid (CA), lactic acid (LA), acetic acid (AA), and propionic acid (PA) are produced after metabolic fermentation of plant residues [10]. It is a well-known fact that microorganisms adapt to their extreme alkaline environment by synthesizing various forms of acidic secretions such as polysaccharides, proteins, nucleic acids and lipids [11]. The ability of CA and oxalic acid (OA) to reduce BR pH has been explored in depth [7]. In that study, the rebound characteristics of pH have been quantified, in terms of rebound rate of pH (RRP) and rebound termination period ($t_{rt}$), to facilitate analytical comparison between the efficiency of remediation techniques to reduce and maintain BR pH. Further, these studies give an insight into the application of microorganisms, which can synthesize organic acids for BR remediation. It is interesting to note that several studies have attempted bioremediation techniques on BR, which include microbial treatments, vegetation, organic amendments and combinations thereon.

In this study, advancements and recent developments in bioremediation techniques are discussed in order to get an overview of the situation status quo. It also encompasses brief comparisons of factors considered for these studies. It is interesting to note that biochemical mechanisms as well as the prevailing physico-chemico-mineralogical conditions are studied to incorporate the better understanding of studied techniques. However, the primary contribution of this study is the experimental findings observed while investigating the efficiency of CA in mitigating BR alkalinity.

## 2 Bioremediation Techniques

Several bio-based remediation techniques including plantation, use of biopolymers and microbial remediation have been discussed in this section. Plantation, mostly growing grass, is suggested on BR impoundments in order to abate its associated hazards such as dusting, erosion, and leaching [12]. The dusting behaviour of BR has also been addressed by utilizing biopolymers such as guar gum or xanthan gum, along with the improvements in its engineering properties [13]. It is, nonetheless, important to note here that exorbitant alkalinity of BR hinders the efficiency of plantation techniques.

On the contrary, the microbial treatment involves identification of strains of microorganisms with metabolic ability to (1) produce EPS inducing aggregation of particles, (2) synthesize acidic secretions to reduce local pH, (3) produce $CO_2$ which could further reduce the pH of BR, and (4) mutate the ionic balance by solubilizing minerals and selective extraction of ions from pore water.

Advancements in these bioremediation techniques of BR are discussed in the following sections. The observations and results of these studies have been analyzed to understand their efficacy and compared to the basics of soil chemistry and remediation, followed by summary of limitations and research prospects in this field.

### 2.1 Producing EPS to induce aggregation of particles

Studies have reported production of EPS by various strains of bacteria and fungi, which lead to precipitation, bridging and cementation around particles, resulting in improvement of engineering properties at microstructure level [14]. EPS is produced by microbes as a defense-mechanism to adapt and survive in extreme conditions along with providing mechanical stability by increasing inter-particle cohesion. Microbial EPS generally consists of lipids, genetically modified nucleic acids, oligosaccharides and polysaccharides, and other forms of proteins [14]. It was also reported in the study that the particular structure of EPS produced varies for each strain bearing genetical mark of the microbial strain or symbiotic community. However, it has been observed in various studies that the composition and structure of EPS is considerably affected by the prevailing environment of microbial communities [15, 16]. It has been identified that nutrient availability, pH and temperature are the most important factors for bacterial communities [15]. Whereas, the most crucial factor affecting EPS for fungal strains is water potential, in addition to prevailing pH and nutrient concentration [16].

Remediation using EPS produced by microbes is an important alternative for improving the engineering behavior of BR. The efficiency of this technique can be further enhanced by physico-chemically modifying the conditions for these microbial strains. It can be hypothesized based on the available literature that the local conditions for these microbes can be rendered more amicable by identifying microbial strains which can alter the pH and chemical composition of BR by any of the aforementioned mechanisms such as synthesizing acidic secretions, producing $CO_2$, metabolically fermenting vegetation residues or mutating ionic balance. However, the interaction of these microbial communities in conditions prevailing in BR and their combined efficiency should be explored in detail before recommending the remediation technique for industrial applications.

