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3 **Recycling Mine Tailings as Precursors for Cementitious Binders – Methods,**
4 **Challenges and Future Outlook**

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6 S. Maruthupandian, A. Chaliasou, A. Kanellopoulos*

7 *University of Hertfordshire, School of Physics, Engineering and Computer Science, Centre for*
8 *Engineering Research, College Lane Campus, Hatfield, AL10 9AB, United Kingdom*

9 *Email: a.kanellopoulos@herts.ac.uk

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11
12 **Abstract**

13 Increase in demand for mineral commodities such as coal, copper, iron, aluminium, gold,
14 tungsten, zinc etc., has led to higher quantity of mineral waste produced such as solids,
15 crushed rocks, overburden soil and tailings. The fine-grained mineral waste left after removal
16 of valuable material from ore is called mine tailing and is one of the major wastes of the mining
17 processes. Mineral wastes from mines, quarries and excavations are typically rich in SiO₂,
18 Al₂O₃, CaO and Fe₂O₃. This chemical composition makes them very attractive candidates to
19 be used in the production of construction materials, as these oxides are also the main
20 constituents of cement as well as of key alkali-activated binders. This contribution aims to
21 provide a comprehensive overview of the nature of mine tailings, the current state-of-the-art
22 in their utilisation in cementitious binders and the future potential. A rational summary of
23 limitations associated with use of mine tailing in cementitious binder due to its low reactivity
24 and potential solutions to overcome it is also provided. The study concludes with how the use
25 of mine tailings in cementitious binder could benefit in achieving the global sustainability goals.

26
27 **Highlights**

- 28 ○ Up to 2060 rapid growth is expected for construction materials.
- 29 ○ Decarbonisation policies will lead in the reduction of volumes of traditional SCMs.
- 30 ○ Tailings' chemical profile yield them suitable alternative cementitious materials.
- 31 ○ Systematic round-robin studies are required to explore tailings' potential

32
33 **Keywords:**

34 Mine tailings, mineral waste, alkali activation, thermal activation, mechanical activation,
35 concrete technology

36
37 **Nomenclature**

38 OECD - Organisation for Economic Co-operation and Development

39 SCM – Supplementary cementitious material

40 PC – Portland cement

41 OPC – Ordinary Portland cement

42 GGBFS – Ground granulated blast furnace slag

43 TEM – Transmission electron microscope

44 SEM - Scanning electron microscope

45 SDG – Sustainable Development Goal

46 XRD - X-Ray Diffraction

47

48 **1. Introduction**

49 The use of natural resources has been the basis of growth in civil infrastructure for at least
50 100 years. At the same time, the exploitation of natural resources and the pollution associated
51 with their extraction and processing have been already stretching the limits of our natural
52 habitat, putting significant pressure on the environment and human well-being. According to
53 the United Nations, the world's urban population has quadrupled since the 1950s, reaching 4
54 billion today, and with an estimated 7 billion people living in cities by 2050 [1]. To sustain this
55 rapid urbanisation rate, billions of tonnes of construction materials are required annually.

56

57 All evidence and projections suggest that civil infrastructure growth will keep expanding to
58 meet the needs of modern urban societies, including essential development for poverty
59 reduction and provision of fundamental infrastructure to the billions currently living in deprived
60 areas, for the decades to come. A recent report from the Organisation for Economic Co-
61 operation and Development (OECD) projects a rapid growth in the production and use of
62 construction materials in the next 40 years [2]. This translates into a massive demand for
63 pristine natural raw materials. Such demand will have a detrimental effect on the environment,
64 depleting natural resources and causing large scale destruction of landscapes [3].

65

66 The manufacture of construction materials is a process that utilises enormous amounts of raw
67 materials and energy, due to the sheer scale of the industrial processes required. Concrete is
68 the dominant construction material and the key element in most infrastructure assets.
69 However, concrete manufacture on a gigatonne scale per annum imposes extremely high
70 energy and resource demands: with >4 billion tonnes of cement being produced annually,
71 accounting for ~8% of global anthropogenic CO₂ emissions, and yielding an annual production
72 of ~2 tonnes of concrete for every person on the planet [4]. In Europe, the construction sector
73 alone is responsible for 36% of CO₂ emissions and for the 40% of all energy consumption [5].
74 Cement manufacture is a major contributor with the whole clinkerisation process consuming
75 ~3.5 GJ/ton [6].

76

77 At the same time, with increase in demand for mineral commodities such as coal, copper, iron,
78 aluminium, gold, tungsten, zinc etc., the mining of ores has increased while low grade ores
79 have started being explored [7]. The lower the quality of the ore, the higher is the quantity of
80 waste produced such as solids, crushed rocks, overburden soil and tailings. The fine-grained
81 mineral waste left after removal of valuable material from ore is called mine tailing and is one
82 of the major wastes of the mining processes.

83

84 Mine tailings can be generally disposed by either direct or indirect methods. The former
85 involves disposal of tailings in rivers/seas, while the latter encompasses disposal of tailings in
86 form of slurry (25-30% solid) into a cell, confinement or dam [8]. Mine tailing management is
87 a crucial issue as the physicochemical characteristics of the wastes have severe impact on
88 the soil and ground water including acid mine drainage [9]. The tailings are mainly composed
89 of oxides of silicon, aluminium, calcium, magnesium, and iron. This composition is similar to
90 that of widely used building materials such as cement, fine and coarse aggregates and
91 supplementary cementitious materials. This, in addition to their vast quantities, gives scope
92 for exploring the use of mine tailings in the construction industry.

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94

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2. The Challenge for Civil Infrastructure

Despite their heavy carbon footprint, shifting away from conventional cements is difficult. Concrete and the related cement-based materials are the construction industry's favourites for a variety of reasons including: (i) ease and low cost of construction compared to alternatives (e.g., steel); (ii) robustness under a variety of exposure scenarios; (iii) ability to construct a large variety of complex geometries; (iv) excellent mechanical performance, and (v) the almost global abundance of raw materials to produce them. Limestone and argillaceous materials (silica, alumina, iron) are extracted from the earth's crust, and through an intensive thermomechanical process converted to clinker, which is ground to be sold as cement. Then potable water and good quality natural aggregates are added to produce concrete. To satisfy this vast demand for concrete production globally, approximately 27 billion tonnes of natural aggregates and nearly 3 billion tonnes of fresh water are required [10]. The environmental impact with respect to depletion of natural resources is enormous. The International Energy Agency in its recently published roadmap for a sustainable transition in the cement and concrete industries has strongly highlighted two important elements: (i) the need to switch to alternative raw materials by 2025 and (ii) to switch to alternative cementitious binders by 2035 [11]. This strategy is also promoted by the European Cement Association in its latest report on decarbonisation of cement [12].

Reducing the environmental impact of cement-based materials is the focus of research the last 15-20 years. At the forefront of these attempts is the use of supplementary cementitious materials (SCMs). These are typically rich in silica and are soluble siliceous, alumino-siliceous, or calcium-alumino-siliceous fine powders that can be used as partial clinker replacement in cements or as partial replacement of Portland cement (PC) in concrete mixtures. SCMs are by-products of industrial operations (e.g., coal fired power plants; steel industry; ferrosilicon industry) and their chemical structure and mineral nature makes them compatible with the cementitious matrix. The three-best known SCMs are fly ash, ground granulated blast furnace slag, and silica fume. These materials do not directly reduce the environmental impact of clinker, but they rather reduce the need for it. In fact, recent studies show strong indications that the use of such minerals to reduce the clinker demand is the most viable option for the industry [13]. However, several problems that are summarised below are associated with the existing SCMs:

(i) Even though SCMs currently account for about 20% of cementitious materials within the UK cement and concrete industries [14], the global reduction in emissions as a result of SCMs use is not more than about 10% [11]. This signals that more amounts are needed and more efficient material use is necessary.

(ii) The existing SCMs rely heavily on specific industrial waste mainstreams which are currently either under major revision or threat. Coal combustion processes which lead to the production of fly ash are expected to considerably reduce to meet the global demand for lower CO₂ emissions, and biomass combustion or co-combustion products are not as suitable for use in most concretes. In the UK it is planned to retire all coal-fired power plants by 2025 and in the Netherlands by 2030 [15]. It is apparent that the global production of fly ash is expected to decline significantly.

(iii) The existing SCMs are not as abundant as natural raw resources. In addition, they are not readily available in large quantities around the world. This results in either excessive use of

144 cement (in absence of SCMs) or the generation of more CO₂ emissions associated with
145 transportation of such compounds in locations where they are not available.

146
147 On the other hand, billions of tones of mineral wastes (from mines, quarries, and excavations)
148 are produced globally every year. For mine tailings alone this quantity is estimated to be in the
149 range of 5 to 7 billion tonnes [16]. Recent studies suggest that the stock of mine tailings
150 generated in China only since 2013 has reached almost 15 billion tonnes [17]. The disposal
151 of such waste is a major environmental challenge and liability, especially in the light of some
152 recent disasters involving the collapse of mineral waste dams leading to considerable loss of
153 life and ecological damage [18–20]. In addition to such catastrophic events, stockpiling such
154 vast amounts of wastes can significantly alter the natural habitat and the biodiversity of the
155 surrounding areas [21].

156
157 Mineral wastes from mines, quarries and excavations are typically rich in SiO₂, Al₂O₃, CaO
158 and Fe₂O₃. This chemical composition makes them very attractive candidates to be used in
159 the production of construction materials, as these oxides are also the main constituents of
160 cement as well as of key alkali-activated binders. The last two years there is a growing interest
161 in utilising such wastes in the manufacture of cement-based composites, alkali-activated
162 binders and ceramics [16,22,23].

163
164 The use of mine wastes in cements can offer a natural solution to a major problem while at
165 the same time their own environmental impact is mitigated. Recycling such wastes in the
166 production of widely used materials is the most promising way to maintain our natural reserves
167 of raw materials and significantly reduce CO₂ emissions associated with the clinker (or other
168 binder) demand. In addition to the predictions from the International Energy Association and
169 the European Cement Association mentioned earlier, the World Business Council for
170 Sustainable Development also considers the recycling of certain mineral wastes as SCMs a
171 very promising solution towards a sustainable future [24]. The World Economic Forum and
172 OECD in their recent reports emphasise that recycling of mineral wastes will gradually become
173 more competitive than mining minerals and the global volumes for mineral waste recycling are
174 expected to grow considerably by 2060 [2,25]. Nonetheless, this process has limitations and
175 unknowns. The variability in waste streams calls for detailed and focused research on this
176 area. This is essential to thoroughly understand the different mineralogical and chemical
177 characteristics of these compounds and hence being able to identify optimum pathways for
178 their use and exploitation in construction materials manufacturing.

179
180 The present work discusses the physical, mineralogical, and chemical properties of different
181 mine tailings and the influence of the mineral composition on their behaviour. The activation
182 of mine tailings based on the mineral composition rather than a conventional chemical
183 composition is assessed in detail. It was observed from past studies that minerals of different
184 reactivity and crystalline structure had same chemical composition and behaved different
185 when used in cementitious binders [26,27].

186
187 The current contribution reviews past studies on the utilisation of mine tailings in cementitious
188 (and “cement-like”) matrices. This includes studies that used mine tailings as SCMs, as a raw
189 material for clinker, as direct cement replacement (with or without activation) and alkali
190 activated binders. For this purpose, scientific articles were collected by combination keyword
191 searches on data bases such as the Web of Science, Google Scholar, Scopus. Mine tailings,

192 mine waste, mine sludge, mineral waste, thermal activation, mechanical activation, alkali
 193 activation, geopolymerisation and valorisation of mine tailings were used as key words
 194 selected based on the abstract and title. Screening of published works was done to avoid
 195 repetition and duplication. Though the papers on masonry units and other civil engineering
 196 applications (e.g., the use of wastes as aggregate replacement) are reported only the
 197 activation process and the properties of the mine tailings were considered from those studies
 198 as anything else was beyond the scope of this paper. The studies confining to cementitious
 199 binders ranging only from 2000 to 2021 have been taken into consideration for the review.
 200 Figure 1 below summarises the steps followed in this analysis.
 201
 202



203
 204 **Fig. 1:** Summary of steps followed to obtain the data discussed in this study.

205
 206 **3. Mine Wastes**

207 **3.1 Sources**

208 Mining is the term used to describe the process of excavation of earth’s crust to extract
 209 minerals and ores. Valuable components or metals are extracted from the minerals and ores
 210 by different mineral separation processes. At the end of the mineral separation process, a
 211 valuable component, which is a concentrate of base metal or mineral, is extracted. As part of
 212 this process, the mineral separation process residues (tailings) are generated [28]. Tailings
 213 do not bear any significant monetary value and occur in form of sludge with a varying water
 214 content ranging from 18 to 32% [29–32]. The quantity of tailings generated varies, depending
 215 on several factors: (i) the type of material extracted; (ii) the process followed and (iii) the quality
 216 of ore. Ince et al [33] reported that for low quality ores in copper extraction about 128 tons of
 217 tailings are generated for the production of 1 ton of copper. In the contrary, high-quality ores
 218 can generate 1.5 tons of tailings for every ton of copper produced [34]. Nonetheless, even the
 219 optimum 1 to 1.5 ratio will lead to the production of immense volumes of waste material that
 220 currently remains unused affecting the environment and local communities.

221
 222 **3.2 Physical, Chemical, Mineralogical properties**

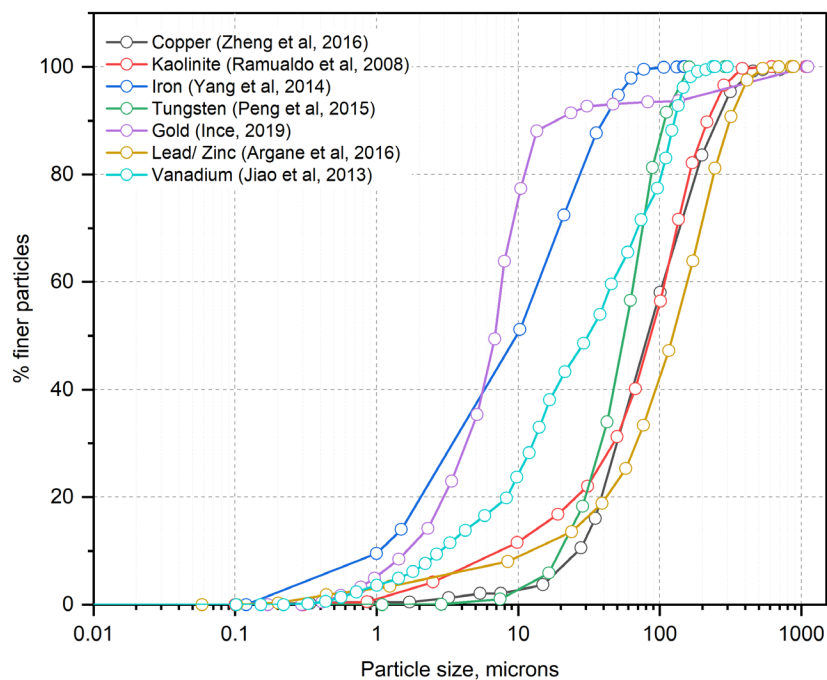
223 **3.2.1 Physical Properties**

224 Mine tailings are crystalline, relatively loose and porous in microstructure [35]. The specific
 225 gravity of such compounds ranges between 2.70 – 4.29, depending on the type of mine and
 226 its composition [36–38]. Mine tailings are reported to have a high-water absorption, up to
 227 7.15% [39]. This can raise concerns if the tailings are intended for use in concrete as

228 aggregates. However, such porous materials can have alternative uses. Recent studies have
229 shown that a percentage of highly porous aggregates can facilitate internal curing processes
230 [40,41]. In addition, it has been demonstrated that porous aggregates can be used as carriers
231 for healants in promoting self-healing processes in cementitious matrices [42,43].
232

233 The pH of tailings is generally in near neutral or slightly alkaline region ranging between 8.0 –
234 9.3 for carbonate rich mine tailings [31] and 6.7 – 10.0 for silicon rich mines [30,44,45]. Though
235 the pH of mine tailings is generally alkaline, the chemical and mineral composition of a mine
236 tailing determines the possibility of acid mine drainage. If a mine tailing has high amounts of
237 sulphide minerals and low amounts of minerals such as carbonates, which will neutralize them,
238 their oxidation in natural conditions may also lead to acid mine drainage [46]. If the mine
239 drainage is neutralized, neutral effluents generated may consist of toxic metals and metalloids,
240 such as Ni, Zn, Co, As, and Sb [47]. This makes the effective and efficient recycling and reuse
241 of such wastes a major environmental concern.
242

243 The particle size of tailings varies amongst cement sized, silt sized and sand sized particles.
244 Argane et al., [46] reported a silt sized tailing of D_{50} 210 -270 μm whereas Bussiere [28]
245 reported a cement sized mine tailing of D_{50} 17 μm . The particle size distribution of mine tailing
246 observed in various studies are as given in Fig. 2.



