# Mine tailings-based geopolymers: Durability, microstructure, thermal and leaching properties

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## Abstract

The mining industry produces a considerable amount of stone waste and tailings, posing an ecological danger. This industrial waste is often disposed of via landfill, which leads to soil degradation and water and air pollution while obtaining valuable land. It can be recycled via a variety of methods, including the promising geopolymerization approach, which converts waste into value. This research investigates recent advancements in the production of geopolymer composites derived from industrial waste and mine tailings as a potential sustainable construction material. This research also provides in-depth analyses of the features and behaviours of mine tailings mixtures utilized in geopolymer production, such as their durability, microstructure, thermal and leaching capabilities. This study also reveals an information gap that must be addressed to progress mine tailings composites for cementitious materials.

## Keywords

Industrial Waste; Geopolymer; Mine Tailings; Durability, Microstructure, Thermal, Leaching

## 1. Introduction

Mine tailings collect in tailings ponds and mine waste dumps, and proper disposal of these wastes is becoming more important [1, 2]. This is owing to the rising output volumes of the metallurgical and mining sectors, as well as the absence of an acceptable way of disposing of the waste generated by these businesses, on the one hand. On the other side, it may be explained by the rising stringency of environmental legislation in the majority of affluent nations worldwide. Lead and mercury, radioactive elements, and other mining tails-related pollutants are actively released into the environment as a consequence of the building of tails, biota, contaminating soils, air, and water, and causing cancer in people. Pollutants from food production and feed waste wreak havoc on valuable farms and natural habitats. The operation of tailing dams increases the chance of man-made disasters happening [3, 4].

Furthermore, mine tailings should be seen as a mineral supply that has been removed from the earth's subsurface, transported, and misused from the perspective of rational natural resource management. The tailings may include trace quantities of target material as well as previously unclaimed components that can be reclaimed using more efficient mining processes [5-10], which is one reason for this viewpoint. The chemical composition of mine tailings, on the other hand, is primarily composed of silicon, aluminum, and calcium oxides, with a percentage ranging from 60 to 90% [1-4, 11, 12]. As a consequence, tailings have the potential to act as an alternate source for addressing a variety of building and industrial needs.[12-14].

A promising trend in the use of mine tailings seems to be the use of mine tailings as geopolymers and precursors of alkali-activated materials or aggregates. [15-17]. Geopolymers are materials that are largely made of amorphous sodium aluminum silicate hydrate. [18]. They are mostly solids that result from the interaction of an aluminosilicate powder and an alkali solution. [19]. According to van Deventer, et al. [18], the geopolymer network is composed of $AlO_4$ and $SiO_4$ tetrahedra connected by oxygen atoms [19]; Positively charged ions (e.g., $Ca_2^+$, $Na^+$, $K^+$, and $Li^+$) present in the cavity framework balance the negative charge. It is possible that using mine tailings as a geopolymer approach will not only slow down the accumulation of mine tailings and reduce the level of ecological contamination, but it will also combine the benefits of geopolymer technology associated with a reduction in carbon dioxide release into the environment, the possibility of utilizing other forms of aluminosilicate waste, and the versatility of geopolymer characteristics [20-24]. There has recently been a significant gain in knowledge across a varied set of professionals in the management of tails in common approaches. Over a dozen studies have been published documenting the efforts done to improve our knowledge of the geopolymerization processes of tails in order to manage the characteristics of geopolymers for applications such as pollution removal. [25-27], sustainable building [28-32], and another particular usage [13-17].

The mine tailings are inhomogeneous and have a complex mineral, aggregate, and chemical composition [11, 33-39]. Furthermore, although having relatively low quantities of valuable components, mine tailings contain hazardous and toxic compounds connected with waste products or mining activities [40-44]. All of these aspects make it more difficult to directly manage mine tailings in order to create geopolymers that fulfill environmental safety regulations in terms of impurity content while also obtaining the needed complicated functional qualities for the manufactured product. [45, 46].