### 2.2 Synthesizing acidic secretions to reduce local pH

The use of acid synthesizing microorganisms in reducing pH poses as a practicable and an alluring alternative for BR remediation. Previous studies have identified various species and strains of microbes which produce acidic secretions such as enzymes, polysaccharides, proteins, nucleic acids, mineral acids and lipids [10, 17-21]. It is reported that microorganisms produce acidic secretions by fermenting organic residues of plants and insects available in the BR ponds [17]. These acidic secretions can be effective in reducing BR pH by donating available $H^+$ to react with alkalinity present in both liquid and solid phases. In addition to that, it has also been observed that various microorganisms undergo autotrophic or heterotrophic metabolisms in the presence of certain chemicals and produce inorganic acids such as sulphuric acid [20, 21]. These microbes present a practicable alternative which can be studied further in order to provide a viable remediation technique of pH mitigation for BR.

### 2.3 Producing $CO_2$ further abating BR pH

Carbon dioxide, whether added mechanically or sequestered from atmosphere or produced by microorganisms, has proved to reduce pH of BR as it reacts with available pore water producing carbonic acid and metal carbonates in some cases. $CO_2$ is generated as an exhaled gas of aerobic and a fermentation byproduct of anaerobic microbes. It is understood that the aerobic $CO_2$ is not efficiently trapped in the pores before releasing into the atmosphere, but fermentation in anaerobic reactions lead to more formation of carbonic acids leading to higher reaction rates with BR alkalinity [9].

### 2.4 Mutation of local ionic balance

It is well established that BR is rich in sodium ions, most of which in reactive or exchangeable form [4]. These exchangeable sodium ions can alter the cation exchange capacity of BR, which has been reported as a cause of pH rebound. In order to reduce the pH and sustain it below the prescribed level, these exchangeable sodium ions have to be extracted or converted to forms that are more inert. Another prospect that can be explored is substituting sodium ions with other metal ions (such as calcium) at these cation exchange sites, which can restrict the adsorbed free sodium from altering BR alkalinity. This has been observed in the case of reduction of BR pH with gypsum. Gypsum solubilizes $Ca^{2+}$ replacing exchangeable sodium by simultaneously producing sulphuric acid leading to reduction of pH in BR. Thus, identifying suitable microorganisms which can solubilize other metal ions or extract $Na^+$ can provide a viable alternative to reduce BR alkalinity. Several strains have been identified [22], which can solubilize $Ca^{2+}$ with change in pH of alkaline media, and surface etching of calcium based minerals. Further, it has been reported that few strains of fungi can selectively extract ions from sodic soils [23]. Identification of such microbial communities, which can alter the ionic balance of BR by selectively extracting alkalinity inducing ions like sodium can lead to further detailed investigations towards finding a strong bioremediation technique.

## 3 Useful Studies on Microbial Bioremediation

Alkaliphiles are microorganisms which find alkaline medium (pH > 9), favorable for their proliferation [24]. They utilize available organic matter as nutrition and synthesize organic acids. Four strains of alkaliphile bacilli were studied by Paavilainen et al. [24] in sodium chloride media. It was reported that various organic acids (carboxylic acids, amino acids, aliphatic and aromatic acids) were produced by these strains depending on the nutrition provided.

On the other hand, haloalkaliphiles require extremely high pH as well as abundance of sodium ions for their proliferation. Alkaliphiles are also found in neutralized or acidic soil and deep sea, but haloalkaliphiles are mostly reported in highly alkaline and saline soils. Few strains of aerobic microbes (amongst genus *bacillus*, *micrococcus*, *pseudomonas* and *streptomyces*) [25] and anaerobic microbes (genus *clostridium*, *amphibacillus*, *thermococcus*) [26-28] are identified from alkaline soils and wastes, which are known to reduce the local pH for their faster proliferation.

