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Invited Review

Mine tailings as a raw material in alkali activation: A review

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Abstract: The mining industry produces billions of tons of mine tailings annually. However, because of their lack of economic value, most of the tailings are discarded near the mining sites, typically under water. The primary environmental concerns of mine tailings are related to their heavy metal and sulfidic mineral content. Oxidation of sulfidic minerals can produce acid mine drainage that leaches heavy metals into the surrounding water. The management of tailing dams requires expensive construction and careful control, and there is the need for stable, sustainable, and economically viable management technologies. Alkali activation as a solidification/stabilization technology offers an attractive way to deal with mine tailings. Alkali activated materials are hardened, concrete-like structures that can be formed from raw materials that are rich in aluminum and silicon, which fortunately, are the main elements in mining residues. Furthermore, alkali activation can immobilize harmful heavy metals within the structure. This review describes the research on alkali activated mine tailings. The reactivity and chemistry of different minerals are discussed. Since many mine tailings are poorly reactive under alkaline conditions, different pretreatment methods and their effects on the mineralogy are reviewed. Possible applications for these materials are also discussed.

Keywords: mine tailings; alkali activation; thermal treatment; mechanical activation; alkaline fusion; heavy metal immobilization

1. Introduction

The mining industry extracts metals that are economically and technically valuable from mined ore. There are different amounts of valuable metals in every mined ore body [1–2], with the leftover material becoming mining waste, mainly mine tailings, which is stored under water in tailings dams without any economic value. The total amount of valuable metal can be extremely low; according to some estimates, 97wt%–99wt% of extracted mineral ends up as mine tailings in tailings dams [3]. The increasing capacity of the mining industry increases the production of different kinds of mining waste residues. The estimated annual solid mine waste productions are 20–25 billion tons, which includes 7–14 billion tons of mine tailings [4].

The properties of the tailings mainly depend on the mineralogy of the mined ore, the geochemistry, the nature of the processing steps, the particle size of the crushed material, and the type of chemicals used in processing. The chemical composition of the tailings that affects the management requirements of mine tailings and tailing dams or the impoundment needs to be designed carefully for a specific mining site. The main minerals in Earth's crust are rich in silicon, aluminum, iron, calcium, potassium, and sodium; as a result, mine tail-

ings rich in these elements are theoretically suitable as a raw material for construction materials. Usually the challenge related to utilization of mine tailings is related to the poor reactivity of their minerals. Additionally, some mine tailings contain a high content of sulfur, heavy metals, and metalloids, since valuable metals are often incorporated into sulfidic minerals. Tailings with heavy metals and sulfates are typically classified as hazardous waste. When sulfidic minerals are in contact with water and oxygen, they are oxidized, which decreases the pH of the surrounding environment, and many heavy metals or metalloids can be dissolved in the acidic conditions [5]. The phenomenon is called acid mine drainage (AMD) that is the most significant concern related to the management of mine tailings in tailing ponds needing careful construction and control during the mining lifetime and after the mine closure. The sulfidic and heavy metal content can hinder the recycling potential of the mine tailings. However, more safety management of hazardous mine tailings should be developed to avoid the environmental and safety problems related to tailings dams.

Since some mine tailings are not hazardous waste, the reuse or recycling potential of mine tailings are attractive solutions compared with discarding tailings into landfills. Currently, a portion of the tailings are utilized as raw materials in

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a paste used to backfill at mining sites. Mine tailings are mixed with a suitable binder material and water, and the generated paste is pumped underground [6]. The final properties of the paste backfill material depend on the physical, chemical, and mineralogical properties of the tailings, the properties of the co-binder, and the amount of water used. In paste backfill technology and stabilization of mine tailings, Portland cement-based binding agent (cement, lime, and other calcium containing materials) is the most commonly used binder. However, Portland cement production yields high CO₂ emissions, which is why it is not ideal for use in paste backfilling. Additionally, when mine tailings have potential acid-generating properties, cement-based paste backfilling is not suitable as the resulting acid can have harmful effects on the generated matrix [6]. Alternatively, pozzolanic products, such as blast furnace slag or fly ash, provide more acid resistance structure and could be utilized in paste backfill technology [7–8].

Since cement-based stabilization is not always the best technology for mine tailings, an alternative method for recycling mining waste is to use the tailings as a raw material in alkali activation for further use as a construction material [9]. Alkali activation is a solidification/stabilization method that can be used mainly with silicon and aluminum-rich raw materials. The raw material is mixed with a suitable alkaline solution, which increases the pH of the mixture, and silicon and aluminum are released to form a new 3D concrete-like structure by sharing oxygen atoms. Additionally, other elements from the precursor, such as Ca, Fe, and Mg, can react in the process to produce a matrix. Alkali activated materials can have high mechanical properties, good durability, and acid and thermal resistance [10]. They also have the capacity to immobilize hazardous components such as heavy metals chemically or physically within their structure [11].