247
248 **Fig. 2:** Particle size distribution of various mine tailings.
249

250 The variation in the size enabled various attempts on recycling the mine wastes as fine
251 aggregates [48,49] and as a binder component [39,50,51]. However, the feasibility of using
252 mine tailings as aggregate or cement mainly depends on its chemical composition, rate of
253 dissolution and its ability to produce hydration products like that of cement.
254

255 3.2.2 Chemical and Mineralogical Properties

256 The composition of mine tailings varies highly due to the variation in source mineral, process
257 of extraction, mineral extracted and quality of the ore. Table 1 shows the chemical composition
258 of various mine tailings discussed across several studies. It can be seen from the table that

259 bauxite, coal, graphite, gold, iron, kaolinite, and lead-zinc tailings are predominantly composed
260 of silicate. Phosphate, tungsten, and zinc tailings have a considerable amount of CaO, whilst
261 bauxite, coal and kaolin tailings have a considerable amount of Al₂O₃. The ratio of silica to
262 alumina (SiO₂/Al₂O₃) of tailings ranges from 0.86 – 9.12, modulus of hydraulicity [52] ranges
263 between 0.02 – 3.98 and the modulus of basicity are between 0.03 – 5.28. Shi et al [53]
264 recommended a basicity modulus range between 0.7 to 1.2 for a material to be suitable for
265 alkali activation. However, these conventional indicators of hydraulicity or geopolymerisation
266 may not be suitable to determine the reactivity of the mine tailings, as the mine tailings are
267 crystalline in nature [48] and aluminates and silicates present are not always available in a
268 form suitable for dissolution that will yield them suitable for participation in chemical reactions.
269 Therefore, the mineralogical composition of the material plays a significant role in determining
270 its reactivity and selection of appropriate and effective treatments can only be done when the
271 fraction of reactive components in a tailing are identified [54].

272
273 Table 2 provides an outlook of various minerals present in different mine tailings. Quartz,
274 Kaolinite, Calcite, Muscovite are some of the minerals most widely found in the tailings.
275 Similarly, many phyllosilicate, alumino silicate, calcium alumino silicate and carbonate minerals
276 were also present. It can be seen that SiO₂ is found in non-reactive quartz form [55,56] or
277 reactive cristobalite form [57,58]. Similarly, TiO₂ occurs as low reactive Rutile [26,59] and in
278 the form of anatase which is more reactive [60]. Minerals may also occur as a member of a
279 series such as feldspar, biotite, phyllosilicates, iron oxides, alumina silicates, with different
280 crystallinity and reactivity [61,62]. Hence, conversion of non-reactive phases to reactive
281 phases and making them more amorphous is important for exploring the potential of mine
282 tailing as binders.

283
284

Table 1: Chemical composition of a variety of mine tailings reported in the literature.

Mine type Chemical Composition	Bauxite [63]	Boron [55]	Coal [64]	Copper [65]	Gold [39]	Iron [66]	Kaolin [29]	Graphite [49]	Phosp hate [67]	Quartz [68]	Tungsten [51]	Vanadium [69]	Zinc [70]	Copper & Zinc [71]	Lead & Zinc[72]	
															Silicate	Carbo nates
CaO (%)	3.15	14.36	1.06 -8.41	8.14	14.89	6.20	0.2	15.547	34.2	0.51	28.02	6.51	33.66	0.84	1.99	19.83
SiO ₂ (%)	32.24	19.57	52.34 - 61.92	44.54	44.62	24.40	59.5	62.50	22.8	79.53	36.52	61.92	6.20	25.16	68.44	7.16
Al ₂ O ₃ (%)	37.39	6.17	19.50 - 36.34	16.18	8.64	10.95	32.9	10.21	2.5	9.52	8.70	7.35	2.06	6.41	9.38	3.09
Fe ₂ O ₃ / TFe (%)	8.67	2.85	6.42 - 8.35	-	26.1	31.15	2.7	5.07	0.9	3.22	11.71	4.12	0.19	19.24	2.22	2.11
SO ₃ / S (%)	-	0.34	0.11 -0.65	5.14	2.31	-	0.1	-	-	-	0.16	7.14	1.46	26.18	1.12	7.51
MgO (%)	0.85	4.26	1.0 - 1.57	-	0.51	0.99	-	2.33	4.1	0.64	1.12	1.24	9.94	1.69	0.48	11.27
K ₂ O (%)	-	2.31	2.65 - 4.02	-	0.55	0.86	0.1	2.26	0.4	3.24	0.05	1.25	0.24	0.41	5.46	0.48
TiO ₂ (%)	2.31	-	0.85 - 1.21	-	0.41	0.42	2.0	-	-	0.52	0.12	0.46	0.09	0.09	-	-
Na ₂ O (%)	0.85	0.18	0.17 - 0.53	-	1.63	0.28	-	-	0.8	0.72	-	2.66	0.30	0.23	0.7	0.1
MnO (%)	-	-	0.06 - 0.14	-	0.09	-	-	-	-	0.16	2.56	-	-	0.07	-	-
Cl (%)	-	-	-	-	0.02	-	-	-	-	-	-	-	-	0.04	-	-
ZnO/ Zn (%)	-	-	27 - 46 ppm	0.65	0.11	-	-	-	-	78.82 mg/kg	0.02	-	1.77	0.31	230 ppm	260 ppm
PbO/ Pb (%)	-	-	-	-	-	-	-	-	-	195.90 mg/kg	-	-	0.15	0.14	4710 ppm	5940 ppm
Cu (%)	-	-	-	-	-	-	-	-	-	75.24 mg/kg	-	-	-	0.19	-	-
Cr ₂ O ₃ / Cr (%)	-	-	-	-	-	-	-	-	-	-	0.03	-	-	0.02	-	-
WO ₃ (%)	-	-	-	-	-	-	-	-	-	-	0.03	-	-	-	-	-
V ₂ O ₅ / V (%)	-	-	-	-	-	-	-	0.308	-	-	0.02	0.42	-	-	50.30 ppm	-
SrO/ Sr (%)	-	0.51	-	-	-	-	-	-	-	-	-	-	-	21.40 ppm	1.12	7.51
BaO/ Ba (%)	-	-	-	-	-	-	-	-	-	-	-	-	0.31	0.008	-	-
P ₂ O ₅ (%)	-	-	-	-	-	-	0.4	-	-	-	-	-	0.27	0.04	-	-
ZrO ₂ / Zr (%)	-	-	-	-	-	-	0.1	-	-	-	-	-	-	0.003	-	-
LOI (%)	13.74	21.06	1.81 - 5.61	18.92	-	6.95	2.0	0.65	19	2.46	2.30	6.93	43.35	18.68	-	-
SiO ₂ /Al ₂ O ₃	0.86	3.17	1.70-2.68	2.75	5.16	2.23	1.81	6.12	9.12	-	4.20	8.42	3.01	3.93	7.30	2.32
Basicity Modulus $\frac{CaO+MgO}{SiO_2+Al_2O_3}$	0.06	0.72	0.03-0.10	0.13	0.29	0.20	-	0.25	1.51	-	0.64	0.11	5.28	0.08	0.03	3.03
Hydraulicity Modulus $\frac{CaO}{Fe_2O_3+SiO_2+Al_2O_3}$	0.04	0.50	0.01-0.08	0.13	0.19	0.09	0.002	0.20	1.31	0.49	0.09	3.98	0.02	0.02	1.60	0.04

*The chemical compositions reported in the table are indicative for a particular type of tailing observed in a study and does not represent a mean or median.

** All values given in the table are in percentage unless otherwise specified.

289 **Table 2:** Mineral composition of a variety of mine tailings (Note: The percentage composition
290 of minerals is reported only in a few studies).

Sl. No	Mineral waste	Mineral	% Composition	Sl. No	Mineral waste	Mineral	% Composition		
1	Bauxite [63]	Kaolinite		7	Low sulphide Lead - Zinc [48] Carbonate dominant (80%)	Hematite	1.07		
		Diaspore				Other	4.03		
		Muscovite							
		Anatase							
		Corundum							
		Quartz							
		Mullite							
2	Boron [55]	Hydroboracite		8	Kaolin [73]	Kaolinite			
		Montmorillonite				Mica			
		Colemanite				Quartz			
		Illite							
		Quartz							
3	Coal [76,77]	Quartz	29 – 37	9	Phosphate [74] [57,75]	Quartz	0.9 – 12.44		
		Mica	25			Apatite	9.6		
		Calcite	15 – 17			Fluorapatite	34.09 - 44		
		Kaolinite	14 – 15			Calcite	11.15		
		Dolomite	5			Dolomite	9.45 – 89.5		
		Feldspar	2			Illite	9.46		
		Amorphous	17			Palygorskite	21		
		Hematite				Hematite	1.4		
4	Copper [65,78–80]	Albite		10	Tungsten [51,81]	Garnet	88.14		
		Gypsum				Amphibole	5.46		
		Sanidine				Chlorite	4.27		
		Quartz				Quartz	2.13		
		Augite				Muscovite			
		Dolomite		11	Vanadium [82]	Quartz			
		Phlogopite				Feldspar			
		Amphibole				Diopside			
		Clinochrysotile				12	Low sulphide Lead - Zinc [48] Silicate dominant (90% silicate)	Quartz	
		Calcite						Orthoclase	
		Talc		Chlorite					
		Clinocllore		Albite					
		Siderite		Barite					
		Hematite		13	Desulphurized pyrrhotite tailings [26,83]	Fluorite			
		Pyrite				Magnetite	2.56		
Mullite		Orthoclase	1.37						
Muscovite		Pentlandite	0.29						
		Pyrrhotite	0.64						
5	Gold [39,50,84]	Albite		14	High sulphide tailings Copper and Zinc [71,85]	Quartz	1.60		
		Gypsum				Rutile	0.30		
		Quartz				Actinolite	29.75		
		Dolomite				Albite	2.76		
		Muscovite				Arsenopyrite	0.01		
		Microcline				Calcite	2.17		
		Pyrite				Chalco Pyrite	0.25		
		Sodium aluminium silicate							
		Sanidine				Clinocllore	26.48		
		Mica				Lizardite	31.81		
6	Graphite [59]	Montmorillonite		14	High sulphide tailings Copper and Zinc [71,85]	Pyrite FeS ₂			
		Quartz	43.15			Anhydrite CaSO ₄			
		K-Feldspar	12.42			Caldecahydrate CaAl ₂ O ₄ 10.H ₂ O			
		Albite	5.08			Quartz SiO ₂			
		Anorthite	3.31			Smithstone			
		Biotite	8.34			Diopside			
		Muscovite	4.82			Calcite			
		Epidote	2.05			Albite			
		Chlorite	3.13			Sodium copper sulfide			
Rutile	0.72								

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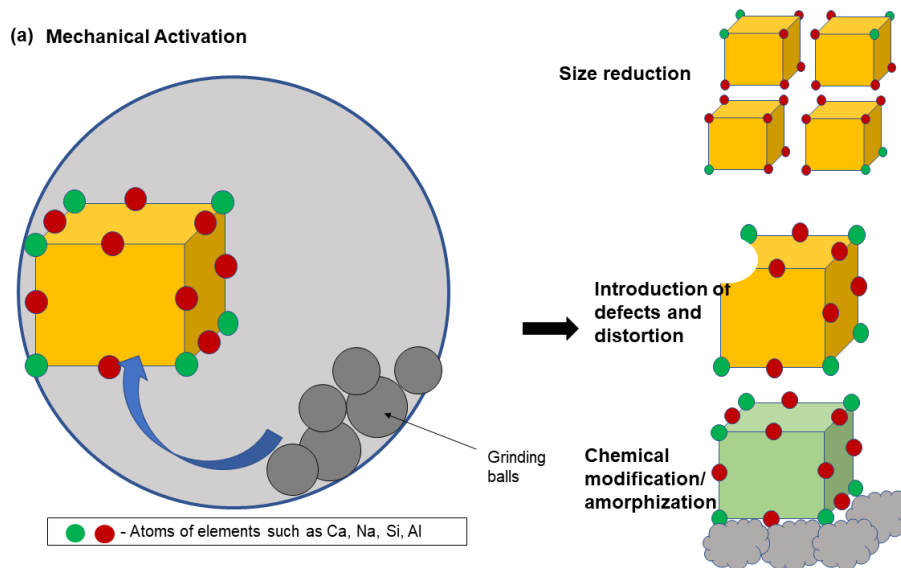
3.3 Activation Processes

293

The structural stability of any material is associated with its chemical reactivity. Materials with more defects in their microstructure, more distortion in their lattices, or materials existing in an

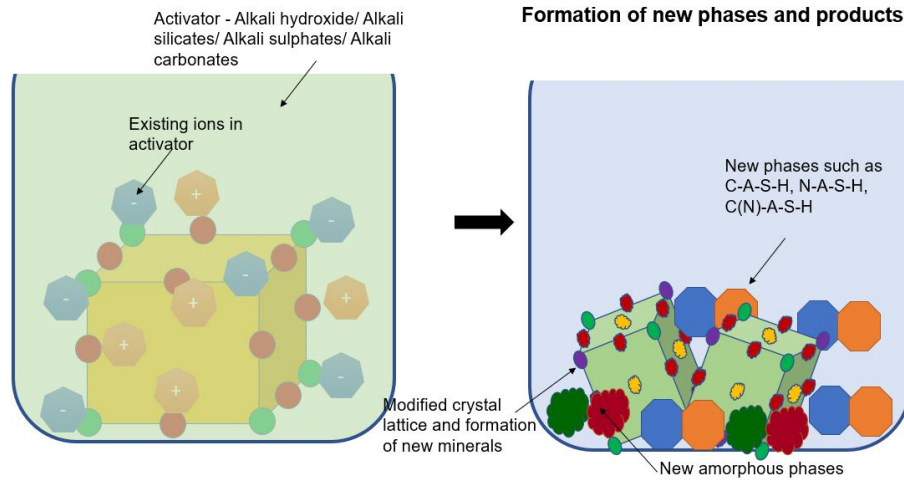
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295 amorphous state are considered to have less stability and tend to have a high reaction activity
 296 [86]. Since mineral wastes, and hence tailings, are highly crystalline their processing is
 297 essential to render them more reactive for use in cementitious systems as part of the binder
 298 phase. This processing is commonly known as “activation”. Activation processes can be either
 299 mechanical, chemical, thermal or a combination of them (see Fig. 3). For use in cementitious
 300 matrices, the mine tailings can be alumino-silicate, calcium-alumino-silicate or predominantly
 301 silicate. The energy required in the processing of tailings should be of sufficient magnitude to
 302 rearrange the crystalline lattice. For example, in case of an alumino-silicate mineral if the
 303 binding energies of the Al-O-Si, Si-O-Si or other structural forms present in the raw material
 304 are strong, energy higher than binding energy may be required to disrupt the bonds in
 305 crystalline lattice and make ions available for further reaction [87]. The thermal activation is
 306 achieved by calcination of the material in a high temperature furnace. The mechanical
 307 activation can be performed by means of ball mill, ring mill, planetary ball mill, vibratory ball
 308 mill, stirred ball mill or jet mill [88,89]. The chemical activation is achieved by use of an activator
 309 to induce chemical reactions for release of Si, Al and other ions and enabling further reactions.
 310 When alkalis are used for this purpose, the process is known as alkali activation [90]. The
 311 following sections discuss the various activation processes found in the literature. Table A1 in
 312 the Appendix provides detailed information about the application of activation processes for
 313 individual minerals as reported in published research.