As a consequence, addressing the challenges connected with the usage of mine tailings-geopolymer composites is particularly beneficial, both in terms of reducing the negative environmental effect and the promise of expanding the resource base of manufactured mineral raw materials. It is very advantageous to tackle the challenges associated with the usage of mine tailings-geopolymer composites. This review starts with a discussion of some of the physicochemical and environmental challenges surrounding the use of mine tailings-geopolymer composites. This study discusses mine tailings-geopolymer composites in depth, which is both a generalization and a detailed research of the relationship between their structural, mechanical, and thermal capacities, as well as their durability and other significant elements. Aside from the helpful properties of the development of the characteristics of mine tailings-geopolymer composites, we thoroughly address the well-known examples of their exploitation in prospective applications.

## 2. Durability properties

Only a few researchers have examined the long-term durability of mine tailings-geopolymer composites. With the help of Caballero, et al. [47], the gold mine tailings-geopolymer was exposed to sulfate and acid solutions as well as high temperatures. According to its findings, as compared to a reference cementitious composite, the rate of loss in compressive strength with immersion time in sulfuric and nitric acid solutions is pretty equal in gold mine tailings-geopolymer composites. Similar results have been seen in magnesium and sodium sulfate solutions, as well as when the solutions are exposed to high temperatures. Ahmari and Zhang [48] discovered that copper mine tailings-geopolymer composites submerged for 120 days in aqueous solutions with pH values ranging between 4 and 7 had a substantial drop (by 58–79% compared to reference specimens) in their plain compressive strength. The high initial Si/Al proportion and partial geopolymerization of the mine tailings, according to the scientists, were responsible for this result. Water absorption and weight loss, on the other hand, were quite minor and had lower values in comparison to the OPC-based binding agent. Another study by Ahmari and Zhang [49] showed that introducing cement kiln dust can improve durability and unconfined compressive strength. The beneficial impact of cement kiln dust was connected to improved aluminosilicate dissolving, production of calcium carbonate, and calcium incorporation into the geopolymer system. Falayi [50] demonstrated that activating with potassium aluminate results in a better resistance of geopolymers to alternate wetting and drying than potassium silicate. In every case, the UCS values dropped more than threefold after 10 wet and dry cycles [51-56]. This makes it difficult to use these composites in places where there is a lot of wet and dry time, and it also makes it important to look into ways to mitigate this.

The utilization of tailings to substitute natural aggregates (gravel or sand) in geopolymer concretes, either partially or completely, might lead to an upsurge in the water absorption and porosity of the latter [45, 46, 57-60]. In turn, this can make these substances more vulnerable to chemical assault, which can have a detrimental impact on their overall durability. Further investigation in this field is needed because of a lack of understanding about these and other characteristics of the durability of mine tailings-geopolymer composites, which suggests a need for future research in this area.

## 3. Microstructure properties

The microstructure of geopolymerization products; the content, structure, and proportion of the produced amorphous and crystalline phases; as well as the existence, distribution, and size of pores, are all useful factors in determining the attributes of mine tailings-geopolymer composites.

Falah, et al. [61] found that rising the sodium silicate content of a copper mine tailings-geopolymer composite by up to 30% densifies the microstructure of the material. It was also discovered that, at such a concentration of sodium silicate, almost the whole geopolymer is changed into fused rectangular prisms, which indicates a full transition to high alkaline conditions. Manjarrez, et al. [62] have discovered that when copper slag is put into its geopolymer, the density of the geopolymer rises as assessed by SEM image analysis. Its results show that copper slag increased the breadth of the amorphous peak in the XRD of copper mine tailings-geopolymer composites, whereas the crystalline peak in the copper mine tailings remained the same after geopolymerization, which is compatible with the SEM findings [63-67].