Mine tailings have recently attracted interest as precursors in alkali activation because of their large amounts of silicon and aluminum. Different kinds of mine tailings (e.g., copper, tungsten, vanadium, gold, boron, and kaolinite tailings), each with their own mineralogical properties, are studied in the following sections. The main challenge is the low reactivity of mine tailings. With poor reactivity, mine tailings can be utilized as a filler material with the proper co-binder. In order to utilize mine tailings as a reactive material in alkali activation, the reactivity of the minerals needs to be increased. The reactivity of the minerals has been increased by mechanical activation (e.g., grinding), thermal treatment (calcination), and alkaline fusion. In alkali activation technologies, the final materials could be utilized in paste backfilling, as construction materials in mining sites or to design the dry land-filling instead of discarding into tailing dams.

The objective for this paper is to review the extensive research conducted recently on the alkali activation of mine tailings. Since mine tailings are waste and the waste-based

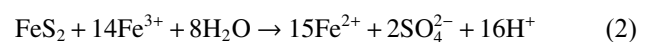
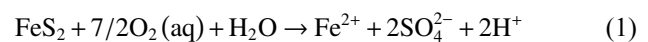
materials cannot be utilized in large-scale applications, there should be laws and policies to allow the use of tailings in construction industries. Before this is possible, there needs to be sufficient data on the use of mine tailings in practical solutions. This paper can be used as a review of how different mine tailings could be used in alkali activation technologies and what are the main conclusions made within this topic. In this paper, the main minerals in mine tailings are discussed and the focus is on understanding the effects of the mineralogy and chemical composition of the different tailings on their potential for alkali activation by analyzing their reactivity in alkaline conditions. Different pretreatment methods for increasing the reactivity of the tailings are introduced, and their effects on the reactivity of different minerals are summarized. The immobilization of heavy metals and sulfates from hazardous mine tailings by alkali activation are also discussed.

2. Mineralogy of mine tailings

Each mine tailing has its own chemical and physical properties, which depend on the mineralogy of the mined ore and the processing steps used to produce the valuable metals. The mineral phases in mine tailings can be divided into two categories: (1) primary phases, including both unreacted and reacted phases present during milling; (2) secondary phases, formed after deposition of the tailings into the environment by precipitation or weathering [12]. The mineralogy, chemical composition, and potential behavior of the tailings under different environmental conditions need to be studied before a proper waste management plan can be designed. Both fresh and weathered mine tailing materials need to be characterized to fully understand the material properties [2].

Minerals are naturally occurring solids with their own structures and chemical compositions. The main minerals in the Earth's crust have been described in detail by Kinnunen *et al.* [13]. Generally, four major classes of minerals are found in the crust. Silicate minerals cover over 90% of Earth's crust, and the most common silicate minerals are feldspar and quartz. Feldspar minerals contain aluminum, calcium, potassium, sodium, and silicon, whereas quartz consists of silicon and oxygen. In addition, mica and olivine (magnesium, iron silicate) are also commonly observed. Carbonates, oxides, and sulfides make up the balance of the minerals.

Many valuable metals, such as Au, Cu, Pb, and Zn, are found in sulfidic minerals, and a large portion of sulfur-rich mine tailings are generated in the mining industry. Sulfidic tailings can cause serious environmental issues because of their oxidation when in contact with oxygen and water. Pyrite is the most typical sulfidic mineral found in Earth. The oxidation of pyrite is shown in Eqs. (1) and (2) [5].



When pyrite starts to oxidize, it yields protons (H^+), Fe^{2+} ions, and sulfates, and decreases the pH of the environment. Fe^{2+} can be oxidized further, releasing Fe^{3+} ions that can act as further catalysts and oxidize more pyrite minerals in the surroundings [5]. As previously mentioned, mine tailings can contain different kinds of heavy metals that can be highly soluble in acidic conditions. When sulfidic minerals are oxidized and the pH decreases, a higher content of leachable heavy metals is released into the mining water. AMD is one of the most serious environmental problems related to the management of mine tailings [5,14]. The bioavailability of hazardous components also depends on the oxidation state of different elements, the mineralogy of the generated tailings, the grain size and surface area of the particles, and the morphology [12]. AMD can be generated both during the mining operation and long after mining closure, increasing the risk for long-term environmental impact. When the leaching of harmful components exceeds the legal limits set for waste, the mine tailings are classified as hazardous; when tailings are discarded in tailings ponds, the ponds require large landfill areas and also careful construction and maintenance even after closure of the mine [5].

Carbonates and silicates, which can neutralize acidic conditions in tailings ponds, also need to be characterized to understand the environmental behavior of mine tailings. Carbonates react with protons, decreasing the acidic condition of the water and thereby providing short-term neutralization of acidic conditions, whereas silicates are long-lasting neutralization minerals. Additionally, during the mining process, some neutralization reagents, such as lime or limestone, are added to the tailings before deposition to control the pH and prevent acid formation [5].

3. Alkali activation

Alkali activation involves a synthesis reaction where silicon and aluminum-rich raw materials are mixed with a highly alkaline solution. When Al and Si ions are released into the solution, they start to form a new 3D network sharing oxygen atoms. Other elements, such as Fe, Mg, and Ca, also contribute to this reaction, forming calcium–(sodium) aluminosilicate hydrate (C(N)ASH) or Mg–Al layered double hydroxide. When calcium content is low, alkali activated materials are called geopolymers. The formed 3D structure can be poorly crystalline or crystalline [15], and the formed matrix can achieve high mechanical strength over a relatively short curing time. In addition, alkali activated material can have high thermal and acid resistance with low CO_2 emissions and energy consumption. These properties make the alkali activated materials a more sustainable alternative for construction materials compared with ordinary Portland cement [11].