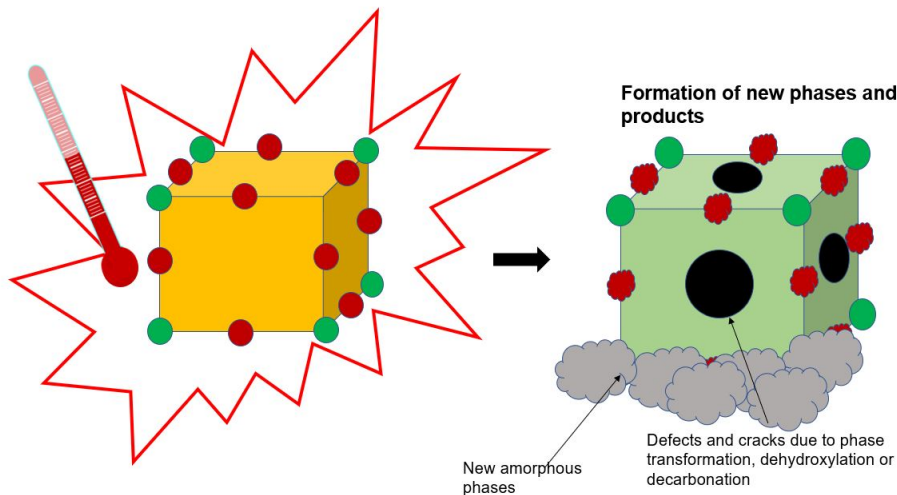
314

(b) Chemical Activation



315

(c) Thermal Activation



316

317 **Fig.3:** Schematic representation of the mineral activation processes: (a) Mechanical; (b) Chemical and
318 (c) Thermal activation.

319

320 3.3.1 Calcination or Thermal treatment

321 As the formation of crystalline phases in the minerals is a function of temperature, calcination
322 is known to impart changes in the crystal structure of the minerals. The general modifications
323 which happen during the calcination are:

324

325 (i) **dehydroxylation** of hydroxides in minerals such as kaolinite at about 580 °C and mica at
326 about 700 °C [73]

327

328 and (ii) **decarbonation** in carbonate minerals such as calcite, dolomite, siderite smithsonite
329 etc [48,91]

330

331 The dehydroxylation of phyllosilicate minerals improves the reactivity as the loss of hydroxides
332 generates more disorder and exposure of Al ions favouring their dissolution. [92]. Therefore,
333 this method of activation has been found suitable for mine waste containing a large amount of

334 phyllosilicate or carbonate minerals. However, minerals such as albite though they exhibit
335 dehydroxylation at a higher temperature do not exhibit any change in crystal structure [93].
336 The calcination also involves alpha – beta transformation of quartz. Such transformations are
337 generally accompanied by increase in volume and hence lead to formation of cracks thus
338 further improving the reactivity by exposing new surfaces available for reaction [94]. Past
339 studies show that minerals such as albite [93], apatite [54], arsenopyrite [95], calcite [64],
340 chlorite [96], clinocllore [97,98], corundum [99], diaspore [63], dolomite [96], feldspar [100],
341 hematite [101,102], hydroboracite [55], montmorillonite [92], talc [103], undergo modifications
342 when calcined at temperatures between 540 °C and 1450 °C. Therefore, thermal treatment
343 shall be effective for mine tailings with these minerals as major phases.

344

345 The choice of treatment temperature also depends on the minerals present in the mine tailings
346 in their raw form. Torgal and Jalali reported that calcination of mine tailings at 750 °C had no
347 effect on the compressive strength of the cementitious matrix containing them. However,
348 further increase of the calcination temperature to 950 °C resulted in an increase in the
349 compressive strength values due to formation of amorphous products [104]. Beyond 950 °C,
350 thermal treatment resulted in the formation of crystalline phases crystalline phases, such as
351 mullite, which reduced the reactivity and hence did not yield an improvement in the measured
352 mechanical properties.

353

354 Calcination in presence of alkali or alkaline roasting or thermo chemical activation

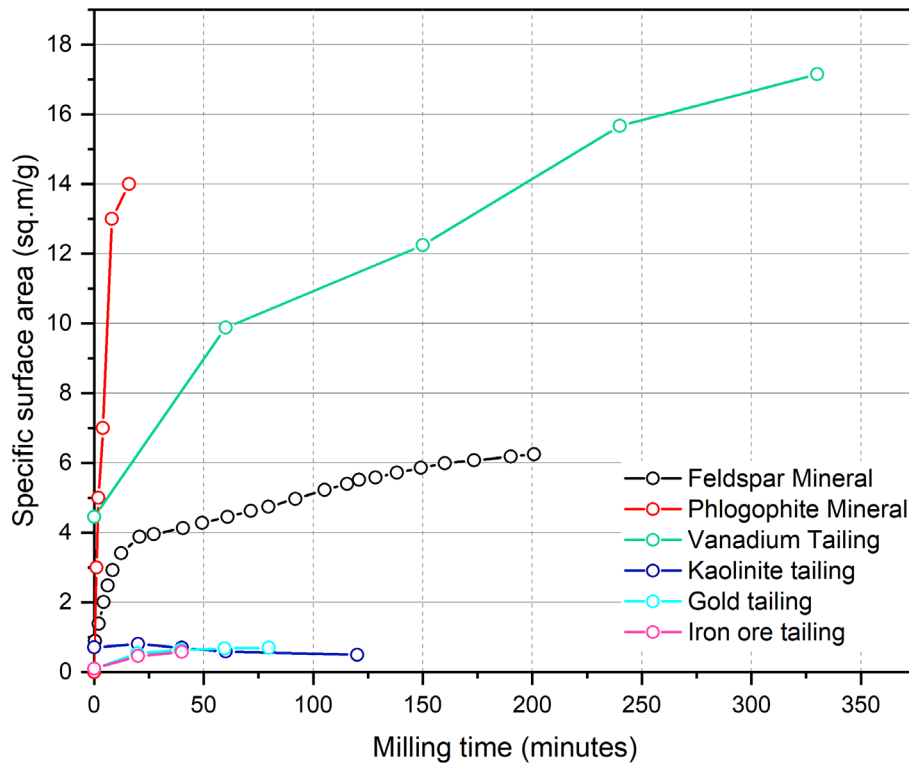
355 Calcination of tungsten mine waste in presence of sodium carbonate yielded a maximum
356 strength at a calcination temperature of 500 °C instead of 950 °C [104]. Minerals such as albite
357 when calcined in presence of NaOH or Na₂CO₃ they completely shift to an amorphous state
358 due to loss of crystalline ordering of Al and Si [93]. Similarly, calcination of mine tailing in
359 presence of alkalis at 600 °C indicated decrease in crystalline silica and albite and formation
360 of biotite. Also, it is important to take into account the disintegration temperature of activator
361 during alkaline roasting, or else the disintegrated compounds may lead to leaching or
362 formation of efflorescence during hydration [104]. Kaze et al [105] in their study reported that
363 the presence of high amounts of iron (almost 50%) led to a structural disorder in clay minerals
364 where Al ions are replaced by Fe ions. These inherent defects result in reduction in crystallinity
365 and requires lower thermal activation energy.

366

367 3.3.2 Mechanical and Mechano-chemical Activation

368 Mechanical activation utilises mechanical actions to induce changes on the microstructure of
369 minerals. Mechanical processing essentially decreases the particle size and increases the
370 surface area of the materials. This in turn improves the reactivity [50]. The most commonly
371 used mechanical activation is grinding which involves a combination of pressure and shearing
372 actions on the material. Research indicates that grinding improves reactivity of tailings by
373 dynamic creation of new surfaces, by creating defects in crystal lattice and phase
374 transformations. [50,69,86,89]. It was also reported that during grinding due to friction and
375 impact of the grinding media on the material a localised increase in temperature may be
376 observed in a small area leading to mechanically activated chemical reactions [89]. The
377 electron energy at such localised contact points is about 10 eV which is higher than energy of
378 thermal treatment at 1000°C (4 eV) [88]. The variation in specific surface area of different
379 tailings with respect to grinding time is given in Fig. 4. A phenomenon called selective grinding
380 occurs when minerals of high and low hardness are present in the same material and the
381 minerals with higher hardness act as a grinding media for minerals of lower hardness and

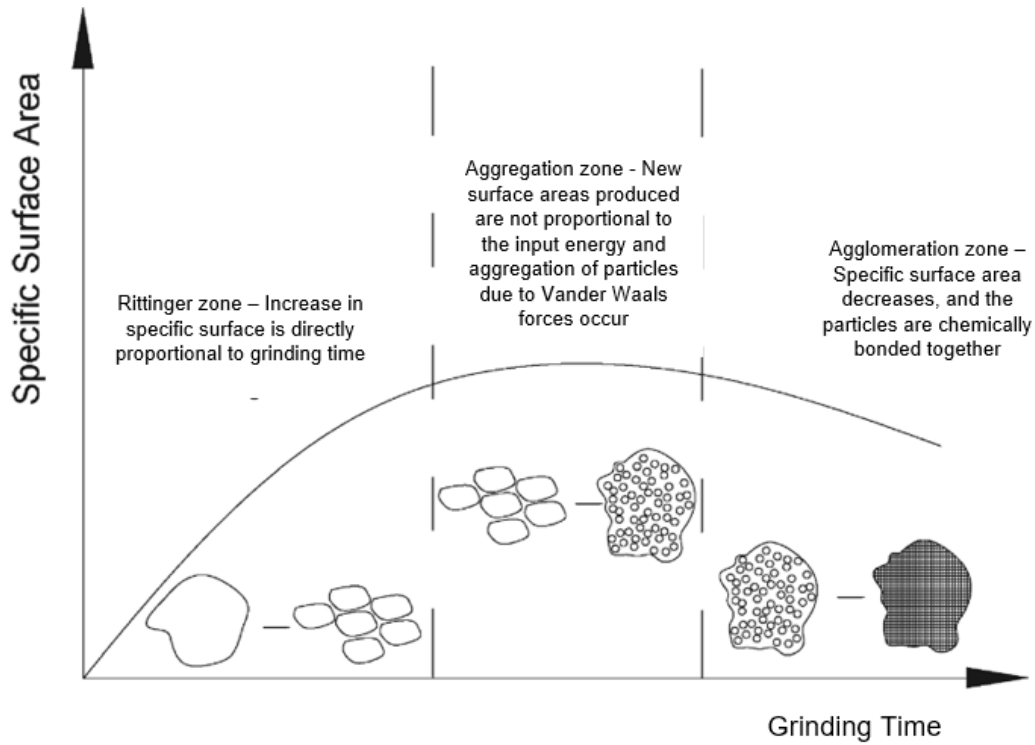
382 accelerate the grinding process [51,89]. Minerals of high hardness such as garnet and quartz
 383 have a particle size larger than other minerals after grinding of the mine waste. A bimodal
 384 particle size distribution curve after grinding represents the presence of minerals with different
 385 relative grind ability present in the same material [51]. This property of different minerals being
 386 downsized by different extent for the same amount of grinding energy applied is called grinding
 387 performance or grindability. And better grinding performance is crucial for economic and
 388 environmental efficiency [106].
 389



390
 391

392 **Fig. 4:** The specific surface area of tailings with respect to grinding time [50,69,107–110].
 393

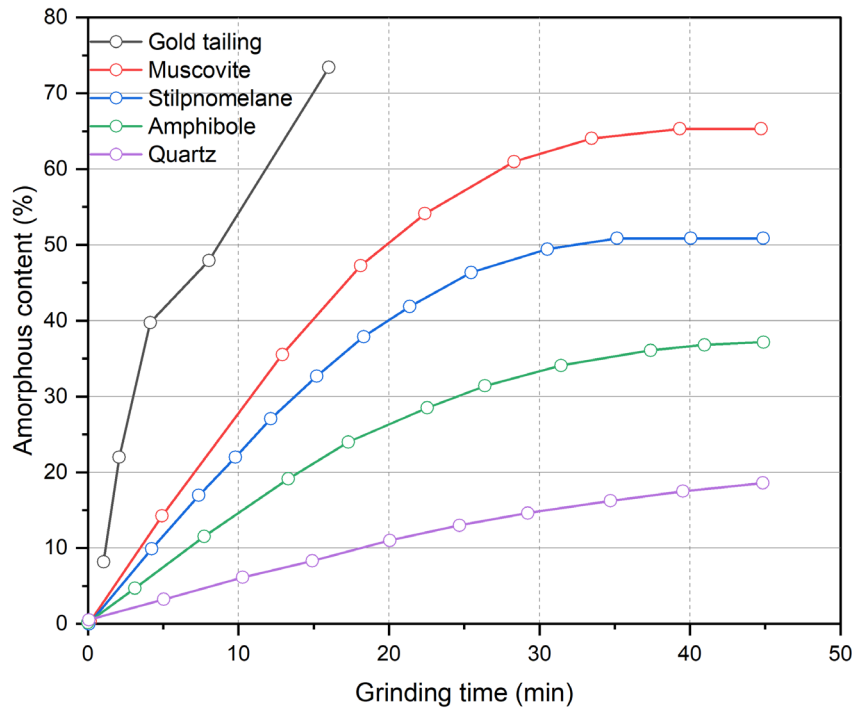
394 In certain cases, grinding also breaks the Al-O-Si, Si-O-Si bonds further improving the
 395 reactivity of minerals. A recent study has shown that the content of reactive Si and Al increased
 396 from 300 to 1500 mg/ L and from 100 to 315 mg/ L respectively after 5.5 hours of grinding
 397 [69]. Similarly, Yu et al. [35] reported an increase in reactive Si and Al content by 26.03% and
 398 96.33% respectively, after 4 hours of grinding. Grinding beyond 3 h leads to increased surface
 399 energy which in turn results in agglomeration of particles. Therefore, an increase in the
 400 particle size is observed after 3 h of grinding in case of copper tailings and 4 hours grinding in
 401 case of iron tailings [35,111]. This phenomenon was explained by Tole et al [89] as given in
 402 Fig 5.



403
 404 **Fig. 5:** Reaction evolution of particles during mechanochemical activation. Image reproduced from [89].
 405

406 The XRD analysis showed that intensity of quartz, endenite, albite, hematite and feldspar
 407 peaks were significantly decreased after 4-5 hours of grinding [35,111]. After melting,
 408 quenching and grinding the reactivity of phlogopite increased from 1% to 40% [54]. Yao et al
 409 [50] reported that the relative crystallinity of muscovite reduced to 37% whereas the
 410 crystallinity of quartz reduced only to 80% after 80 minutes of grinding. Clinocllore exhibited
 411 a complete collapse of mineral structure after 16 minutes of milling [112]. The increase in
 412 amorphous content of different mine tailings and minerals with grinding is given in Fig. 6 while
 413 in the literature researchers have also used TEM (see Fig. 7) to confirm the formation of
 414 amorphous phases pre- and post- mechanical treatment.

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Fig. 6: Amorphous content of different tailings and minerals with respect to grinding time

421
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424
425

Fig. 7: TEM images of kaolinite-talc mixture: (a) before and (b) after grinding. The well-developed pseudo hexagonal crystals were converted to shapeless amorphous materials after 60 minutes of grinding. Image reproduced from [108] with permission from Elsevier.