Few species of alkaliphiles and haloalkalihiles (such as *vagococcus*, *marinobacter*, *paenibacillus*, *roseinatrobacter*, *rhodobaca*) have been isolated from various soda lakes of India, which prefer a highly alkaline environment of pH 10 to 10.5 and high sodium concentration [29-32]. *Vagococcus* generate exopolysaccharide (a sugar based EPS). Whereas, marinobacter is known to be highly adaptive to its local environment, and used as an alternative to treat contaminated soils [33]. It is interesting to note that *paenibacillus polymyxa* is resistant to chemicals while presenting favorable response to mutations and genome engineering [34].

## 4 Microbial Bioremediation of BR

Bioremediation of BR by certain microbes have proved to be sustainable and eco-friendly [35, 36]. Low levels of injured bacterial cells grew (from <10 to more than 109 cells/g) with addition of required nutrients leading to formation of organic acids in the BR, which might have lowered its pH from 13 to 7 [37].

The microbial strain identified in the aforementioned studies belongs to a wide variety of species: *bacillus*, *lactobacillus*, *leuconostoc*, *micrococcus*, *staphylococcus*, *pseudomonas*, *flavobacterium* and *enterobacter*. It is interesting to note that vegetation growth was observed to be supported in the BR post-treatment. *Cynobacteria*, also known as blue green algae, is considered efficient for remediation of problematic soils [36] and extraction of various elements from them as well as resistant to different forms of various metals [38]. With this in mind, four different strains of *cynobacteria* (i.e. *oscillatoria sp.*, *lyngbya sp.*, *phormidium sp.*, and *microcystis sp.*) were investigated upon in order to identify their efficacy in bioremediation of BR [36]. It was reported that *oscillatoria* and *phormidium* when combined with *aspergillus tubingensis* could act as an effective remediation technique of BR. Microbial technique for pH reduction was also explored by [39], which led to the isolation of EEL02 strain of *bacillus thuringiensis* from the BR impoundments. It was reported that PA, AA, and $CO_2$ (leading to the formation of carbonic acid) were generated by this strain with glucose and peptose as nutrition and at optimal conditions of 25℃ and pH 10, which led to substantial decrease in BR pH within a short span of time. However, no study on the efficacy of these microbes in abating the post-treatment pH rebound of BR has been conducted. Table 1 summarizes the key findings of pertinent studies.

**Table 1.** Summary of available literature

| Authors | Key findings |
|---|---|
| | Bioremediation Techniques |
| Fuller et al. [12] | BR impoundments abate the hazards associated with it: such as dusting, erosion, and leaching |
| | Producing EPS to Induce Aggregation of Particles |
| Flemming and Wingender [14] | EPS production by various strains of bacteria and fungi, results in improvement of engineering properties. |
| Gorret et al. [15] and Papinutti [16] | The composition and structure of EPS is affected by the prevailing environmental conditions (i.e. nutrient availability and concentration, pH, temperature and water potential) of microbial communities |
| | Synthesizing Acidic Secretions to Reduce Local pH |
| Sugaya et al. [10], Mussel et al. [17], Kubicek et al. [18], Wu et al. [19], Schippers and Sand [20], Friedrich et al. [21] | Microbes produce acidic secretions (such as enzymes, polysaccharides, proteins, nucleic acids, mineral acids and lipids) by fermenting organic residues of plants and insects available in the BR ponds which helps in reducing BR pH. |
| Schippers and Sand [20], Friedrich et al. [21] | Various microorganisms undergo autotrophic or heterotrophic metabolisms to produce sulphuric acid which regulates pH mitigation. |
| | Producing $CO_2$ Further Abating BR pH |
| Santini et al. [9] | Fermentation in anaerobic reactions lead to more formation of carbonic acids leading to reduction in pH of BR |
| | Mutation of Local Ionic Balance |
| Kolo et al. [22] | Suitable microorganisms solubilize $Ca^{2+}$ with change in pH of alkaline media which reduces BR alkalinity |
| Wu et al. [23] | Extraction of ions from sodic soils by few strains of fungi |