Different kinds of aluminosilicate materials can be utilized

as raw materials in alkali activation depending on the reactivity (extent of dissolution, surface area, etc.) of silicon and aluminum under alkaline conditions. The most studied materials are metakaolin and blast furnace slag, and different industrial wastes have been also studied [16–17]. The properties of the alkaline activator affect the final structure of the alkali activated materials. Notably, with higher concentration, higher mechanical strength can be achieved. However, upon a certain limit, the higher concentration can degrade the strength. The most common alkaline reagents are hydroxides (such as NaOH and KOH) and silicates (such as sodium silicate). For some raw materials, usually with higher Ca content, Na_2SO_4 or Na_2CO_3 are suitable alkaline sources. Additionally, $Ca(OH)_2$, CaO or MgO can be also used as an alkaline activator [18]. The alkali activator of each material needs to be studied carefully.

Alkali activated materials can immobilize harmful components, such as heavy metals, within their structure because of their good mechanical properties. The immobilization is based on either chemical interaction or physical encapsulation [19]. Based on the immobilization capacity, alkali activation is an attractive method for mine tailing stabilization. Metakaolin has been used to study the heavy metal immobilization mechanism during alkali activation. When Al and Si form a 3D network, a negative charge occurs based on the charge difference between the ions in the matrix. Cationic species can then balance the negative charge of the matrix. Cationic elements are introduced by the used alkaline solution, the raw material or heavy metals from the waste material. By utilizing different kind of co-binder materials, the other phases participate in the immobilization process making the immobilization mechanism more complex, which is why the immobilization mechanism is challenging to analyze [20]. In physical encapsulation, heavy metals can be entrapped in the material matrix decreasing possible leaching from the structure. Additionally, a high pH of the activating solution and formed matrix can facilitate the precipitation of heavy metals with low solubility [19].

3.1. Alkali activated mine tailings

The reactivity of the raw materials for alkali activation can be studied by testing the solubility of different elements by keeping the material in alkaline conditions for 24 h and analyzing the released elements by inductively coupled plasma spectrometry (ICP) [21]. Additionally, the reactivity of the material can be estimated by evaluating the crystalline structure of the material using X-ray diffraction (XRD) and scanning electron microscopy (SEM). Even though most mine tailings have a large amount of silicon and aluminum, most tailings are poorly reactive under alkaline conditions because of their highly crystalline structure [13,22–25]. The poor reactivity decreases the recycling capacity of mine tailings in alkali activated materials. Many researchers have utilized

poorly reactive mine tailings as raw materials with reactive co-binder (mainly metakaolin or blast furnace slag) in alkali activation. Additionally, the reactivity of the mine tailings can be improved by different treatments [26–27]. However, some tailings, such as lithium, are still inert after thermal treatment [27]. Lithium tailings can be used as inert fillers and have been evaluated in ceramic materials with some co-

binders such as ladle slag/slag [28]. The decision of using mine tailings as a binder/inert filler should be based on their reactivity, which depends on the mineralogical composition of the material and how well the tailings respond to the treatment processes. The final materials can be used in different applications such as dry landfilling, paste backfilling, or construction (Fig. 1).

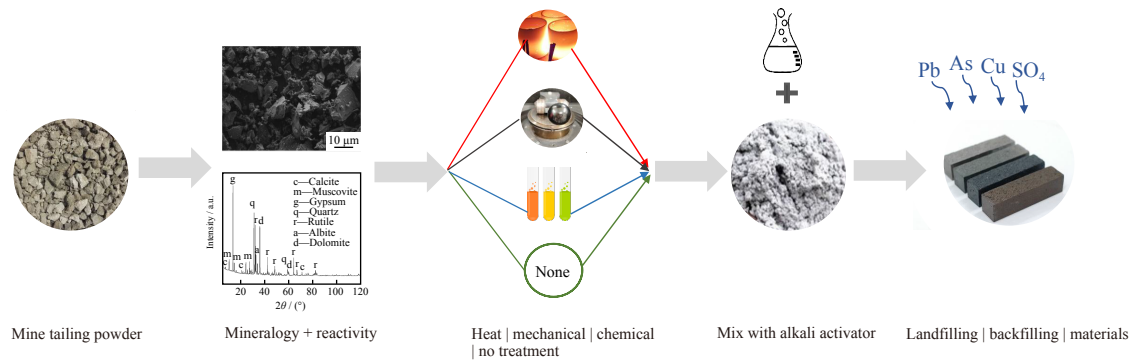


Fig. 1. Alkali activation process for mine tailings.