426 3.3.3 Chemical Activation

427 Alkali activation of mine tailings refers to the use of alkalis to stimulate the potential activity by
428 dissolution. It is not to be confused though with the term “alkali activation” that refers to
429 alternative to cement binders, in some cases known as “geopolymers”. In both cases, the
430 alkalis used for this purpose are known as activators. In tailings treatment, the most widely
431 used activators are MOH, M_2SiO_3 , M_2SO_4 and M_2CO_3 where M refers to either sodium or
432 potassium [53]. The dissolution of silica and alumina from the source material is generally
433 dependant on: (i) the strength Si-O-Al bonds; (ii) the formation of soluble species from the
434 source; (iii) the pH of the solvent medium (i.e. activators) and (iv) the temperature of the
435 system in which reaction takes place [16,113]. Studies showed that the time and rate of
436 release of Al and Si ions play a crucial role in strength gain. Though the rate of dissolution of
437 Si can be increased with increased alkali concentrations, the concentration of Si, Al, Ca in the
438 binder, the glass content and the fineness of the precursor, played the most important role in
439 strength gain [16,113]. Most studies therefore used alkali activation in combination with other
440 activation methods such as grinding and calcination to attain the optimum conditions
441 [29,55,112].

442
443 When it comes to geopolymerisation, it was found that grinding improved the rate of reaction
444 due to increase in the specific surface area [69]. Xu and Deventer [114] suggested that alkali
445 activation can be performed both on calcined and non-calcined materials, and calcined
446 materials had better reactivity due to their more amorphous nature. Their study also reported
447 that geopolymerisation of multiple minerals combined is possible, showing alkali activation can
448 be suitably applied for mine tailings. Torgal et al [115] observed that the compressive strength
449 increased with increase in molarity of the activator while past studies indicate that the better
450 strength was achieved when the SiO_2/Al_2O_3 ratio of the system (precursor + activator) ranged
451 between 3.0 – 3.8 [116]. Liew et al [117] found that the optimum SiO_2/Al_2O_3 , SiO_2/Na_2O ,
452 Al_2O_3/Na_2O and H_2O/Na_2O molar ratios were 3.10, 2.69, 0.87 and 14.23 respectively, for
453 calcined phyllosilicate minerals whereas Xu and Deventer [62] observed a higher compressive
454 strength for alkali feldspar minerals when the Si/Al ratio was maintained between 2.21 – 2.38.

455
456 In general, when the precursor’s Si/Al ratio value does not fall within the above-mentioned
457 range, an additional source of silica as sodium silicate activator and/or alumina are added.
458 The most commonly added materials are metakaolin, ground granulated blast furnace slag,
459 fly ash, milled glass waste, [67,70,71,118,119]. Kiventera et al [119] adopted Si/Al value less
460 than 3, between 1.5 - 2.5 and Na/Al ratio in the range of 1.0 – 1.29, to proportion a mixture of
461 fly ash, ground granulated blast furnace slag and gold mine tailing. The mix was found to have
462 better strength and microstructure when metakaolin was added and the Si/Al was 2.5 and
463 Na/Al was 1.0. Peng et al., [51] activated tailing rich in garnet and quartz, using $Ca(OH)_2$,
464 gypsum and sodium silicate as different activators and found that the reactivity of mine tailings
465 improved by 67.65%, 27.73% and 36.29 % respectively. Others have used NaOH in presence
466 of fly ash and metakaolin to activate calcined tailings comprising calcite, quartz, dolomite and
467 mullite [67]. When fly ash was used quartz, dolomite, calcite, mullite, hydro-sodalite, chabazite
468 were present in the activated paste whereas the use of metakaolin led to the formation of
469 quartz, dolomite, calcite, hydro-sodalite and sodium aluminium silicate [67]. The presence of
470 dolomite, calcite and quartz phases before and after activation indicated that they were
471 unaffected by alkali activation [67]. However, the interlocking of unreacted large particles
472 contributed to strength development after hardening [51]. Jiao et al. [82] activated ground
473 tailing using Na_2SiO_3 and found Si/Al = 3.3, Na/Al = 1.0 and H_2O/Na = 1.6 were optimum for

474 activation. The studies with both sodium and potassium activators indicated that a geopolymer
475 paste with highly reactive precursor when used with a low viscous activator such as KOH or
476 K_2SiO_3 exhibits cracks due to accelerated evaporation of water which otherwise would have
477 been available for geopolymerisation [87]. Koohestani et al [85] reported a formation of silica
478 gel when the pH of the system was 8.7. Evidence in the literature suggests that high calcium
479 content in the mine tailings can turn the tailing itself to an activator [30]. Furthermore, mine
480 tailings can be used as an inert filler in geopolymers in presence of a co-binder [47]. In case
481 of alkali activation of minerals rich in Fe [105,120], it was observed that Al^{3+} ions were replaced
482 by Fe^{3+} . This resulted in formation of Si-O-Fe linkages and hence Fe^{3+} binders are formed.
483 Peys et al [120], from their study reported that the synthesis of this phase is observed when
484 the Na/Fe^{3+} is almost equal to 1 and Si/Fe^{3+} is almost equal to 3. The study also reported that
485 the Fe^{2+} ions after dissolution is oxidised to Fe^{3+} during or after the synthesis of geopolymer.

486
487 The major advantage of alkali activation of mine tailing is that, unlike the other two methods
488 of activation, alkali activated mine tailings can be used as a standalone cement. Alkali
489 activation also: (i) helps in encapsulation of heavy metals and metalloids and thus prevents its
490 leaching; (ii) it has low energy consumption demand and (iii) has better acid resistance
491 [119,121]. Mine tailing alkali activation can be considered akin to engineering the same waste
492 to cater different needs by modifying the activators, e.g. tungsten mine waste when activated
493 with NaOH and $Ca(OH)_2$ exhibited optimum mechanical properties with shrinkage less than
494 OPC whereas when activated with Na_2CO_3 had high early age strength but performed poorly
495 under water [122]. However, the disadvantage in using mine tailings is the effect of presence
496 of Cu^{2+} , Fe^{2+} , Fe^{3+} in the formation of geopolymer chains, which is still not well established.
497 Also, the alkali activation may form hydration products which are unstable thus the binder
498 exhibits high early strength and decrease in strength at later ages [104].

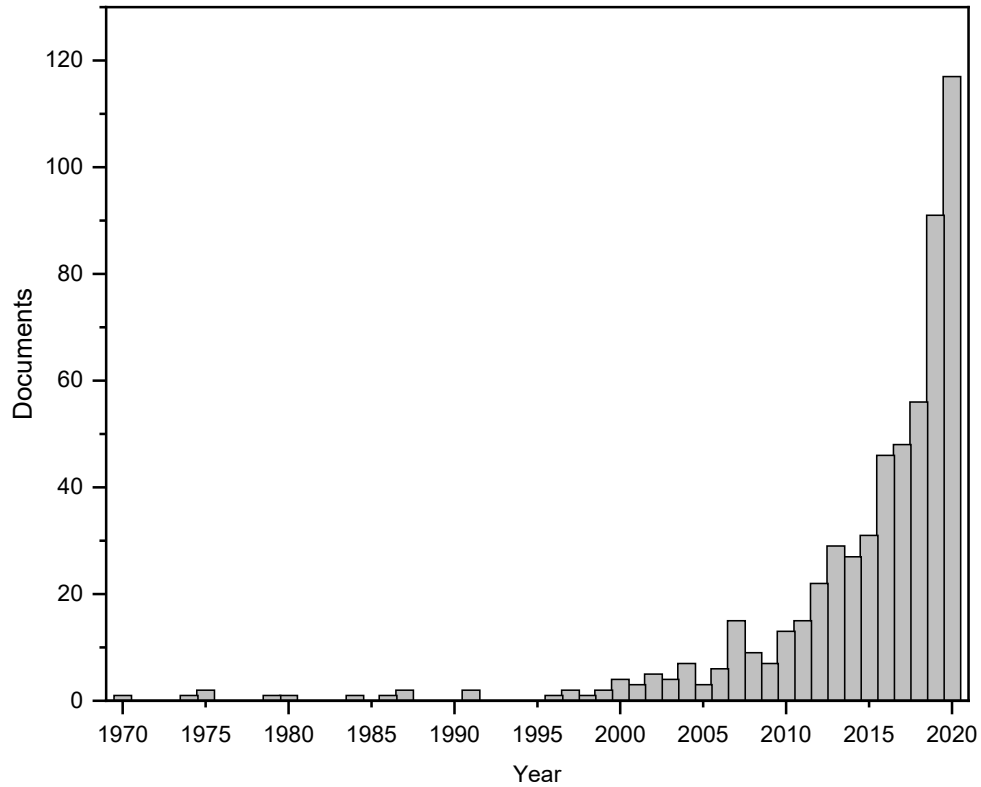
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500 **4. Mine Tailings in Cementitious Binders**

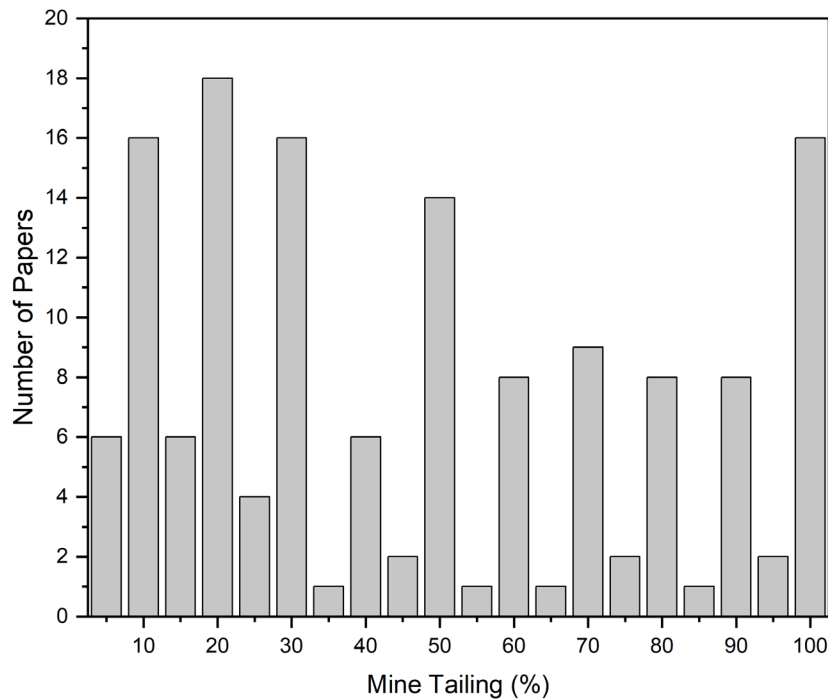
501 The use of mine tailings in construction related applications gradually gains a lot of interest.
502 These applications range from backfilling to the use of tailings as part of a binder. The growing
503 interest in the area is reflected by the continuously growing number of studies published (Fig.
504 8). The utilisation of tailings as part of the binder in particular has significantly increased the
505 last decade (between years 2012-2021) with a growing number of studies reporting various
506 percentages incorporated (Fig. 9). It is interesting to note that a very wide spectrum of %
507 replacement is covered ranging from as low as 5% to as much as 100%.

508

509



510
 511 **Fig. 8:** Number of studies discussing the use of mine tailings in construction related applications (data
 512 obtained by Scopus).
 513



514
 515 **Fig. 9:** Number of studies reporting the utilisation of various % of mine tailings in cementitious binder.
 516

517 **4.1 Mineralogy and Microstructure**

518 Understanding the mineralogy and microstructure of binders containing mine tailings is very
 519 important towards properly understanding their behaviour. The studies found in the literature

520 are still limited but some useful initial conclusions have been drawn already. Generally, the
521 microstructure and mineralogy of the binders containing mine tailings were affected by various
522 factors including the percentages of tailings, type of usage (as admixture, SCM or cement
523 replacement), water content, activator, and presence of mineral admixtures.

524

525 4.1.1 Alkali Activated Binders

526 In geopolymers with alkali activated calcined Kaolin mine tailings, poorly crystalline pseudo
527 zeolitic alumino silicate gel, anatase and kaolinite were observed when $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio was
528 high (sodium silicate activator). However, in systems activated only with NaOH, the crystalline
529 products of hydrosodalite and Trona were identified [29]. Sedira et al. [98] activated tungsten
530 mine waste with brick waste using NaOH and Na_2SiO_3 . Poorly formed K-A-S-H, C-A-S-H, N-
531 A-S-H gels were observed indicating alkaline activation of the products was not complete.
532 Samples with more brick waste had lesser pore diameter and pore volume than the mixes with
533 mine waste alone. Similar observation was made with the use of GGBFS, where the unreacted
534 particles of the mine tailing and slag found to be surrounded by newly formed reaction products
535 [123]. An area of concern is that mixes with mine tailings can potentially lead to matrices with
536 increased porosity [124]. Silva et al [125] reported that when alkali activated, chamosite is
537 converted to crystalline phase zeolite type (sodalite). With use of sodium silicate activators,
538 minerals such as gypsum, quartz, sodium aluminium silicate, magnesium sulphate,
539 magnesite, pyrite were formed in the alkali activated paste whereas sodium aluminium silicate,
540 quartz, magnesium sulphate, pyrite, akermanite, magnesite were present in the raw tailing.
541 Sedira et al [123] reported that mixes with sodium silicate and sodium hydroxide exhibited
542 more cracks due to higher shrinkage when compared to mixes with sodium hydroxide only.
543 Also, the mixes with higher solid to liquid ratio had a more cohesive microstructure. Wang et
544 al [126] in their study with garnet tailings showed that on third day as given in Fig 10(a), the
545 matrix appeared as if many particles of 2-5 μm are glued together. This can be attributed to
546 the high crystallinity of the raw material. However, at later ages (7 days and 28 days) it the
547 original large sized particles present formed a three-dimensional crosslinked structure. Fig. 10
548 (d-f) show that the initial dissolution of the particle has been accelerated by the presence of
549 metakaolin (20%) in the system. It is evident from the absence of large sized particle in Fig.
550 (d) of geopolymer paste made of garnet tailing with metakaolin captured at 3 days.

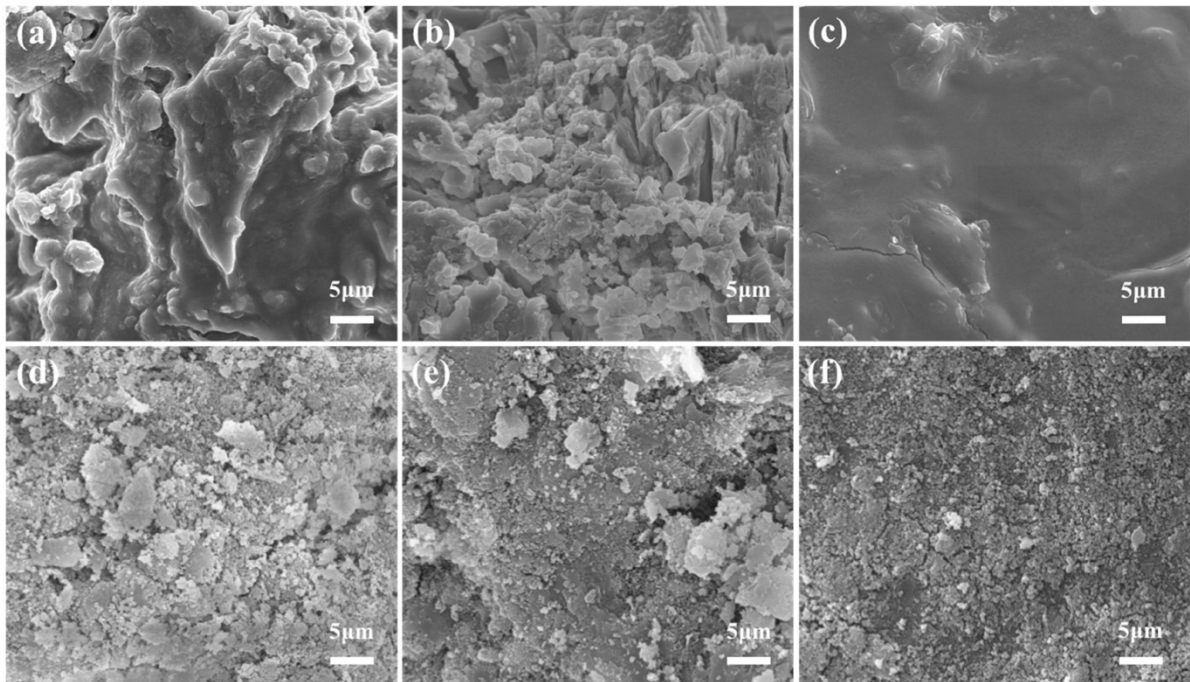


Fig. 10: Microstructure of garnet mine tailing based geopolymer at 3, 7 and 28 days (Image reproduced from Wang et al [125]).