The XRD examination findings of its 28-day-cured geopolymer also reveal a lowering in the ferocity of crystalline peaks, suggesting that the dissolution of the Al and Si components in the geopolymerization process has progressed farther than previously thought. SEM pictures of copper mine tailings-geopolymer composites obtained in the work by Ren, et al. [68] show that raised aluminum sludge levels lead to the development of more geopolymer gels. In addition, they verified that there were no unreacted particles at an aluminum sludge concentration of 21% in their experiment. According to Ahmari and Zhang [49] investigation, as shown in Fig. 1, the enhanced microstructure of copper mine tailings-geopolymer composites is due to the incorporation of cement kiln dust, which leads to the creation of more geopolymer gels, as seen by an increased Si/Al ratio [69-74].

Due to the incorporation of iron mine tailings into fly ash-geopolymer composites, Duan et al. demonstrated that the geopolymer became denser by producing more C–S–H [75, 76]. They also analyzed the microstructure of their geopolymer after it had been subjected to elevated temperatures and discovered that it had suffered no considerable damage to its microstructure after seven heating cycles at 200 ºC. Increased numbers of pores and fractures were found after 800 ºC exposure in fly ash-geopolymer composites that did not include iron mine tailings, but this was not the case in fly ash-geopolymer composites that included iron mine tailings after the same exposure.