3.2. Mine tailings as a filler material in alkali activation

Mine tailings with the low reactivity can be utilized as an inert filler in alkali activation with the proper co-binder material [9]. Studies related to the utilization of mine tailings in alkali activation as filler materials are summarized in Table 1. Davidovits [29] showed that alkali activation can be utilized as a stabilizing method for mine tailings from base metal, coal, uranium, and potash mining sites using metakaolin as the co-binder. According to the research, alkali activation can provide a high immobilization capacity for hazardous elements with high mechanical performance in the final matrix. van Jaarsveld *et al.* [30] studied mine tailing consolidation by alkali activation from a mineralogical point of view. They analyzed the geochemistry of the produced tailings in different conditions, which is important to understand before utilizing them in alkali activated materials. The material mainly contained Si, Al, Mg, Fe, Na and SO_3 , and by mixing with fly ash, good mechanical properties, durability, and permeability were achieved when the proportion of tailings in the mixture was less than 70wt%. Above this limit, the mechanical properties of the matrix started to decrease. Additionally, by adding other additives, such as clay, the permeability of the matrix increased [30].

Ahmari and Zhang [10,31–32] and Manjarrez and Zhang [33] studied the alkali activation of copper mine tailings (including quartz, albite, potassium aluminum silicate, and gypsum) with and without co-binders using NaOH as an activator. However, the co-binder was essential in producing high mechanical properties with good durability. The final strength of alkali activated copper mine tailings was improved using fly ash in the mixture. In addition, higher mech-

anical properties were achieved by optimizing the curing conditions, including controlling the pressure or using an elevated curing temperature (90°C). This resulted in a final product with properties suitable for use as a construction material [22,31]. Alkali activated copper mine tailings have also shown high capability to use in road construction materials.

Wan *et al.* [34] studied mine tailings rich in dolomite, calcite, kaolinite, and quartz that were from a vanadium mining site in China. The tailings were used as a raw material in alkali activation with metakaolin as a co-binder and sodium silicate as an activator. Wang *et al.* [35] studied metakaolin as a co-binder for molybdenum mine tailings rich in quartz, calcite, calcium, aluminum, and iron phases. Notably, with higher metakaolin content, greater amount of tetrahedral Si coordination of geopolymer gel was formed, and higher mechanical properties of the final material were obtained. Calcium silicate hydrate (CSH) and belite were also formed in the structure. Similarly, the alkali activated gold mine tailings, rich in dolomite, quartz, mullite, and sulfur, required the suitable co-binder, such as blast furnace slag or metakaolin, since pure mine tailing based material does not harden [24,36]. The strength of the final matrix was controlled by changing the amount of the co-binder, and with only 5wt% of blast furnace slag, final material can be used in paste backfilling with 5 MPa compressive strength after 28 d curing. With higher co-binder content, a matrix appropriate for use as a construction material was produced. Using a high concentration of NaOH (15 M), the workability of the produced paste significantly decreased, which can have a strong impact on the properties of the produced material.

Alkali activation in paste backfill technology has interested researchers as potential replacement of ordinary Port-

land cement (OPC) as the co-binder. The workability of the fresh produced paste need to be analyzed to ensure pumping the paste underground without clogging the pumps. Additionally, the mechanical properties need to be studied to ensure the safe underground stability of the material. By using alkali activated blast furnace slag with gold mine tailings, rich in quartz, albite, mica, sanidine, and microcline, in the preparation of paste backfill material, the workability decreases with increasing compressive strength of the final matrix. Greater strength of the matrix than Portland cement-based paste under optimum curing conditions and with optimal alkali activator concentration (a combination of NaOH and Na₂SiO₃), indicates the suitability of alkali activated slag as a co-binder in paste backfill technology [37]. Alkali activated blast furnace slag provides better acid resistance when used for sulfide-rich tailings paste backfilling compared with Portland cement-based material. Additionally, when alkali activated slag was used as a co-binder, the mechanical strength was still high after 360 d curing [38]. Alkali activated fly ash can be utilized as a co-binder for sulfidic-rich mine waste rocks to produce paste backfill materials or materials for construction [39]. By utilizing sulfidic waste rock as the aggregate, the acid generation potential of the waste in landfills is highly decreased, and environmentally-acceptable materials can be prepared. This is one of the most important benefits of using alkali activation in paste backfill technology instead of OPC.

Alkali activation has also been studied to produce a cover layer that can be prepared on top of the tailings dams. The aim is to produce a material with high water and erosion resistance to isolate the rest of the tailings below the cover. This technology has been studied for low alumina content containing mine tailings by Falah *et al.* [40]. Mine tailings were mixed with alkaline reagent (Na₂SiO₃ with different concentrations) and water, and the paste was cured at different curing temperatures (40 and 60°C). The curing time and temperature have critical roles that impact the mechanical strength properties. The water permeability depends on the alkaline activator concentration. With higher the concentration, the lower the water absorption because of the lower porosity of the final matrix. The materials could be used in construction or as lightweight thermal or acoustic panels [40].

Mine tailings can be recycled in alkali activation technology as inert filler with suitable co-binder materials. For each mine tailing, the mix design needs to be studied carefully since the mineralogy, chemical composition, and physical properties affect the co-binder materials, alkali-activators, and process conditions. The highest possible amount of mine tailings in a mixture need to be evaluated to make the technology economically viable. The availability of a co-binder also plays a critical role since the mining site is not always near the industry/manufacturing location that produces a suitable co-binder.

4. Pretreatment methods

Since mine tailings have shown poor reactivity under alkaline conditions, several pretreatment methods have been investigated to enhance the reactivity of different minerals. Mechanical activation, heat treatment or calcination, and alkaline fusion have been the most extensively studied. The effect of each method on the reactivity of different minerals is discussed in detail in this section.