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553
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4.1.2 Portland Cement Replacement and Supplementary Cementitious Material

556 In the literature, one can find several combinations of how mine tailings are incorporated in
557 cementitious matrices. Mixes with tailings, lime powder (10%) and OPC indicated the
558 formation of quartz, calcite, dolomite, pyrite, siderite, muscovite, clinocllore, hematite and
559 ettringite [65]. Phases such as portlandite, quartzite, alite, quartz, ettringite, calcium ferric
560 aluminate hydrate, albite and epidote were found in mixes with 20% mine tailings and OPC.
561 This indicates that all phases in mineral tailings cannot be consumed by hydration [127].
562 Martinez et al. reported that total porosity increased by 28% and mean pore size decreased
563 by 42% with 50% replacement of OPC by activated coal mine waste [76]. Ince [39] reported
564 both reduction in pore size and pore volume with increase in percentage of mine tailing. The
565 study reported that the porosity decreased from 37.5 % to 32 % for 30% cement replacement.
566 Cheng et al [86] reported that mixes with mine tailing had a flocculent structure in their calcium
567 silicate hydrates (C-S-H) accompanied by lesser quantity of portlandite crystals and attributed
568 this to the lower Ca/Si value. However, the overall microstructure was more compact and
569 uniform. A similar observation was made by Liu et al [128] in samples of 90 days age, whereas
570 at 3 days it was seen that the Ca/Si value was high due to faster dissolution of Ca from the
571 cementitious system than Si from the mine tailings. This gradually reduces with age and
572 hydration and resulting in secondary reactions leading to formation of flocculant C-S-H as
573 seen in Fig. 11.

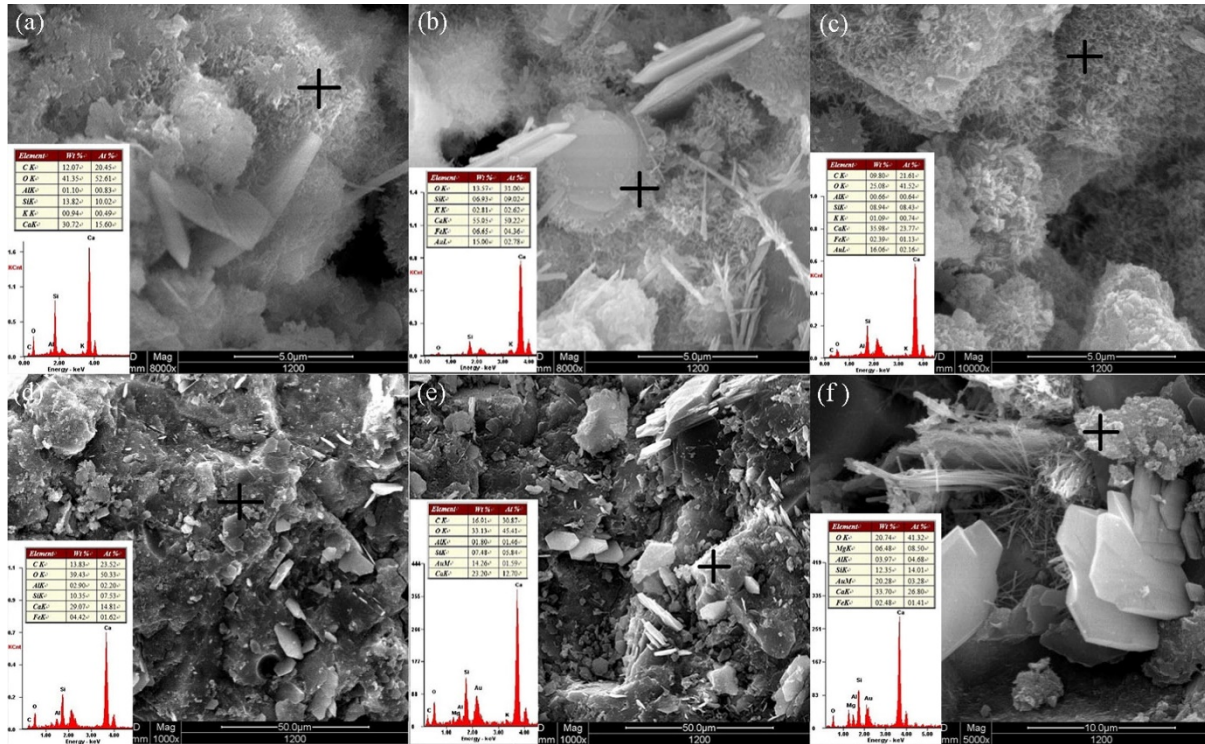


Fig. 11: Microstructure of cement paste with 0, 15, 45 % mine tailing at ages 3 (a-c) and 90 (e-f) respectively (Image reproduced from Liu et al [128]).

4.2 Hydration and Fresh Properties

Early age properties are rather critical for any binder. Fresh properties define placement and handling whereas hydration kinetics affect setting times and hence have a knock-on effect on the evolution of properties. It is well known fact in concrete technology that the addition of powdered additions has an impact on early age properties. While the published data on traditional SCMs is quite rich and conclusive, for mine tailings it is still early days. The available research is very limited and far from conclusive.

4.2.1 Alkali Activated Binders

Rao and Liu [129] reported workability as a function of alkali concentration and reported mixes made with activator at pH 14 had better workability than activator at pH 12, whereas Kiventera et al., [130] reported that the workability decreased with increase in concentration of alkalis in case of the highly sulphidic mine tailing. Perumal et al., [131] reported that the setting behaviour of alkali activated mine tailing was mainly dependant on the minerals present in it for the same activators used. Koohestani et al [85] reported that increase in sodium silicate delayed the setting of pastes. While it can be comprehended that the workability of the mine tailing is a function of minerals present and the concentration of activator, it can be seen clearly from Fig. 10 that the crystallinity of the mine tailing had an impact on hydration and the matrix formation of the pastes after alkali activation. The mechanism of polymer formation involves the following steps: i) dissolution; ii) poly-condensation; iii) gelation and iv) hardening [53,61,85,87]. This mechanism is summarised for mine tailing in Fig. 12.

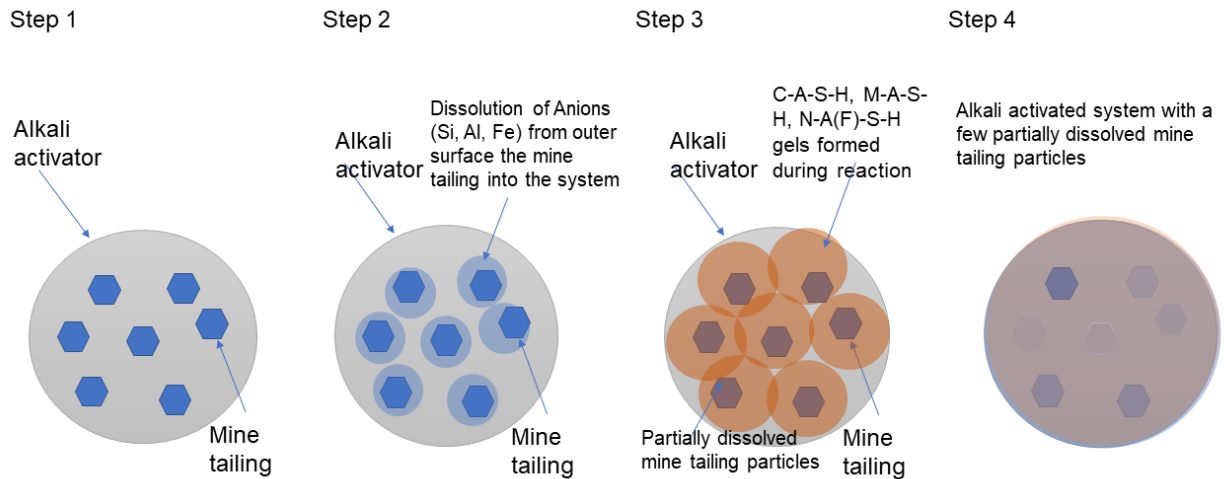


Fig. 12: Mechanism of alkali activation of mine tailing: Step 1: dissolution; Step 2: polycondensation; Step-3: gelation and Step 4: hardening.

4.2.2 Portland Cement Replacement and Supplementary Cementitious Material

The general trend observed in the published studies is that increasing mine tailings addition in cementitious binders increases the water demand [33,36,39,71]. The addition of coal mine tailing as a raw material for clinkering increased the setting time due to its high lead (Pb) content [132]. The replacement of cement tailing by 30% phosphate mine waste led to an increase in setting time, bleeding and delayed the induction phase of hydration. The study confirms the use of phosphate tailing as direct cement replacement not only has a dilution effect but also a retarding effect due to dissolution of phosphate. Increasing the proportion of phosphate tailings led to a significant drop in the yield stress, plastic viscosity, and an increase in the thixotropy of the mix [74]. Other studies do not show significant effect on setting times with 10% addition having no effect on initial setting time whereas 20% addition increasing the setting time only marginally [64]. Ince et al [33] reported that the increase in quantity of mine tailing replacement from 0% to 20% led to decrease in setting time in both lime mortar from 1250 minutes to 1180 minutes and cement based mortar from 280 minutes to 220 minutes. The yield stress and plastic viscosity of the mortar decreased with increase in percentage of mine tailing replacement, and were found to be 30.4% and 35.8% lesser than control concrete for a replacement percentage of 30% [127]. Of course, the rheological effects depend on the fineness and reactivity of mine tailings and therefore more detailed and extended research is needed. There is also some early evidence that heat of hydration and induction period are also affected. Liu et al showed that the addition of mine tailings resulted in dilution of calcium ions in the cementitious system and hence delayed the saturation point and further strength gain [128]. Therefore, it can be considered that in the initial days of hydration, mine tailings act only as fillers. To summarise, the presence of mine tailing provides additional Si required for the pozzolanic reaction and facilitate the production of secondary C-S-H. However, this Si is available at a much later age due to slow reactivity of the mine tailing and sometimes leads to formation of C-S-H with a low Ca/Si ratio resulting in a low dense C-S-H. Therefore, to achieve binder of better mechanical performance it is imperative to adjust the percentage replacement or improve the reactivity of mine tailing to facilitate the additional Si is available at the right time and quantity for reaction. Also, the presence Cu^{2+} , PO_4^{3-} and impurities such as oil from production process affect the strength gain.

635 **4.2.3 Filler**

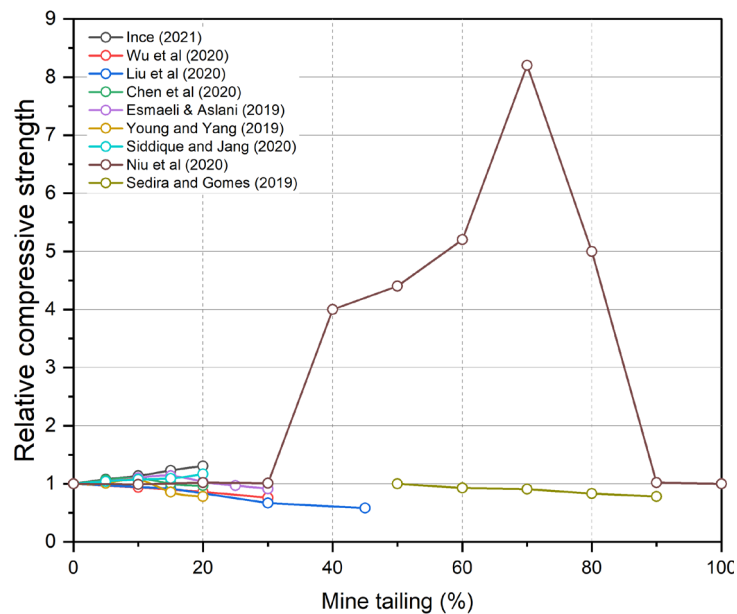
636 Using tailings as fillers, Siddique et al reported that with increase in mine tailing the flow of
637 mortar decreased even for the same water to binder ratio [133]. Similarly, Onuaguluchi and
638 Eren [36] found that flow decreased with increase in percentage of mine tailing and Zheng et
639 al [74] reported that the addition of mine tailing of a particle size higher than cement led to
640 lesser total surface area and hence reducing the water demand. The study also reported a
641 delayed setting time due to release of phosphorous ions which acts as retarder into the
642 hydrating system. Studies reported that as a filler the mine tailings increased the water
643 demand due to its higher surface area which in turn could delay the hydration and setting.
644 Also, as mine tailings might act as a reactive filler, it is important to consider its influence both
645 in packing density as well as chemical reaction.

646

647 **4.3 Mechanical Properties**

648 Studies on mortars containing mine tailings have shown compressive strength up to 75 MPa
649 as given in Table 3 and the relative compressive and flexural strengths are given in Fig 13 and
650 14. From all existing data it is evident that there is no generic trend of increase or decrease in
651 strength with the increase in percentage of mine tailings. The absence of specific patterns
652 can be attributed on various factors including particle size, type of cementitious binder, curing
653 temperature, type and extent of activation and the limited number of studies currently existing.
654 However, a point of convergence between studies is that 20-30% replacement appears to be
655 an optimum value so far.

656

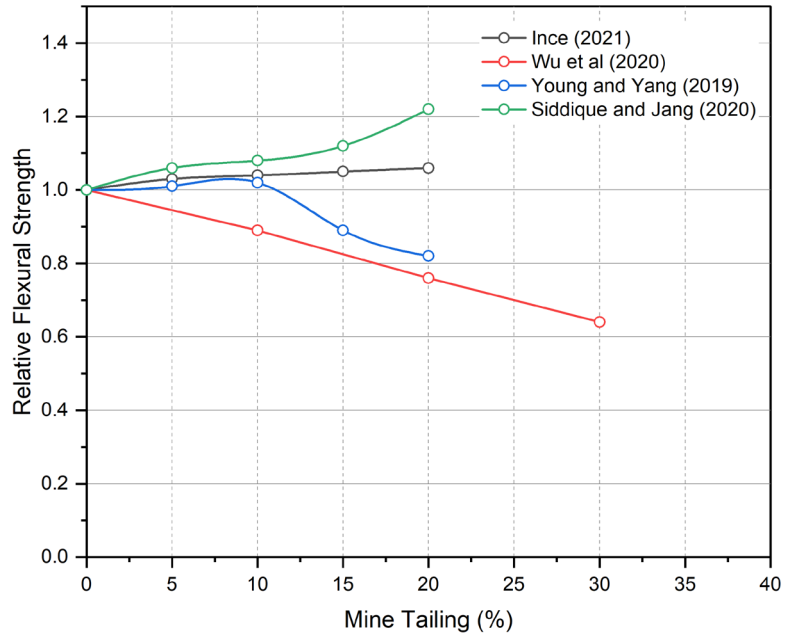


657

658

659 **Fig. 13:** Variation in compressive strength of samples with different percentages of mine tailings.

660



661

662

Fig. 14: Variation in flexural strength of samples with different percentages of mine tailings.