| | |
|---|---|
| | provides strong bioremediation technique. |
| | Useful Studies on Microbial Bioremediation |
| Paavilainen et al. [24] | Alkaliphiles utilize available organic matter as nutrition and proliferate in alkaline medium. It produces organic acids (i.e. carboxylic acids, amino acids, aliphatic and aromatic acids) |
| Duckworth et al. [25], Podkovyrov and Zeikus [26], Kodama and Koyama [27], Keller et al. [28], | Certain aerobic microbes and anaerobic microbes reduces the local pH for their faster proliferation |
| Kulkarni et al [29], Nandy and Deo [30], Malu et al. [31], Upasani and Desai [32] | Alkaliphiles and haloalkalihiles (such as *vagococcus*, *marinobacter*, *paenibacillus*, *roseinatrobacter*, *rhodobaca*) prefer a highly alkaline environment of pH 10 to 10.5 and high sodium concentration. |
| | Microbial Bioremediation of BR |
| Prasanna et al. [35], Dubey and Dubey [36] | Microbial bioremediation of BR by certain microbes (*bacillus, lactobacillus, leuconostoc, micrococcus, staphylococcus, pseudomonas, flavobacterium* and *Enterobacter*) is sustainable and eco-friendly. |
| Hamdy and Williams [37] | Growth of bacterial cells leads to formation of organic acids in the BR by lowering its pH from 13 to 7. |

## 5 Experimental Findings

### 5.1 Materials used

BR samples used in the present study were collected in disturbed and wet state from the impoundment site of Vedanta Aluminium Limited at Kalahandi district in the state of Odisha, India. Samples were subjected to oven drying at 110±0.5℃ for at least 24 hours to remove entrapped moisture before pulverizing within the required size. Atterberg's limits, specific gravity, particle size distribution and pH of different samples were determined with the help of standard equipment in compliance with relevant ASTM standards. Table 2 presents the results obtained on basic characterization of BR. This sample of BR can be classified as inorganic silt of low plasticity (ML) according to the Unified Soil Classification System (USCS).

In the present study, to examine the effect of organic acid on pH of BR, CA procured from CDH (Central Drug House (P) Ltd, New Delhi, India) was used as an additive at different concentrations varying from 0.1 to 6 M. CA is a naturally occurring organic acid in plants, also can be synthesized by *aspergillus niger*.

**Table 2.** Characteristics of bauxite residue

| Property | Value |
|---|---|
| Specific gravity, $G_s$ | 3.06 |
| Consistency limits (%) | |
| $w_L$ | 42 |
| $w_P$ | 35 |
| $w_{PI}$ | 7 |
| Percent fraction | |
| Sand (%) | 04 |
| Silt (%) | 71 |
| Clay (%) | 25 |
| pH value | 11.4 |
| USCS | ML |

### 5.2 Sample preparation

Acid solution, instead of water, of pre-decided concentrations was added to BR samples of quantity calculated based on optimum water content. These CA admixed BR samples were then used to prepare compacted cylindrical samples using the moist tamping method to attain compaction state of maximum dry unit weight at optimum water content. The samples were sealed in zip-lock bags and preserved inside a humidity chamber in order to maintain the moisture equilibrium.

### 5.3 Testing methodology

Morphological characteristics of BR samples are examined using MERLIN compact Field Emission Scanning Electron Microscope (FE-SEM) (make, ZEISS, Germany). Representative samples in briquette form are prepared from the cured compacted cylindrical samples stored in humidity chamber. Each briquette is sputtered with gold coating using Q150R ES Sputter Coater (make, Quorum, UK) before conducting SEM analysis in order to obtain clear morphological characteristics.