4.1. Mechanical activation

The aim of mechanical activation is to increase the reactivity of different components, such as Al, Si, Fe, Mg, and Ca, by grinding the material before using in the alkali activation. The increased reactivity is due to the higher amorphous content of the reactive elements after milling [25]. Additionally, the surface area is increased as the particle size decreased. The most important parameter to optimize is the grinding time needed for each kind of material to ensure the most cost effective and sustainable process. The amorphous content, solubility of different elements, particle size distribution, and phase composition are usually analyzed before and after grinding the material. The research results related to mechanical activation are summarized in Table 2.

Jiao *et al.* [41] studied mechanical treatments aimed at improving the final properties of alkali activated mine tailings from a vanadium mining site, which mainly consisted of quartz, feldspar, diopside, mullite, and hematite. Differences in the morphology were shown before and after milling of the tailings. The shape of the particles was more homogeneous and the mean particle size decreased from 31 to 2 µm following a 60-min grinding time, which produced the higher mechanical strength in the final alkali activated material. The amorphous content also increased, and the amount of crystalline quartz was decreased. In addition, grinding produced more solubility of Si and Al. The final alkali activated materials were prepared by mixing mine tailings, fly ash, and solid sodium silicate powder, and the prepared dry mixture was then mixed with water. The resulting material had higher thermal resistance properties than Portland cement composites.

Similar results were obtained for copper mine tailings rich in quartz, albite, dolomite, and chlorite [42]. A 93% higher Al solubility and a 23% higher Si solubility were observed after grinding copper mine tailings for 3 h compared to the non-grounded mine tailing material. No new phases were formed during milling, but a less intensive peak of crystalline silica (albite) was obtained as shown in the corresponding XRD analysis [42]. Antigorite (Mg₃Si₂O₅(OH)₄)-rich mining waste was mechanically activated by Kalinkina *et al.* [25]. The tailings sample was milled with a planetary mill (0.5–10 min). Based on this research, dehydroxylation was clearly related to the formation of more amorphous material

Table 1. Summary of studies using different mine tailings as filler materials in alkali activation methods

Mine tailings	Process parameters	Main elements/phases	Mineralogy	Obtained final properties	Ref.
Metal, coal, uranium, and potash mine tailings	Liquid toxic waste and a geopolymer precursor mixed; curing at 60°C	—	—	High immobilization of heavy metals; high mechanical performance	[29]
Kaltails tailings	Fly ash as a co-binder	Si, Fe, Al, Ca, Mg, Na, and SO ₃	—	Maximum mine tailing content of 70wt%; stronger hardpan achieved with greater fly ash content	[30]
Copper mine tailings	Fly ash as a co-binder; curing at 90°C; pressure used in preparation	—	Quartz, albite, potassium aluminum silicate, and gypsum	Improved mechanical strength with higher co-binder content and elevated curing temperature	[22, 31–32]
Copper mine tailings	No co-binder; curing at 35°C	—	—	Low calcium containing tailings suitable to utilize in road construction	[33]
Sphalerite flotation tailings	Metakaolin as a co-binder; curing at 60°C for 6 h	CaO, MgO, SiO ₂ , and Al ₂ O ₃	Calcite, dolomite, kaolinite, and quartz	Geopolymer gel, belite, lead silicate glass, and CSH gel formed; higher strength obtained and more geopolymer structure formed with higher metakaolin addition	[34]
Molybdenum mine tailings	Metakaolin as a co-binder; curing at room temperature	—	Calcite, andradite, quartz, dolomite, and calcium aluminum silicate	The improved strength of the final material with metakaolin	[35]
Gold mine tailings	Blast furnace slag and metakaolin as co-binders; curing at room temperature	—	Dolomite, quartz, chlinoclore, muscovite, albite, and gypsum	Metakaolin used for adjusting the scarce Al content in the mixture; the effect of co-binder content on the mechanical properties of the final material	[24, 36]
Gold mine tailings	Slag, gypsum, cement, lime, and silica fume as co-binders	—	Quartz, albite, mica, sanidine, and microcline	Appropriate material to utilize for paste backfill technology	[37]
Copper–zinc mine tailings	Blast furnace slag as a co-binder; sodium silicate and sodium hydroxide as activators	Fe, Si, and SO ₃	Pyrite, quartz, albite, calcite, dolomite, muscovite, and chlorite	A great potential of alkali activated slag to replace OPC in paste backfill technology; high long-term mechanical properties achieved	[38]
Sulfidic mine waste rock	Fly ash as a co-binder	—	Chlorite, quartz, illite, and pyrite	Suitable to utilize sulfidic mine waste rock as aggregate in alkali activation	[39]
Cu/Ni mine tailings	30–60 s grinding; curing at elevated temperature	—	Tremolite, chlorite, quartz, talc, magnesium silicate hydrate, dolomite, and calcite	Suitable material for mining closure by optimization of curing conditions and activator concentration	[40]

during the milling process. Additionally, a high solubility of silicon was obtained. Milled tailings with cementitious properties were used as a binder without any alkaline reagent [25]. Phyllite-rich material has also shown cementitious properties after milling [43]. Phyllite, a naturally occurring metamorphic rock, mainly consists of clay minerals, with quartz, albite, muscovite, and chamosite. The amorphous content increased by the mechanical activation. The reduction of muscovite and chamosite were clearly evident, while the amount of quartz

remained constant. The albite content decreased similarly to that reported by Yu *et al.* [42].