663

Table 3: Compressive strength variation observed in various studies

As cement replacement and supplementary cementitious material									
Sl. No	Type of tailing	Mix	Admixture	w/b ratio	Activation Process	Percentage replacement	Max. Compressive strength (MPa)	Variation in compressive strength compared to control concrete (%)	Study
1	Gold	Mortar	-	0.62	-	30% as cement replacement	37	13.9	[39]
2	Copper	Mortar	-	0.485	Mechanical activation	10% as cement replacement	46.5	35.5	[36]
3	Coal	Mortar	-	0.50	Thermo mechanical activation	10% as cement replacement	64	- 8.3	[64]
4	Tungsten	Mortar	GGBFS	0.52		10% as cement replacement	35	-14.6	[31]
5	Coal	Mortar		0.30	Thermal activation	6 % as cement replacement	59	-1.7	[134]
6	Coal	Mortar		0.50	Clinkering	7% as clinker raw material	60	1.7	[132]
7	Coal	Mortar	-	0.50	Mechano-Thermal activation	20 % as cement replacement	55.7	-11.9	[76]
Alkali activated cements									
Sl. No	Type of tailing	Mix	Admixture		Activation Process	Percentage replacement	Maximum Compressive strength achieved (MPa)		Study

1	Vanadium	Mortar	Sodium hydroxide and Metakaolin	Thermo chemical activation (Alkaline roasting)	70 % mine tailing and 30% metakaolin	55.7	[135]
2	Kaolin	Paste	Sodium hydroxide, Sodium silicate,	Thermo mechanical treatment followed by Alkali activation	100% mine tailing	75	[29]
3	Boron	Mortar	Sodium silicate, sodium hydroxide	Thermo mechanical treatment followed by alkali activation	100 %	30.7	[55]
4	Tungsten	Mortar	calcium hydroxide, sodium hydroxide, sodium silicate	Thermal treatment followed by alkali activation	100%	75	[115]
5	Tungsten	Mortar	Sodium silicate, sodium hydroxide Potassium Hydroxide and ground granulated blast furnace slag	Alkali activation	90% mine waste and 10% GGBFS	33	[123]
Alkali activated material as a replacement of OPC							
1	Tungsten	Mortar	Sodium silicate, gypsum, lime	Mechano-chemical activation	30% cement replacement	56.95	[51]

666 4.3.1 Alkali Activated Binders

667 The mineral Albite, calcined in presence of alkali, when used as a one part cement achieved
668 a strength of up to 44 MPa at 28 days [93]. Tungsten mine waste calcined in presence of
669 Na_2CO_3 , yields high early age strength up to 45 MPa on alkali activation with NaOH, and the
670 strength decreases at later ages due to the formation of unstable compounds due to ineffective
671 activator combination [104]. The molarity of NaOH solution found to have an effect on the
672 development of compressive strength of binders with activated tungsten mine waste [104].
673 More specifically, about 18 MPa, 30 MPa and 70 MPa were achieved using 8 M, 14 M, and
674 24 M NaOH solution for activation of the waste, in presence of $\text{Ca}(\text{OH})_2$. Geopolymers
675 synthesized from non-calcined raw materials display a higher increase in the compressive
676 strength in the later stage of the geopolymerisation reaction [129]. Jiao et al [135] indicated
677 the curing temperature played an important role in strength gain of geopolymers with mine
678 tailings where the compressive strength increased from 35.1 MPa to 55.7 MPa when the
679 curing temperature was increased from 20 °C to 60 °C. At room temperature curing, the
680 strength gain happens at a later age about at 14 days [85]. The feasibility of development of
681 sodium silicate from mine tailing was studied. It was found that the sodium silicate produced
682 from the tailings were able to successfully activate a metakaolin based system as geopolymers
683 and achieve a strength up to 32.1 MPa as compared to a strength of 40 MPa by commercial
684 sodium silicate [136].

685

686 4.3.2 Portland Cement Replacement and Supplementary Cementitious Material

687 Malagon et al [132] used 7% of coal mining waste as raw material for producing cement clinker
688 and observed a 9-14% decrease in strength due to hindering of hydration by the Cu ions
689 present in the mine waste. A replacement of 30% cement with phosphate tailings causes a
690 decrease in flexural strength by 25% and compressive strength by 34.7%, 38.9%, 40.7% at 3
691 days, 28 days and 90 days, respectively [74]. Similar observation was made by Liu et al [128]
692 where the compressive strength decreased from about 88 MPa to 35 MPa when the
693 replacement percentage was 45%.

694

695 Martinez et al., [77] suggested when activated coal mine waste was used as cement
696 replacement up to 20% attained strengths were at the 42.5 MPa class grade range whereas
697 increasing the replacement to 50% performance dropped to 32.5 MPa class. Esmaeli and
698 Aslani [44] proposed an optimum replacement percentage for copper mine tailings at which
699 the compressive strength was greater than the control mixes by 5% and elastic modulus and
700 split tensile strength were greater by 10% and 12 %, whereas Vargas and Lopez [91] reported
701 all percentage of replacements lead to decrease in strength and use of copper mine tailing as
702 an addition to the matrix as a filler improved the strength. Similarly, Liu et al found that at all
703 water cement ratios the strength reduced with increase in graphite tailing content by up to 29%
704 - 42% for 15% cement replacement due to free graphite and hydrocarbon oil leading to
705 improper bonding and hydration [137]. Ince [39] reported a maximum compressive strength of
706 about 37 MPa was achieved when the percentage replacement of cement by mine tailing was
707 30%, proving to be the optimum and all mine tailing mixes had compressive strength values
708 higher than mixes with 100% cement. In their recent study, Ince et al [33] reported that the
709 compressive strength of mixes increased with increasing percentage of mine tailings for both
710 cement and lime mortars up to a replacement of 20% as shown in Fig 8. The mortars
711 developed by Ince et al [33] were then used to join masonry units and it was found that the
712 dewatering of the mortar (i.e. water movement towards the masonry) had an impact on the
713 evolution of properties. When dewatering happened before curing the cement mortar mixes

714 with 20% mine tailing had approximately 20% lesser strength than that of control. The study
715 also reported a reduction in split tensile and flexural strengths of the dewatered cement mortar
716 with use of mine tailings whereas the same improved with increase in percentage of mine
717 tailings for dewatered lime mortar. The mechanisms that underpin better behaviour in lime
718 mortars are not yet well studied. Jian et al [138] observed that addition of up to 2-6% of mine
719 tailing as a clinker raw material affected negatively the strength development of the
720 cementitious matrix, with an exception of 2% replacement at 60 days. More specifically, for
721 0%, 2%, 4% and 6% replacement the strength of the cementitious matrix at 28 days was 44.1
722 MPa, 18.6 MPa, 11.3 MPa and 5.7 MPa respectively; while at 60 days it was 66.8 MPa, 71.5
723 MPa, 48.9 MPa and 25.3 MPa respectively.

724

725 4.3.3 Filler

726 Existing data suggests that the use of mine tailings as fillers in cementitious matrices is also
727 not conclusive. Some suggest that the filler effect can lead in the improvement of mechanical
728 properties probably due to better packing, while others report reduction in properties. Sidique
729 and Jang showed that incorporation of 5%, 10%, 15% and 20% mine tailings led to an increase
730 of 3%, 1%, 10% and 16% in compressive strength and 2%, 5%, 24% and 17% in flexural
731 strength respectively [133]. All the tailing added mixes had better resistance to abrasion and
732 impact as compared to control concrete. However, Zheng et al [74] reported a decrease in
733 performance of mortar mixes with increase in percentage of mine tailings. The study reported
734 a 25% decrease in flexural strength and a 35.9% decrease in compressive strength for a 30%
735 addition of mine tailing as a filler. This decrease in performance compared to control mixes
736 could be due to the dilution effect as the study aimed reduction in cement usage by replacing
737 cement by a filler.

738

739 It can be outlined from the past studies that obtaining a binder of high compressive strength
740 up to 75 MPa is possible with the use of different mine tailings. However, the published results
741 are not yet conclusive while at the same time there is a large spread of processing techniques
742 and replacement percentages (for the tailings used). Therefore, there is a need for a
743 systematic approach and clear understanding of: the mineralogy (and its variability) of tailings;
744 suitable activation techniques; optimum duration of processing; choice of activator(s);
745 optimum concentration of activator(s) and the percentage of replacement of cement in
746 traditional cementitious binders.

747

748 **4.4 Durability**

749 The durability of the mine tailing binders was in most cases found to be a function of porosity
750 and microstructure. The performance of mine tailings with respect to corrosion behaviour,
751 chloride penetration and resistivity were affected by the presence of conductive elements such
752 as graphite and iron. Though, the past studies do not indicate a standard trend in improvement
753 or reduction in durability performance with increase in percentage of mine tailing, it was always
754 observed in certain cases that optimum ranges for mine tailings' utilisation exist and within
755 these optimum ranges the binders performed well.

756

757 4.4.1 Alkali Activated Binders

758 Jiao et al [82] reported a high thermal stability of geopolymer mixes with mine tailings. The
759 compressive strength of the samples was unaffected up to an exposure temperature of 600°C,
760 and it increased up to a temperature of 900°C due to densification. Ahmari and Zhang [139]
761 found that there were no visible efflorescence, disintegration or cracks even after four months

762 of immersion in solutions of pH 4 and 7, however the unconfined compressive strength
763 decreased by 59.3% and 53.3% from 11 MPa respectively, after immersion. A similar result
764 was reported by Kiventera et al [130]. Their study showed that mine tailings mixed with pure
765 NaOH produced a binder which performed poorly after immersion in water. However, the same
766 performance was improved by use of ground granulated blast furnace slag in combination with
767 tailings as a precursor. Falah et al [140] reduced the water absorption of alkali activated
768 concrete with mine tailing by using 30% submicron sized mine tailings. Perumal et al [112] in
769 their study reported that alkali activated mine tailing pastes exhibited a water absorption of
770 35%, 30%, 26%, 22.5% and 20% when the grinding time was varied from 1,2,4,8, 16 minutes
771 respectively.

772

773 4.4.2 Portland Cement Replacement and Supplementary Cementitious Material

774 In general, water absorption of the mixes with mine waste were found to be higher than that
775 of mixes with OPC and increased with increase in percentage of mine tailings [36,76]. When
776 OPC was replaced by 50% coal mine waste, the water absorption increased by 32% [76].
777 However, Esmaeli and Aslani [44] reported that the water absorption decreased up to an
778 optimum percentage of replacement and increased beyond that. In another study [44] mixes
779 with optimum replacement ratio of 15% and 20% mine tailings had marginally lower water
780 absorption (~2%), when compared to control concrete (2.4%). The depth of water absorption
781 was also reported to decrease up to 20% replacement and increase beyond 20% cement
782 replacement. Though the depth of water penetration increased after 20% replacement, it was
783 still found to be less than control concrete (29.3 mm) at 30% replacement (24.2 mm) [44].
784 Similarly, Cheng [86] reported an optimum replacement percentage of 30% beyond which the
785 performance of samples after immersion in water starts declining. Similar increase in trend
786 was also observed in case of water absorption under pressure where the depth of water
787 penetration for 0%, 20% and 50 % cement replacement was 3.6 mm, 6.5 mm and 15.8 mm
788 respectively [76]. The study also reported primary absorption coefficient through capillary
789 sorption measurements where it was similar for samples with 50% replacement and control
790 mixes whereas samples with 20% replacement performed better than control. Ince et al [33]
791 reported that water penetration depth and porosity increased with increase in percentage of
792 mine tailings for cement mortar and decreased with increase in percentage of mine tailings for
793 lime mortar. It is evident, that the reported data is scattered and to a certain extent confusing.
794 Two studies that focused on freeze thaw performance concluded that the reduction in pore-
795 size (as a result of the addition of mine tailings) led to a poor freeze thaw performance with
796 the mixes containing graphite mine tailings reported a mass loss of about 5% after 25 cycles
797 of freeze and thaw [39,45].

798

799 Ince reported that carbonation depth decreased from 9.8 mm to 5.6 mm for 30% cement
800 replacement [39] whereas Cheng et al [86] reported that the carbonation depth increase with
801 increase in percentage of mine tailings. However, the mixes with mine tailings had better
802 compactness and were reported to be less porous. On the chloride ion penetration front,
803 mixes with mine waste exhibited a better chloride resistance, irrespective of the type of mine
804 waste used. The samples with 50% coal mine waste had a chloride resistance 5 times greater
805 than that of control samples and samples exposed to chloride indicated the formation of
806 Friedel's salt which was in line with the behaviour of samples with fly ash [77]. The chloride
807 migration test indicated that the samples with mine tailings performed better than the control
808 sample in chloride migration test. The samples with mine tailing fell in the category of high
809 chloride diffusivity and the control samples were in the category of very high chloride diffusivity

810 [44]. A study on the corrosion development, found that the corrosion onset time increased
 811 linearly with increase in percentage of mine waste, indicating that the mine tailings delayed
 812 the initiation of corrosion [141]. Graphite tailing improved the electrical conductivity of concrete
 813 which, on the other hand, can be problematic when it comes to corrosion. The surface
 814 resistivity decreased from 62 kΩ·cm for normal concrete to 32 kΩ·cm for concrete with 15%
 815 graphite tailing [49]. The surface change for samples with graphite tailing was almost twice as
 816 that of control. The surface temperature reduced rapidly with increase in percentage of
 817 graphite tailing [45].

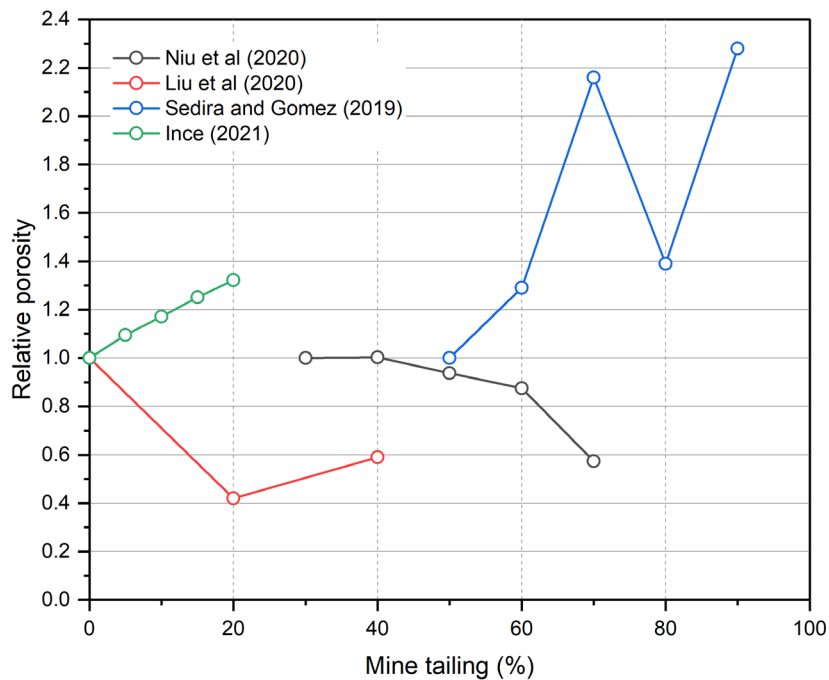
818

819 **4.4.3 Filler**

820 Incorporation mine tailings as fillers increased density and lowered water absorption and total
 821 void content. After 24 hour of exposure to water the mixes with 20% mine tailings recorded a
 822 11% decrease in rate of water absorption [133]. The mixes with 10% mine tailings were highly
 823 resistant to sulphate attack. Though the mixes with 15% and 20% mine tailing exhibited higher
 824 strength loss and weight loss compared to mixes with 10% mine tailings, all the mixes with
 825 mine tailings performed better than control mixes. Similarly mixes with 10% mine tailing were
 826 more resistant to acid attack [133]. Mass loss and depth of chloride penetration decreased
 827 with increase in percentage of mine tailing, depth of penetration was 10.7 mm for control mixes
 828 whilst 3.5-3.7 mm for mixes with mine tailing, similarly 4.3% and 3.4-3.7% mass loss for both
 829 the mixes. The mass loss of the sample when exposed to HCL was 4.3% for control and 3.4%
 830 - 3.7 % for samples with mine tailing [36]. Addition of mine tailing also increased the drying
 831 shrinkage, however the drying shrinkage of mixes with 10% mine tailing was lower than the
 832 mixes with 5%, 15%, 20% and 25% mine tailings [74].