### 5.4 Results and discussions

Fig. 1 presents SEM images of BR samples preserved under maintained moisture and temperature conditions after admixing with CA solutions. The overall morphological features of untreated BR can be observed in Fig. 1 (a). It can be observed from the figure that the untreated BR particles are mostly angular with few fluffy aggregations, which might be due to sodalite and calcite found in it [40]. Fig. 1 (b) shows the CA treated BR compacted sample. Here prominent aggregations can be observed over almost every BR particle. This might have occurred because of changes in mineralogical phases of BR due to its reaction with CA.

Figs. 1(c) & (d) presents the microbial growth observed in CA treated and compacted BR samples. It is interesting to note here that microbial colonies have proliferated, connecting and bridging between BR particles. This observation is encouraging as it is difficult for microorganisms to adapt and proliferate in the extreme conditions of high pH and complex chemico-mineralogical properties. However, further detailed investigation is underway to characterize the microbial communities observed in the present study.

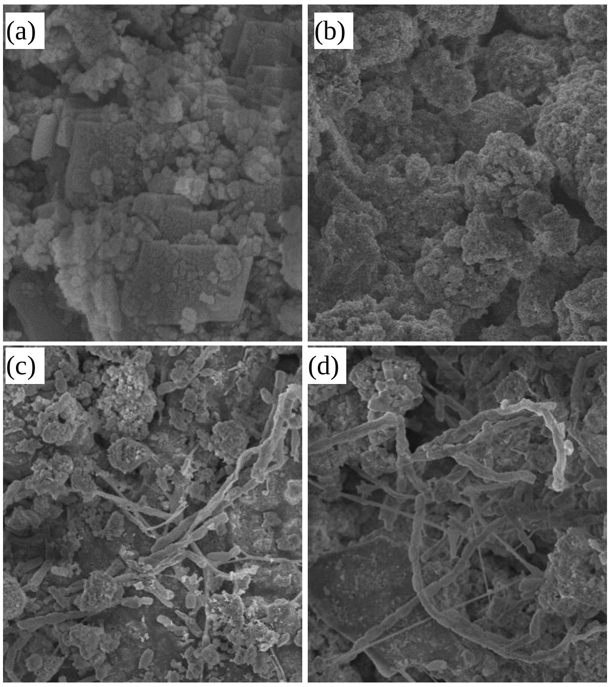

**Fig. 1.** SEM images of (a) untreated BR, (b), (c) and (d) CA treated BR samples

## 6 Conclusions

Bioremediation technique that primarily implements microorganisms seems to provide promising avenues for pH reduction of BR with scope for enhancement of geotechnical behavior. These techniques are considered considerably lesser energy intensive as well. This review study has discussed various possible approaches for utilizing micro organisms based bioremediation techniques. However, the climatic, geoenvironmental and biochemical variations of BR generated and disposed by alumina industry worldwide need to be taken into consideration while choosing any of these techniques for further detailed study. BR impoundments might also be hosting to a few species of microorganisms, which can be studied upon. Furthermore, pH abatement of BR through acid secretion, $CO_2$ generation, selective ions extraction/solubilization as well as aggregation of BR particles through EPS synthesis seems to provide practicable mechanisms for bioremediation of the alkaline industrial waste. The efficacy of such native microorganisms along with externally procured microbial communities in remediating the extremely alkaline BR needs to be investigated in order to provide a more ecofriendly and cost-effective alternative, which can render BR more environmentally benign.

Future endeavors in this regard should be dovetailed to address the bioremediation techniques from three aspects: (a) isolating suitable microorganisms for bioremediation for pH reduction and/or EPS generation, (b) identifying and characterizing biochemical additives that could mutually benefit with the identified bioremediation techniques and (c) conducting a comparative study on the identified techniques to recommend the most ecofriendly and cost-effective approach for industrial application.

Appreciable contribution can be done to the vast ocean of knowledge in the field of geoenvironmental engineering in general and waste utilization, in particular, while attending to the technical and economic aspects of research avenues discussed in this review. The outcome of identified research approaches might assist in providing a green and economical alternative towards utilizing BR.

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