4.2. Heat treatment

Heat treatment prior to alkali activation has shown potential for improving the reactivity of mine tailings. The increased reactivity is based on the transformation of the crystalline structure into an amorphous structure and the removal of hydroxyl groups from the starting material. Heating can

Table 2. Summary of studies related to the mechanically activated mine tailings prior to alkali activation

Mine tailings	Process parameters	Mineralogy	Obtained mineralogical changes	Ref.
Vanadium tailings	60 min grinding; 10:1 weight ratio of grinding ball to powder	Quartz, feldspar, diopside, mullite, and hematite	Decreased amount of crystalline quartz; higher mechanical strength of alkali activated material obtained with higher leaching of Al and Si and higher amorphous content	[41]
Copper tailings	1–4 h grinding; 10:1 weight ratio of grinding ball to powder	Quartz, dolomite, chlorite, and albite	Decreased content of albite (lower crystalline silica content)	[42]
Antigorite (Mg ₃ Si ₂ O ₅ (OH) ₄)	600 s grinding; 6:1 weight ratio of grinding ball to powder	Mg and Si minerals	Cementitious properties obtained in milled mine tailings	[25]
Phyllite waste	9–15 min grinding	Clay, quartz, and albite	Reduction of chamosite and muscovite; decreased albite content	[43]

enhance the dissolution of aluminum and silicon units from the initial material, increasing the reactivity of the material under alkaline conditions [44–46]. In addition, more amorphous content has been detected in the heat-treated mine tailings. The thermal resistance of each mine tailing depends on its mineralogy, and the behavior of various minerals under different temperatures is shown in Table 3.

The earliest studies on heat treatment were conducted by Pachego-Torgal *et al.* [47–49]; they introduced a thermal treatment prior to alkali activation to improve the reactivity of tungsten mine tailings rich in quartz and muscovite. A high thermal resistance was observed for quartz, while a portion of the muscovite (12%) remained constant even after heating above 950°C [49]. Similar observations were made by Kiventerä *et al.* [50], who calcined sulfidic tailings at 600 and 900°C, while the main phases that included muscovite remained constant. A reduction in the dolomite phase was observed at 600°C [50] and 750°C [26], which can yield more reactive Ca and Mg to participate in the alkali activation reaction, and then can improve the mechanical strength of the final material. Boron mine tailings, containing Ca and Mg minerals (i.e., colemanite and hydroboracite), turned into amorphous material at 600°C [51]. Illite, which has properties closely related to muscovite, decomposed at 800°C, and albite decomposed above 900°C [26,51–52].

For kaolinite tailings, calcination at 750°C is the most effective method to increase their reactivity. It is likely that kaolinite turns into metakaolin during decomposition [26]. Notably, it is important to optimize the calcination temperature since metakaolin reacts further and yields crystalline and insoluble mullite, which begins to appear at temperatures greater than 800°C [15].

Moukannaa *et al.* [53] studied the use of heat-treated phosphate mine tailings as a raw material in alkali activation with fly ash and metakaolin. Dolomite and calcite were reduced during alkali activation, while the other phases, such as quartz, fluorapatite, and mullite, remained constant. In addition,

copper tailings indicated higher reactivity after calcination at 600°C, at which point the decomposition of crystalline chloride was observed [42].

Phlogopite has high thermal resistance and thus needs a high calcination temperature [21]. Increased Si and Al solubility was detected when phlogopite was heat-treated at 1600°C. In such case when the required temperature is relatively high, the treatment needs to be optimized using fluxes, such as CaO or Na₂O, to maintain a suitable temperature for practical application. Excessively high temperatures can recrystallize the amorphous phase in the heat-treated material, thereby reducing the reactivity and mechanical strength of the final alkali activated material [26].

4.3. Alkaline fusion

Alkaline fusion is another possible method to increase the reactivity of mine tailings prior to alkali activation, and a few studies related to this mechanism have been published. In alkaline fusion, the raw material is mixed with an alkali source and then heated at a high temperature. The temperature is increased above the melting point of the alkali source used in the mixture. After heating, the treated material is alkali activated with an alkaline solution [54]. Kouamo *et al.* [55] studied alkaline fusion with poorly reactive volcanic ash, obtaining a higher amount of soluble aluminosilicates by conversion of minerals such as muscovite, anorthoclase, and diopside, and thereby increasing the reactivity of the ash. Partial decomposition of quartz, albite, and biotite minerals was observed by Tchadjié *et al.* [56], who studied alkaline fusion with granite waste. The most important parameter affecting the reactivity of the material was the amount of alkaline reagent used. Alkaline fusion has also been tested with gold and phosphate mine tailings [57–58]. For the gold mine tailings, the method was tested by mixing an Al₂O₃ additive with the mine tailings at various contents, and NaOH pellets were added to the mixture. Fusion was performed at 550°C for 1 h at an increase rate of 10°C/min. After heating, the mixture