833

834 Figure 15 shows published data on the variation of measured porosity for different
 835 percentages of mine-tailings used in the binders.



836

837 **Fig. 15:** Variation in porosity of samples with different percentages of mine tailings.

838

839 **4.5 Environmental Properties - Leaching of Heavy metals**

840 Table 1 indicates that mine tailings are in general obtained with the presence of heavy metals
841 such as Pb, Zn, Cd, Cu, Ar, Cr, V, Zr. This always poses a danger of leaching of heavy metals
842 into the environment over the period when mine tailing is used as a construction material.
843 Immobilising heavy metals is very critical in this discussion. Heavy metals can become a part
844 of a 3-dimensional chain and thus get immobilised, and this process is known as locking. The
845 ionic size and valence of the heavy metals are the major factors influencing locking.

846

847 4.5.1 Alkali Activated Binders

848 Wan et al., [70] studied immobilisation of lead from lead nitrate addition by alkali activation of
849 sphalerite floatation mine tailing (Zinc tailings) using sodium silicate and reported that about
850 97.9 – 99.5% of heavy metals were immobilised when metakaolin was used as an additive
851 and 92.26% were immobilised when only mine tailings were used. Similarly, for the same
852 amount of alkali activated of tungsten mine waste the heavy metals leaching decreased with
853 increase in percentage of GGBFS. In either case, the leaching value was less than the
854 permissible value given by Korean guidelines, TCLP toxic limits [142] and European guidelines
855 [143] for cadmium, zinc, copper and lead [31]. Gomes et al., [144] reported that alkali activated
856 tungsten mine wastes when water cured stabilizes at a pH of 7 after about a week. This makes
857 it fall within the range of 2-8.5 in which arsenic leaching is negligible. Peng et al., [51] reported
858 that the Pb in tungsten mine waste are solidified in the garnet crystal and therefore cannot
859 leach.

860

861 4.5.2 Portland Cement Replacement and Supplementary Cementitious Material

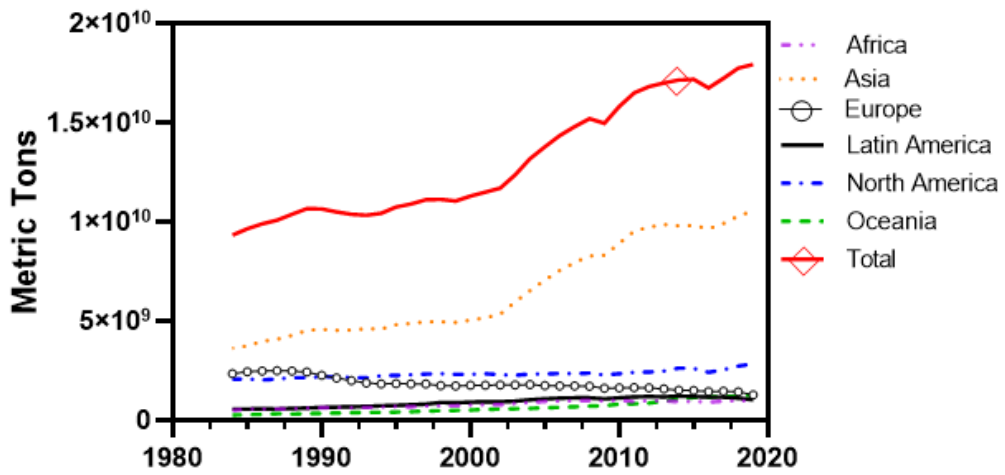
862 Ince [39] found that when sulphidic gold mine tailing was used as a cement or sand
863 replacement the leaching was always within the limits specified by US Code of Federal
864 Regulations (CFR). And it was also observed that when the percentage of replacement was
865 higher the leaching was lower due to pore structure refinement. The Cr, Pb and Cd leached
866 was 0.153, 0.0004, 0.00007 mg/ L when the percentage replacement was 10% and it
867 decreased to 0.129, 0.0003 and 0.00005 mg/L respectively, when the percentage replacement
868 was 30%. When compared with other standards it can also be seen that these wastes qualified
869 as non-hazardous as per the TCLP toxic limits [142] and as non-toxic as per the framework
870 for eco-toxic materials as per European guidelines [143]. Gou et al., [23] also reported that the
871 clinkering and hydration of cement with mine tailing prevents the leaching of heavy metals
872 when mine tailings are used as a cement replacement. In a similar line, Nouairi et al., [145]
873 reported that 75% of Pb, 85% of Zn and 80% of Cu were immobilized by hydration of Zn-Pb
874 mine tailings in mortar. Pyo et al., [68] reported that though the As, Pb, Zn and Cd content of
875 quartz tailing were higher than the other silica powder their values were within the permissible
876 limits. The study also reported the reduction in leaching due to physical and chemical
877 encapsulation of heavy metals when used in ultrahigh performance concrete as
878 supplementary cementitious material.

879

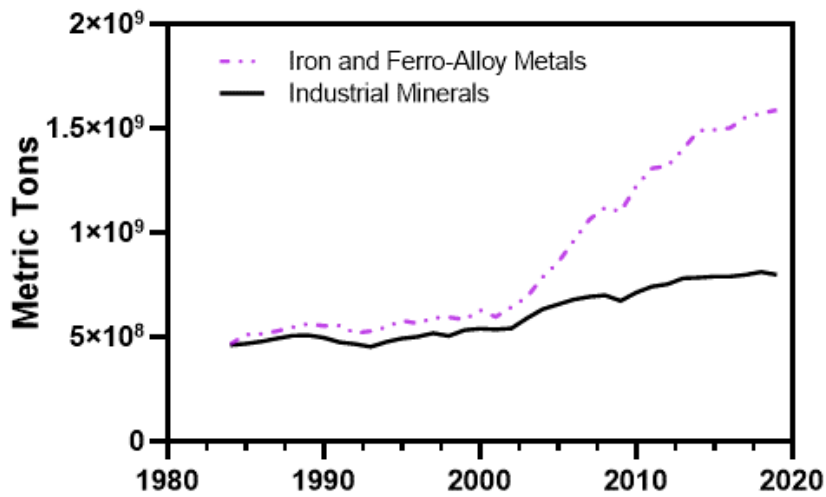
880 **5. Future Outlook**

881 Raw materials form the basis of our technological development in all sectors of our economies.
882 From building and construction operations to the manufacture of bespoke electronic
883 components, raw materials and minerals are important. World mining data shows significant
884 increase in the extraction of mining products the last 35 years (Fig. 16).

885



(a)



(b)

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Fig. 16: Production of mining products between 1985 and 2020: (a) all mining products per geographical region and (b) global production of two major mining commodities (Data obtained from World Mining Data [146]).

Since 2000, there is a steady increase of mining production except for Europe. For Asia and Australia, this increase has been considerable (Fig. 17).

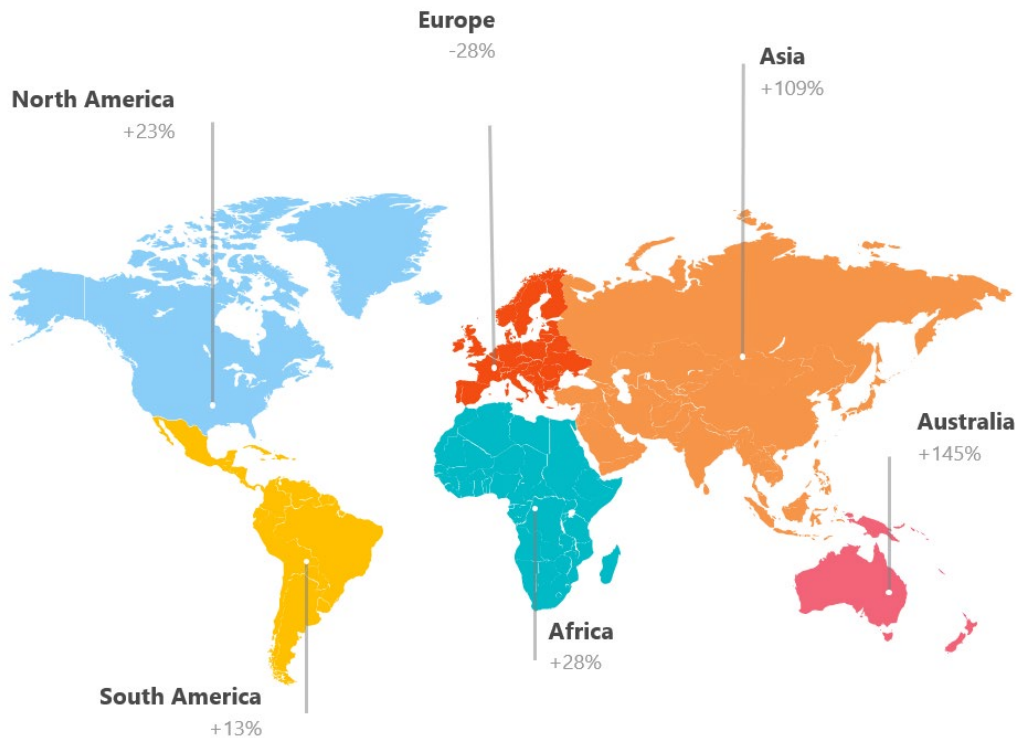


Fig. 17: Production growth of mining products per geographic region between 2000 and 2020 (Data obtained from World Mining Data [146]).

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At the same time, the sustainability goals (SDGs), which in many countries are now part of the legislation, have created the perfect growth conditions for low-carbon technologies. From wind turbines to solar panels and electric cars, these developments require vast volumes of minerals for production and manufacturing. Therefore, the road to a low-carbon sustainable future will be a very mineral intensive road. Industrial level mining directly and positively impacts: the development of good jobs and economic growth (SDG8); provides raw materials that support the development of renewable energy technologies (SDG7) which in turn facilitates innovation and infrastructure (SDG9). At the same time, it can negatively impact terrestrial ecosystems (SDG15) as well as sustainable clean water consumption and availability (SDG6). The effects of mining are also found to impact attaining the goals related to life below water (SDG14), actions related to climate change (SDG13), ensuring healthy lives for all communities (SDG 3) and responsible consumption and production patterns (SDG12) (www.undp.org). Taking these into consideration, the Responsible Mining Foundation proposed a framework for responsible mining [147]. This report declares tailings management as a major environmental responsibility and efficient tailing management shall improve good health (SDG3), sustainable cities and communities (SDG11), responsible consumption (SDG12), climate action (SDG13), life below water (SDG14). When the quantity of tailings disposed in land and into water bodies and confined dams are reduced contamination of land and water is avoided and hence improves life on land (SDG15) and clean water and sanitation (SDG6).

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On the other side of this discussion is the construction sector, which is equally, if not more, responsible for heavy environmental burden. The civil infrastructure sector is global, is huge and utilises extraordinary volumes of materials for the manufacture of core construction composites. This contribution has highlighted the chemical and mineralogical similarities

926 between tailings and pristine raw materials used in the manufacture on cement-based binders.
 927 However, the current state-of-the-art in the field of utilising tailings in such binders albeit
 928 showing some promising results is limited and largely inconclusive as discussed in the
 929 preceding sections. A list of limitations for such use is given in Table 4. All these challenges
 930 highlight the importance of extensive and systematic research on this area, something that
 931 can lead to large scale valorisation of mineral waste streams in large manufacturing processes
 932 such as cement and concrete.

933
 934 **Table 4:** Challenges for use of mine tailings in cementitious binders and potential solutions
 935

Challenges	Potential solutions
Variability in property and quality of various mine streams	It is difficult to assign a single method of reuse/recycle for all wastes. However, proposing a classification system based on certain key parameters from characterisation and providing customised solution for each category shall be more practical from industry point of view. Clustering tailing streams and mapping their properties can allow the development upper/lower limits in their use, increasing the confidence of post-processors and/or end-users.
High crystallinity and non-reactive nature of mine tailings.	The reactivity of the mine tailings shall be improved by mechanical, thermal, or chemical activation. The field of mechanochemistry can provide vital assistance on this issue.
Presence of heavy metals and metalloids such as Hg, Pb, Cr, V, Cd, Mn, Zn etc in variable quantities is observed in mine tailings. This may leach into the environment when used in binders in construction.	Heavy metals and metalloids get encapsulated and are prevented from leaching when used in cementitious binder [23,145]. Also, under the umbrella of circular zero waste economy, options are being explored to mine heavy metals from the mine tailings before their disposal. This may help in removal of heavy metals prior to their disposal and eliminate the risk of leaching.[148].
Presence of Cu^{2+} , PO_3^{2+} which interfere with the hydration and setting processes.	The presence of such ions shall be noted, and the percentage of replacement shall be decided as to keep these ions within permissible limits. Cement technology methods can be utilised to alleviate hydration and/or setting issues.
Lack of guidelines and standards. The existing guidelines and standards do not explicitly provide a methodology for	More research and development should be carried out in this area to strengthen the knowledge data base to provide a rational approach and guideline towards use of mine

<p>suitability assessment of mine tailing as an SCM or precursor for alkali activation.</p>	<p>tailings or any new waste in cementitious binder. Currently with advent of Horizon Europe program and the United Nations SDGs, various research and innovation projects are spearheaded in this direction.</p>
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936

937 Use of mine tailing in cementitious binders not only contributes to environmental, innovation
 938 and health related benefits as discussed. It can also contribute to SDGs related to economy
 939 and our sustainable urban/rural habitats such as the development of good jobs and economic
 940 growth (SDG8) and making human settlements safer, more resilient and sustainable (SDG11).

941

942 The argument that the variability in mine waste streams is a limiting factor should not be a
 943 “preventing” factor to commit to the use of mine tailing in cementitious binders. First of all, the
 944 field has not been explored sufficiently and therefore there is no hard evidence that variability
 945 is an issue or that it cannot be tackled. There is variability in fly-ash and ground granulated
 946 blast furnace slag as well. This did not prevent their extensive utilisation in concrete technology
 947 for almost 3.5 decades. Secondly, the fact that the research in this area is limited can be used
 948 as a motivation for research and innovation to strengthen the means of implementation and
 949 revitalize the global partnership for sustainable development (SDG17). It should contribute to
 950 capacity development, science and technology towards these goals. Cement-based materials
 951 research has dealt with such issues before. Not every fly-ash, slag or silica fume is the same
 952 for example. Nonetheless, intensive research and development created the necessary
 953 confidence on these, once novel and high-risk, supplementary cementitious materials, and
 954 they have been used extensively for more than three decades now.

955

956 It is now time to draw our attention on different sources of cementitious materials and binders.
 957 As discussed at the start of this article, it is now a common belief, even at institutional level
 958 (e.g., European Cement Association), that new cementitious materials must be developed.
 959 Mine tailings provide a great opportunity of sourcing raw materials for new binders without
 960 further exploitation of natural resources. Detailed and systematic studies will contribute
 961 towards this goal improving the level of confidence between engineers and manufacturers.
 962 The impact of successful reuse of mine tailings will be enormous contributing significantly
 963 towards the global sustainability goals.

964

965 **6. Conclusions**

966 The review of the past studies indicate that the mine tailings can be used as a direct cement
 967 replacement or supplementary cementitious material or alkali activated cements based on the
 968 chemical, mineralogical and physical properties. Their usage in most cases is after activation
 969 of minerals in the mine tailing.