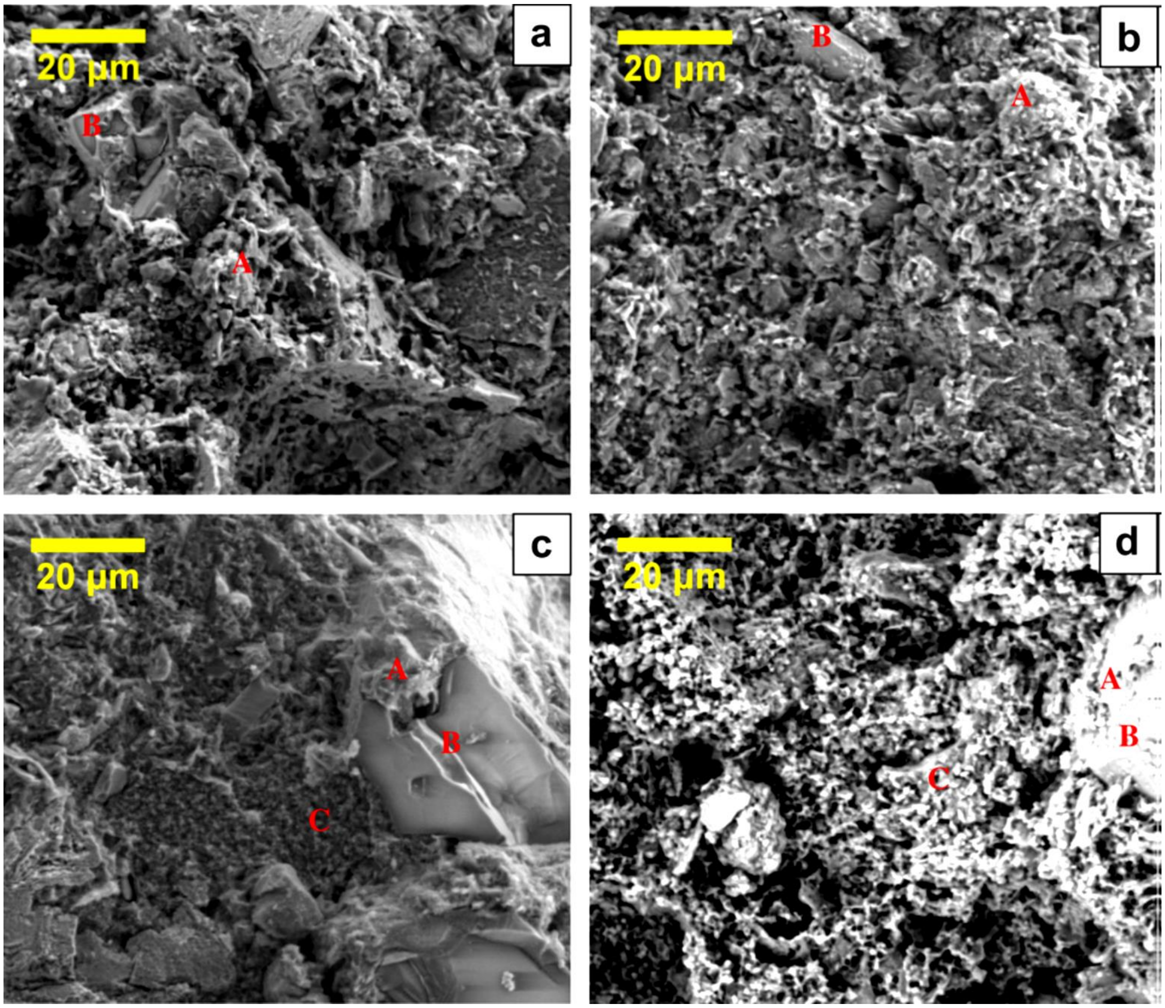


Fig. 1. SEM image of geopolymer brick samples made at 15 molarity NaOH, 16% water content, and cured for 7 days at 90 ºC: (a) 0% cement kiln dust, (b) 5% cement kiln dust, (c) 10% cement kiln dust, and (d) 10% cement kiln dust and after immersion in water for 7 days. (a and c indicate the binder stage, while b indicates the unreacted stage ) [49].

## 4. Thermal properties

As previously stated, geopolymers, in contrast to OPC binders, are recognized for their high thermal stability and the ability to retain strength even after being subjected to high temperatures [77, 78]. This is because of the unique characteristics of its structure, which is formed by branched $AlO_4$ and $SiO_4$ tetrahedral frameworks [77, 78]. The type of aggregate used to make geopolymers also plays a key role in the advancement of their thermal properties. This is because geopolymers can be made with several types of aggregates, such as aluminum-silicate aggregates. It should be noted that, when geopolymers have tails, a careful study of how these materials change and how well they work like insulation and fire-resistant materials is needed to figure out if they can be used [79-84].

Ye, et al. [85] investigated the impact of raised temperatures on the characteristics of a geopolymer made from bauxite tailings and slag. They discovered that the compressive strength of geopolymer is somewhat boosted after exposure to 200 ºC but that it rapidly reduces after exposure to 600 ºC. However, the drop in compressive strength was substantial between 600 and 1000 ºC, with a little gain in compressive strength at 1200 ºC. Anorthite ($CaAl_2(SiO_4)_2$), a type of ceramic, was discovered to be associated with an increase in strength, which could be attributed to self-healing and densification caused by sintering. The noticed drop in compressive strength at temperatures reaching 800 ºC is because of the dissolution of the amorphous stage as well as an extra thermal mismatch between the contracting gels throughout the contracting process. There is also physical harm in the form of cracking on the surface of samples. This is also in line with the findings of the compression experiment, which showed that there is no severe cracking on the sample when it reaches 400 ºC. It gets more violent as the temperature rises, so it starts at 600 ºC and goes up to 1200 ºC. Also, the width of micro-pores in its geopolymer gets bigger as the temperature of the material gets higher.

According to Jiao, et al. [86], the strength gain of mine tailings-geopolymer composites when subjected to high temperatures has also been reported. As a result of sintering, the geopolymers produced by the alkali-activated of vanadium tailings with high silica content demonstrated an improvement in compressive strength at temperatures above 900 ºC. This was accompanied by a lowering in the content of unreacted aluminosilicate precursor particles and the development of a denser microstructure by means of sintering, as shown in Fig. 2. As illustrated in Fig. 3, heating to 1000 ºC reduces bulk density and strength while increasing fracture and porosity. This effect was revealed to be caused by volume expansion and severe thermal incompatibility.