Table 3. Summary of studies related to heat treated mine tailings prior to alkali activation

Mine tailings	Main minerals	Calcination temperatures	Changes in mineralogy	Other comments	Ref.
Tungsten tailings	Muscovite and quartz	950°C	high thermal resistance for muscovite	Increased amount of amorphous phase; thermal behavior similar to phyllosilicate minerals	[47–49]
Gold tailings	Muscovite, dolomite, and quartz	600 and 900°C	Reduced dolomite content at 600°C	Muscovite and quartz remaining constant; no sulfur removed	[50]
Kaolinite tailings	Muscovite, kaolinite, and quartz	750°C	Kaolinite turned into amorphous metakaolin	Possible recrystallization of amorphous phase beyond optimum temperature	[26]
Phosphate mine tailings	Quartz, calcite, fluorapatite, dolomite, mullite, and feldspar	750°C (2 h)	Quartz, fluorapatite, and mullite remaining constant; dolomite and calcite content reduced; feldspar content collapsed	High mechanical strength achieved with 50wt% phosphate tailing in the mixture	[53]
Copper tailings	Quartz, albite, chlorite, and dolomite	400–600°C	Crystalline chlorite dissolved	—	[42]
Boron mine tailings	Colemanite ($\text{Ca}_2\text{B}_6\text{O}_{11}\cdot 5\text{H}_2\text{O}$), hydroboracite ($\text{CaMgB}_6\text{O}_8(\text{OH})_6\cdot 3\text{H}_2\text{O}$), quartz, illite, and calcite	600, 700, and 800°C	Colemanite and hydroboracite turned into amorphous phase at 600°C; illite and calcite decomposed at 800°C	Similar properties of illite with muscovite	[51]
Lithium mine tailings	Quartz, albite, and microcline	750 and 900°C	Albite decomposed partially at 900°C and turned into amorphous phase	No improvement in reactivity	[26]
Apatite tailings	Phlogopite	1600°C	Phlogopite turned into amorphous phase at 1600°C	Temperature optimized for practical applications; using fluxing agents (CaO or Na_2O)	[21]

was mixed with Na_2SiO_3 . The main factor affecting the reactivity was the content of Al_2O_3 rather than the curing temperature. When alkaline fusion was used with phosphate tailings, decomposition of the illite, palygorskite, and dolomite phases and an increased content of the Na-rich crystalline phases were observed.

5. Heavy metal immobilization

The mechanical properties of alkali activated mine tailings have drawn interest, but only a few studies have involved the immobilization of heavy metals through alkali activation of mine tailings. The main research results are shown in Table 4. The nature of the tailings needs to be characterized carefully, and evaluating the mineralogy and chemical composition of the tailings is the most important. If the tailings contain hazardous components such as heavy metals, radioactive materials, acid-generating minerals (sulfidic minerals), and process chemicals, they are classified as hazardous waste [13]. The geochemistry needs to be considered when planning the proper alkali activation method and possible applications for the final material.

The first studies of immobilizing mine tailings by geo-

polymerization were performed by Davidovits [29]. Metakaolin was used as a co-binder and has been used in other studies to understand the immobilization mechanism more within the matrix. The immobilization of harmful elements can be either chemical or physical. In chemical immobilization, the harmful substance can be involved in the structure as a charge balancing element in a geopolymer or other formed phases; another method is precipitation by high pH. In physical encapsulation, the heavy metal particle is entrapped in the formed matrix so that the element does not leach from the structure [19,59].

The heavy metal content can affect the final properties of the alkali activated matrix, and a reduction in mechanical strength has been observed with excessively high metal contents [34,60]. According to some research studies [30], when leaching of Si and Al increases, more heavy metals leach out, which is related to potential breakdown of the matrix produced. The efficiency of the immobilization is also dependent on the chemical property of each metal element. High immobilization of Cu, Fe, Zn, Cu, Pb, Ni, Mn, and Al has been achieved by the alkali activation of different mine tailings [10,36,61]. Anionic elements, such as oxyanions, are poorly immobilized by alkali activation because of their high solu-

bility under alkaline conditions. When mine tailings contain metalloids, such as As or V, the leachability increases after alkali activation as compared with the leaching test before alkali activation [36], possibly because of the soluble properties of oxyanions at a wide range of pH values. However, improved immobilization of As has been obtained with longer curing time [36].

Among oxyanions, a high sulfate content can be a challenge to achieve effective stabilization. When sodium-based alkaline activators are used, sulfates can form sodium sulfates with high solubility. The calcium content in the mixture can play a critical role in oxyanion and sulfate immobilization because of the formation of calcium-based elements [50,62]. Arsenic can form calcium arsenate or calcium arsenite, and sulfates can form gypsum, which has low solubility. An attractive method for the stabilization of heavy metals, oxyanions, and sulfate-containing tailings is via the formation of a needle-like mineral named ettringite in the binding system. Ettringite can be formed with Ca, Al, and SO₄ under proper pH conditions, and it can also include other elements, such as iron. Ettringite has shown a high capability for immobilizing heavy metals within its structure either through chemical bonding or physical encapsulation [63–64]. A possible co-

binder to encourage ettringite formation could be metallurgical slags, such as blast furnace slag or ladle slag, depending on the chemical composition and mineralogy of the initial mine tailings [50,65].