970

971 The process of activation involves creation of structural defects by grinding, heating or by alkali
 972 activation. The choice of activation method is based on the minerals present in the mine
 973 tailings. Calcination has been found to be highly effective in converting the crystalline phases
 974 phyllo-silicate, hydroxide and carbonate minerals to amorphous phases. The grinding process
 975 improves the reaction by increase in surface area, by reduction of particle size and by
 976 developing new fractured surfaces and suitable of minerals of low hardness such as mica,
 977 talc, clay minerals and other phyllo-silicates. When two different minerals of different hardness
 978 are present the possibility of differential grinding has also been reported. Both grinding and

979 calcination are highly energy intensive and are counterproductive beyond the optimum
980 duration. This emphasizes the significance the mineral phases have on the selection of
981 activation methods. Alkali activation is also a potential route for processing mine tailings. With
982 the change in alkali concentration and choice of alkali activators a strength range varying from
983 15 MPa – 75 MPa or even higher has been achieved. However, this process demands further
984 understanding on formation of polymeric chains of C-A-S-H, N-A-S-H, K-A-S-H, C-(N)-A-S-H,
985 N-F-(A)-S-H in presence different amounts of Ca, Si, Fe and Al ions and their stability. When
986 a combination of two or more alkali activators are used the compatibility of one activator with
987 another, and the minerals present in the precursor, play a critical role in terms of long-term
988 strength. Activation processes can be complementary (e.g., mechano-chemical) but they
989 always have to lead to the maximum benefits at minimum possible consumed energy.

990

991 The past studies indicate a high potential for use of mine tailings in cementitious binders.
992 However, published literature also points out the lack of an activation method or mix design
993 procedure which is more systematic rather than trial and error. Undoubtedly, mine tailings
994 provide a great source of raw materials for new binders without further exploitation of natural
995 resources. Intensive and systematic research in this area will improve the level of confidence
996 of engineers and manufacturers and can have a significant impact towards attaining the global
997 sustainability goals.

998

999

1000 **Acknowledgements**

1001 Financial support for the lead author from the Doctoral Training Alliance DTA3/COFUND Marie
1002 Skłodowska-Curie is gratefully acknowledged.

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1027 **Appendix – Published data on reactivity and activation processes**

1028

1029 **Table A1:** Reactivity and activation processes of minerals present in mine tailings according to the existing literature

1030

Mineral/ group reported	Mineral	Reactivity/ Property in raw form	Activation possibility	Literature
Albite		A crystalline alumina silicate mineral with formula $\text{NaAlSi}_3\text{O}_8$. Slow reactive and stable up to 1000 °C under normal conditions	Albite was completely amorphized when calcined at 1000 °C or 1150°C in presence of 50% NaOH or Na_2CO_3 respectively. Albite was observed to be stable at temperatures below 1000°C	(Feng et al., 2012; Perumal & Illikainen, 2019)
Amphibole		Inosilicate minerals of double chain SiO_4 tetrahedra with a better dissolubility as compared to Quartz	The amphibole phases decreased significantly when mixed with blast furnace slag, cement clinker and finely ground to a powder and mixed with warm water and cured at a temperature of 48 ± 1 °C. Also, shall be used as a precursor for geopolymer.	(Huang et al., 2012; Kinnunen et al., 2018; Luukkonen et al., 2018)
Anatase		Anatase is a mineral form of TiO_2 and is crystalline. Anatase is less stable as compared to Rutile.	TiO_2 shall be immobilised in presence of phyllosilicates such as kaolinites. Admixtures prepared with anatase, alkali and phyllosilicates by calcination at 800 °C were found to improve compressive strength by about 37%	(Zhang et al., 2017)

Ankerite	Ankerite is $\text{Ca}(\text{Fe, Mg, Mn})(\text{CO}_3)_2$, and has properties similar to dolomite	Being a carbonate mineral ankerite is prone to decomposition at a temperature range of 650 °C to 850 °C	(Argane, Benzaazoua, et al., 2016)
Anorthite	Anorthite $((\text{Ca,Na})(\text{Si,Al})_4\text{O}_8)$ is a member of plagioclase feldspar member series	Quartz, calcite, and chlorite decompose to form anorthite at a firing temperature of about 1100 °C and contribute to the strength bricks and ceramics. However, Anorthite partially melts to liquid phase at 1200 °C. In presence of NaOH/ KOH dissolution of Si and Al in Anorthite happens to form reactive components	(Choi et al., 2009; Li et al., 2017; H. Xu & Van Deventer, 2002; Yang et al., 2014)
Apatite	Apatite is a group of phosphate mineral including hydroxyapatite $(\text{Ca}_5(\text{PO}_4)_3(\text{OH}))$, fluorapatite $(\text{Ca}_5(\text{PO}_4)_3\text{F}_2)$ and chlorapatite $(\text{Ca}_5(\text{PO}_4)_3\text{Cl}_2)$	Apatite indicated no/ lesser solubility in presence of alkali. However, shall be used in heavy metal encapsulation in ceramics. In a basic environment at a temperature of about 550 °C the phosphates from apatite and Al react leading to decrease in apatite peaks.	(Kinnunen et al., 2018; Zheng et al., 2015)
Arsenopyrite	Arsenopyrite (FeAsS) is a hard Iron Arsenic Sulphide crystal. p	When arsenopyrite was heated to 550 °C using microwaves Pyrite was formed. Arsenopyrite reacts with alkali and leaches As.	(Kiventerä et al., 2016; Pyo et al., 2018)

Augite	Augite is a chain silicate mineral with a stoichiometric formula $(Ca, Mg, Fe)_2(Si, Al)_2O_6$	Augite reacted when mixed with blast furnace slag, cement clinker and finely ground to a powder and mixed with warm water and cured at a temperature of 48 ± 1 °C.	(Huang et al., 2012; Hua Xu & Van Deventer, 2000)
Barite	Barite is a Barium sulphate mineral of formula $BaSO_4$	When used in mortar without treatment remained unaltered even till 180 days	(Argane et al., 2015)
Biotite	Biotite is a phyllosilicate mineral within the mica group of formula $K(Mg,Fe)_3AlSi_3O_{10}(F,OH)_2$ for the i^{th} member	Biotite underwent partial decomposition in presence of sodium silicate	(Moukannaa et al., 2019)
Caldecaydrite	$CaAl_2O_4 \cdot 10H_2O$	It can be used in combination with other silicate minerals or other source of silica shall be considered for the use in binders	(Paiva et al., 2019)
Calcite	Calcite is calcium carbonate mineral of formula $CaCO_3$	The decomposition occurs completely at about 800 °C under calcination and at about 550 °C under alkali fusion.	(Frías et al., 2016; Moukannaa et al., 2019)
Chlorite	Chlorite is a group of phyllosilicate minerals and shall contribute to pozzolanic properties in raw form	Reactivity shall be improved by calcination	(Simonsen et al., 2020)
Clinochlore	Clinochlore is a member of chlorite group of minerals with the formula $(Mg,Fe)_6(Si,Al)_4O_{10}(OH)_8$	Structural decomposition of Clinochlore occurs at about 505 °C of calcination.	(Sedira et al., 2018; Yang et al., 2014)
Colemanite	Colemanite is a borate mineral of formula $Ca_2B_6O_{11} \cdot 5H_2O$	Colemanite decomposed forming CaO upon calcination up to 600 °C	(Aydlın & Kızıltepe, 2019)
Corundum	Corundum is an Al_2O_3 mineral phase with better stability and crystallinity	Corundum disappeared on sintering above 1100 °C	(Liu et al., 2015)

		indicating decomposition and fusing to form other minerals depending on the elements present in the matrix	
Diaspore	Diaspore is an aluminium hydroxide mineral $\text{AlO}(\text{OH})$	Diaspore turns to Corundum when calcined at a temperature of $800\text{ }^{\circ}\text{C}$ for 1 h. The specific temperature of decomposition was at $510\text{ }^{\circ}\text{C} - 540\text{ }^{\circ}\text{C}$.	(Ye et al., 2017). (Vangelatos et al., 2009)
Diopside	Diopside is a pyroxene mineral of formula, $\text{CaMgSi}_2\text{O}_6$	Alkaline roasting of diopside in presence of NaOH at $450\text{ }^{\circ}\text{C}$ led to its decomposition.	(Jiao et al., 2013)
Dolomite	Dolomite has a general formula of $\text{MgCO}_3 \cdot \text{CaCO}_3$.	Can play a role as an activator of pozzolans and shall also be used as a replacement of limestone. The peaks of dolomite, decreased significantly when mixed with blast furnace slag, cement clinker, and finely ground to a powder and mixed with warm water and cured at a temperature of $48 \pm 1\text{ }^{\circ}\text{C}$. The decomposition of dolomite by dehydration and decarbonation processes complete at $850\text{ }^{\circ}\text{C}$ of calcination.	(Huang et al., 2012; Moukannaa et al., 2019; Simonsen et al., 2020)
Feldspar	Feldspars are group of silicate minerals with calcium, aluminium, and potassium in it.	Completely dissolves in presence of a 10 M NaOH and shall be used as a	(Luukkonen et al., 2018; Yao et al., 2021; Young & Yang, 2019)

		precursor for alkali activated materials. Also decomposes while clinkering at a temperature of 1450 °C Grinding up to 200 minutes increased the pozzolanic activity index of feldspar from 61% to 84%.	
Fluorite	Fluorite or fluorspar is a calcium fluorite mineral CaF_2	When used in mortar without treatment remained unaltered even till 180 days	(Argane et al., 2015)
Garnet	Garnet is a group of aluminium silicate minerals. It has high hardness, structural and chemical stability, and low cementitious property. It is also reported to have solidified chromium in it. It	Garnet minerals exhibited low dissolution in presence of alkali such as KOH and NaOH. However, garnet exhibited excellent geopolymerisation in presence of 20% metakaolin and 30% sodium silicate	(Peng et al., 2015; Wang et al., 2019; Hua Xu & Van Deventer, 2000)
Gypsum	Calcium sulphate mineral reacts with water and leads to expansion., $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$	Percentage shall be limited to 3.5 – 4% in the cement. Gypsum also reacts with alkalis and participates in geopolymerisation	(Kiventerä et al., 2016; Simonsen et al., 2020)
Hematite	Hematite is an Iron mineral of formula Fe_2O_3 and is abundant on the earth's crust.	Hematite in an alkaline medium produced by sodium metasilicate and in presence of other minerals reacted to form kinoferrosilite. Hematite also exhibited a thermal stability up to a temperature of 950 °C.	(Buchwald et al., 2009; He et al., 2012; Krivenko et al., 2017)

		At temperatures greater than 1000 °C Hematite decomposed to produce magnetite and oxygen.	
Hydroboracite	Hydroboracite is a hydrated borate mineral of formula, $\text{CaMg}(\text{B}_3\text{O}_4((\text{OH})_3)_2 \cdot 3(\text{H}_2\text{O}))$	Hydroboracite decomposed at 600 °C of calcination	(Aydlın & Kızıltepe, 2019)
Illite	Illite is a type of phyllosilicate clay mineral	At 10% of NaOH, Illite was dissolved at a temperature of 550 °C and Na-rich sodium aluminium silicate was formed. The dehydroxylation of illite generally occurs between 600 °C and 800 °C but the presence of alkali elements enhances the release of H ₂ O by hydrolysis and then the completion of the dehydroxylation process, however this hydroxylation has smaller effect on crystallinity.	(Fernandez et al., 2011; Moukannaa et al., 2019)
Kaolinite	$\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$ or kaolinite is an alumina silicate crystalline mineral with Si/Al ratio equal to one. It in natural form shall act as an inert filler for supply of Al ³⁺ ions	Turns to amorphous metakaolinite phases on calcination 750°C. This is one of the most important minerals used in geopolymerisation, A study considered geopolymerisation of sixteen different minerals mixed with kaolin. All sixteen mixes indicated higher strength in case of kaolinite addition,	(Komnitsas & Zaharaki, 2007; McIntosh et al., 2015; Perumal et al., 2019; Hua Xu & Van Deventer, 2000)

		due to increased dissolution of alumina in the matrix. However, kaolinite on calcination at 950°C recrystallizes and reduces the reactivity	
Magnetite	Magnetite is an iron mineral with chemical formula, Fe ₃ O ₄	Magnetite exhibited lesser dissolution and reactivity in presence of alkali and phases were unaltered even after 28 days.	(Komnitsas & Zaharaki, 2007)
Mica	Mica is a group of phyllosilicate mineral.	It shall contribute to pozzolanic properties. The behaviour shall be improved by calcination till dehydroxylation	(Simonsen et al., 2020)
Montmorillonite	A soft phyllosilicate clay mineral system. On contact with water montmorillonite can swell many times its original volume	600-800 °C of calcination exhibited decrease in surface area and increased consumption of Ca(OH) ₂ . However calcined montmorillonite does not have much influence on the microstructure. However, montmorillonite on grinding to a nano size becomes amorphous and improves microstructure and reduces permeability of concrete	(Chang et al., 2007; Fernandez et al., 2011)
Mullite	Mullite is an alumina silicate mineral of formula Al ₆ Si ₂ O ₁₃	Mullite when formed in bricks and ceramics as a product of firing up to 1200 °C contributed to mechanical	(Chen et al., 2013; Cristelo et al., 2020)

		properties. However, in raw materials Mullite phases were found to be inert in presence of alkali.	
Muscovite	Phyllosilicate mineral of aluminium and potassium of formula $KAl_3SiO_{10}(OH)_2$	80 minutes of grinding reduces the crystallinity of muscovite by 37%. However, muscovite is identified to be mostly inert in case of alkali activation	(Cristelo et al., 2020; Yao et al., 2019)
Orthoclase	Orthoclase is a Feldspar mineral with formula $KAlSi_3O_8$	Orthoclase is capable of dissolution in alkali environment and hence geopolymerisation	(H. Xu & Van Deventer, 2002)
Palygorskite	Magnesium aluminium phyllosilicate clay mineral with water absorption capacity about 300% or more	Addition to cement leads to formation of interconnected fine pores.	(Lindgreen et al., 2008)
Pentlandite	Pentlandite is a sulphide mineral of formula $(Fe,Ni)_9S_8$	Pentlandite on oxidation leads to acid mine drainage	(Kossoff et al., 2014)
Phlogopite	Phlogopite is an end member of the biotite series of mica group $(KMg_3AlSi_3O_{10}(OH)_2$	The peaks phlogopite, decreased significantly when mixed with blast furnace slag, cement clinker and finely ground to a powder and mixed with warm water and cured at a temperature of $48\pm 1^\circ C$. Melting, quenching and grinding of phlogopite improved alkali activity by 40%. Calcination of phlogopite up to $1200^\circ C$ improved the	(Hartikainen & Helinä Hartikainen, 2008; Huang et al., 2012; Kinnunen et al., 2018; Sreenivasan et al., 2017)

		reactivity and at 1600°C was completely amorphous.	
Pyrite	Iron sulphide mineral (FeS ₂) in presence of water and oxidation leads to acid mine drainage	On hydration in presence of OPC pyrites produce ettringite and in presence of alkalis pyrites undergo alkali oxidation	(Argane et al., 2015; Kiventerä et al., 2016)
Pyrrhotite	Iron sulphide mineral of formula Fe _{1-x} S,	Leads to acid mine drainage on oxidation and is more reactive than pyrite	(Kiventerä et al., 2016; Sephton et al., 2019)
Quartz	Inert silicate mineral, SiO ₂	Mine tailings with high amount of quartz has been used as aggregates	(Kiventerä et al., 2020)
Rutile	TiO ₂ . More inert compared to anatase. Contributes to photocatalytic effect		(Zhang et al., 2017)
Sanidine	K- Feldspar K(AlSi ₃ O ₈)	Sanidine acted as a source of AL in alkali environment, however complete dissolution did not take place.	(Ahmari & Zhang, 2012, 2013)
Siderite	Crystalline Iron carbonate mineral	Transformed to magnetite on calcination	(Castro-Gomes et al., 2012; Obenaus-Emler et al., 2020)
Smithsonite	ZnCO ₃	Calcination leads to decarboxylation	(Argane, El Adnani, et al., 2016)
Talc	Talc is a phyllosilicate mineral	Milling more than 30 minutes led to reaggregation of particles.	(Hajimohammadi & van Deventer, 2016)

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