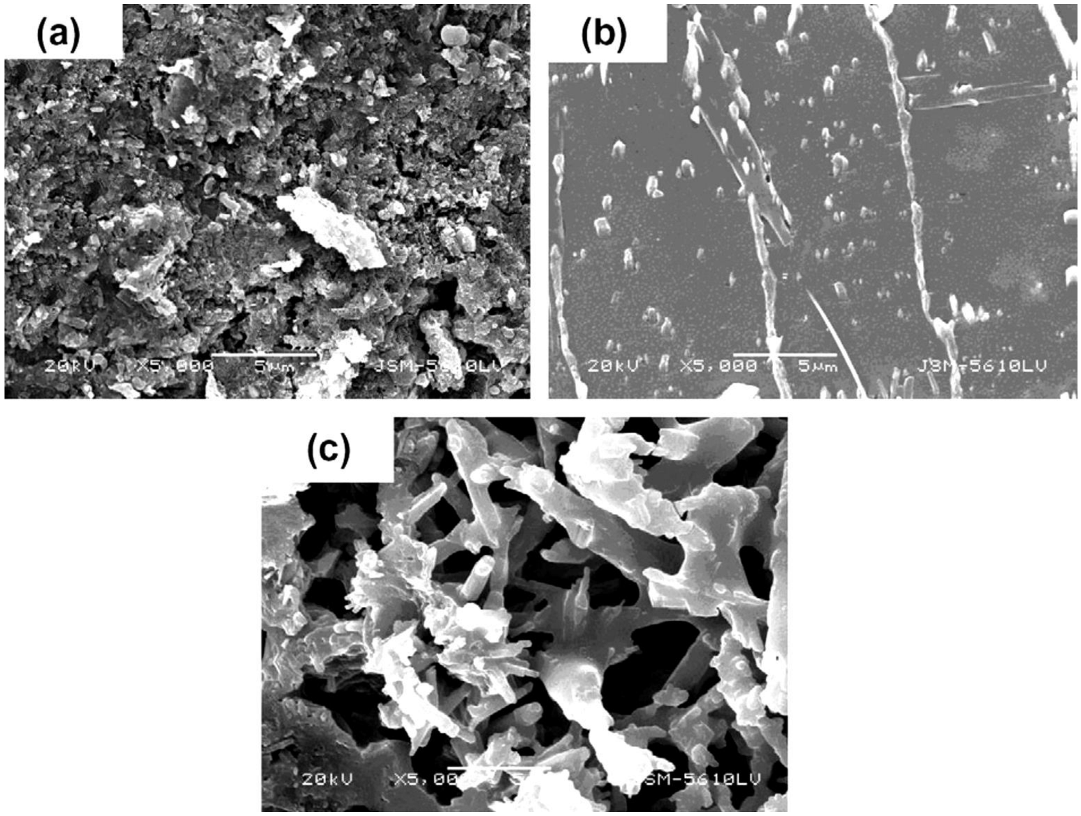


Fig. 2. SEM microanalysis of the geopolymer specimen: (a) ambient temperature; (b) at 900 ºC; and (c) at 1050 ºC [86].