Another method for the immobilization of sulfidic tailings is to use a MgO-activated blast furnace slag as a binding agent for sulfidic-containing mine tailings [66]. This study showed high immobilization of the heavy metals and a good pH buffering capacity resulting from the production of hydrotalcite. The formation of brucite from MgO has been suggested as the reason for the high immobilization of Pb, Zn, and Ni. Based on the high immobilization capacity of alkali activation, this method could be utilized to design dry land-filling with mine tailings or produce a cover layer for tailings landfill areas. For these applications, the leaching of different elements, the durability under different environmental conditions, cold climate resistance, and hydraulic conductivity should be studied.

6. Discussion and conclusion

Mine tailings are the most generated solid waste from the mining industry, and most of the mine tailings are dumped

Table 4. Summary of studies related to immobilization of heavy metals from mine tailings by alkali activation

Mine tailings	Elements/phases	Binding system	Curing conditions	Immobilization	Ref.
Copper mine tailings	Cu, Ni, Zn, Mn, and Pb	Fired coal fly ash + NaOH/Na-Sil	Curing for 72 h at 60°C	Immobilization of all elements during curing of 7 weeks	[61]
Copper mine tailings	Cu, Al, Fe, and Zn	NaOH (15 M)	Pressure used for compacting the samples; curing at 90°C	Effective immobilization of all elements	[10]
Gold mine tailings	Cr, Cu, Ni, Zn, As, V, Sb, and Mn	GBFS/MK as a co-binder; NaOH/Na-Sil as an activator	Curing at room temperature	Effective immobilization of Cr, Cu, Ni, Zn, and Mn; poor immobilization of As, V, and Sb; curing improved immobilization of 18 months	[36]
Sphalerite mine tailings	Pb	Metakaolin as a co-binder	Curing for 6 h at 60°C and for 7 d at room temperature	Effective immobilization of lead when the content was less than 5wt%; the decreased immobilization with too high content of lead	[60]
Dolomite, calcite, kaolinite, and quartz rich mine tailings	Pb	Metakaolin as a co-binder (0–50wt% of mixture)	Curing for 6 h at 60°C and for 7 d at room temperature	The decreased UCS with too much lead (>6wt%); effective immobilization of lead; immobilization related to geopolymer gel formation in matrix	[34]
Gold mine tailings	Cr, Cu, Ni, Zn, As, V, Sb, Mn, and SO ₄	Ca(OH) ₂ as an alkaline reagent; GBFS as a co-binder	Curing at room temperature	Effective immobilization of heavy metals, oxyanions, and sulfates	[50]
Sulfidic tailings	As, Cr, Cu, Ni, Se, Zn, Mo, Pb, Se, Cl, and SO ₄	MgO activated GBFS as a binding system	Curing at room temperature	More effective immobilization of all elements with Mg activated GBFS than an OPC based binding system	[66]

Note: Na-Sil—Sodium silicate; GBFS—Ground granulated blast furnace slag; MK—Metakaolin; UCS—Unconfined compressive strength; OPC—Ordinary Portland cement.

into a controlled landfill area under water coverage because of a lack of economic value. Landfilling was not considered to be a problem until the environmental impact of tailings drain and flood from tailings pond caused fatalities [67]. As a result, strict legislative norms have created pressure on the mining industry to investigate alternative disposal options.

The potential use of mine tailings as the raw material in alkali activation has attracted many researchers, and many laboratory scale studies have been performed. The most important parameter to study is the mineralogy of the mine tailings. Based on the mineralogy, most of the tailings show poor reactivity under alkaline condition. The utilization of mine tailings as a raw material in alkali activation is challenging because of low reactivity. The mine tailings can be used as an inert filler in alkali activation technology with a suitable co-binder, such as metakaolin, fly ash, or blast furnace slag. Curing conditions, curing time, alkali activator and its concentration, and co-binder materials all play critical roles in this approach, and the parameters need to be studied carefully for every system. Another option is to improve the reactivity of the initial material by using thermal treatment, mechanical activation, or alkaline fusion prior to alkali activation. If the reactivity is improved by a pretreatment method, the treated mine tailings can be used as a binder material in alkali activation. In all cases, the mechanical or thermal treatment does not improve the reactivity of the initial material which is why all systems need to be designed properly. Alkali activated mine tailings could be utilized as construction materials on mining sites and for road construction, or they could be recycled as paste backfill materials underground. In addition, the method could be also used prior to landfilling as a solidification and stabilization approach to dealing with hazardous mine tailings.

Valuable research needs to be conducted at the laboratory scale and in pilot testing to ensure environmentally friendly and the long-term stable properties of the final structure before the practical applications of these materials. The location of the mining sites can be challenging, and the recycling of mine tailings cannot be easily performed which need to be considered carefully. Since mine tailings are waste, and waste-based materials cannot be utilized in large-scale applications directly, there should be laws and policies to allow the use of tailings in construction industries. The authors recommend and emphasize the need for more research work in the area of mine tailings to bring confidence to policy makers and industrial partners regarding their application in construction materials.

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