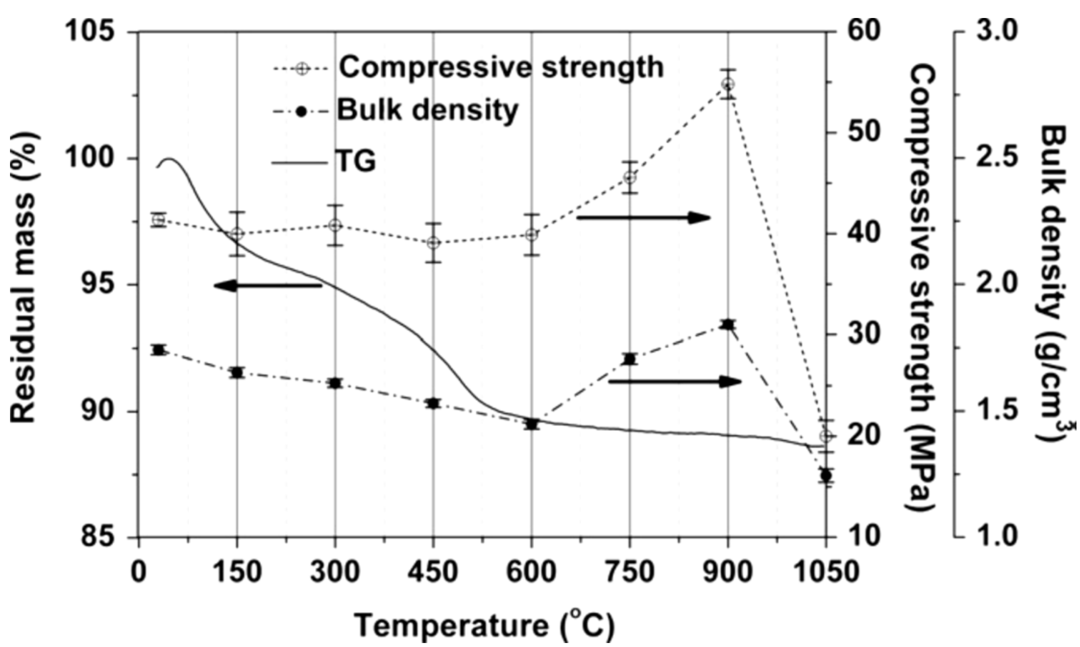


Fig. 3. Compressive strength, residual mass, and bulk density of the geopolymer specimen at high temperatures [86].

## 5. Leaching behavior

The presence of various heavy metals in mine tailings is a major environmental concern. To prevent their spread in soils and groundwater due to leaching, solidification (stabilization) through geopolymerization can be considered as one of the sustainable methods for neutralizing tailings containing toxic elements. In this regard, leaching characteristics are important indicators describing the effectiveness of heavy metal immobilization in geopolymers. As a result, making mine tailings-based geopolymers requires extra care when choosing the best ways and parts to make them [87-92].

The ability to successfully immobilize the heavy metals contained in lead-zinc tailings via physical and chemical ways was demonstrated by Zhao, et al. [93] in geopolymer based on coal gangue and blast furnace slag. Although an increase in tailings in prepared samples led to an increase in the concentration of $Zn^{2+}$, $Pb^{2+}$, and $Cd^{2+}$ in the leaching solution, these values remained within acceptable limits [93]. The obtained geopolymer samples were characterized by a compact structure, wherein the crystalline phase $Zn^{2+}$ was found; the amorphous phases were characterized by the content of $Pb^{2+}$ and $Cd^{2+}$.

Heavy metal cations can form chemical bonds with reactive components during polycondensation, which can lead to the formation of new phases. The formation of the PbO/BaSiO3 phase was observed by Hu, et al. [94] in rare earth tailing-based geopolymers. This is because $Pb2^{+}$ and $Ba2^{+}$ interact with unbridged oxygen or the Si/Al chain, which makes sure that the heavy metals stay in place inside the framework.

Ahmari and Zhang [49] reported no effective immobilization of arsenic and molybdenum due to geopolymerization in copper mine tailings-based geopolymers [48]. The authors also suggested a methodology to predict trace elements in geopolymers (Fig. 4). The experimental leaching data in their investigation correlates well with the proposed paradigm. Many studies have examined the efficiency of gold mine tailings-based geopolymers in immobilizing heavy metals [50, 95]. It is observed that the immobilization efficiency of Cr, Cu, Zn, Ni, and Mn in gold mine tailings, metakaolin, and slag blended geopolymer is higher than 98% with the only exception of arsenic and vanadium (Va), whose leaching is higher in that geopolymer [95].

In gold mine tailings-based geopolymers, the immobilization efficiency of heavy metals is higher in PA and KOH activated gold mine tailings geopolymers than in those synthesized by PS and KOH [50]. Kiventerä, et al. [96], Kiventerä, et al. [97] also reported effective immobilization of sulfate and arsenic in gold mine tailings-based geopolymer using calcium hydroxide and slag. After 7 days of curing, their geopolymer contains over 90% sulfate and over 95% arsenic, with other heavy elements immobilized as well. Wan, et al. [98], Wan, et al. [99]reported that lead (Pb) can be effectively immobilized in the mine tailings-geopolymer. They found that the formation of geopolymer gel in the binders is very important to the immobilization of Pb.

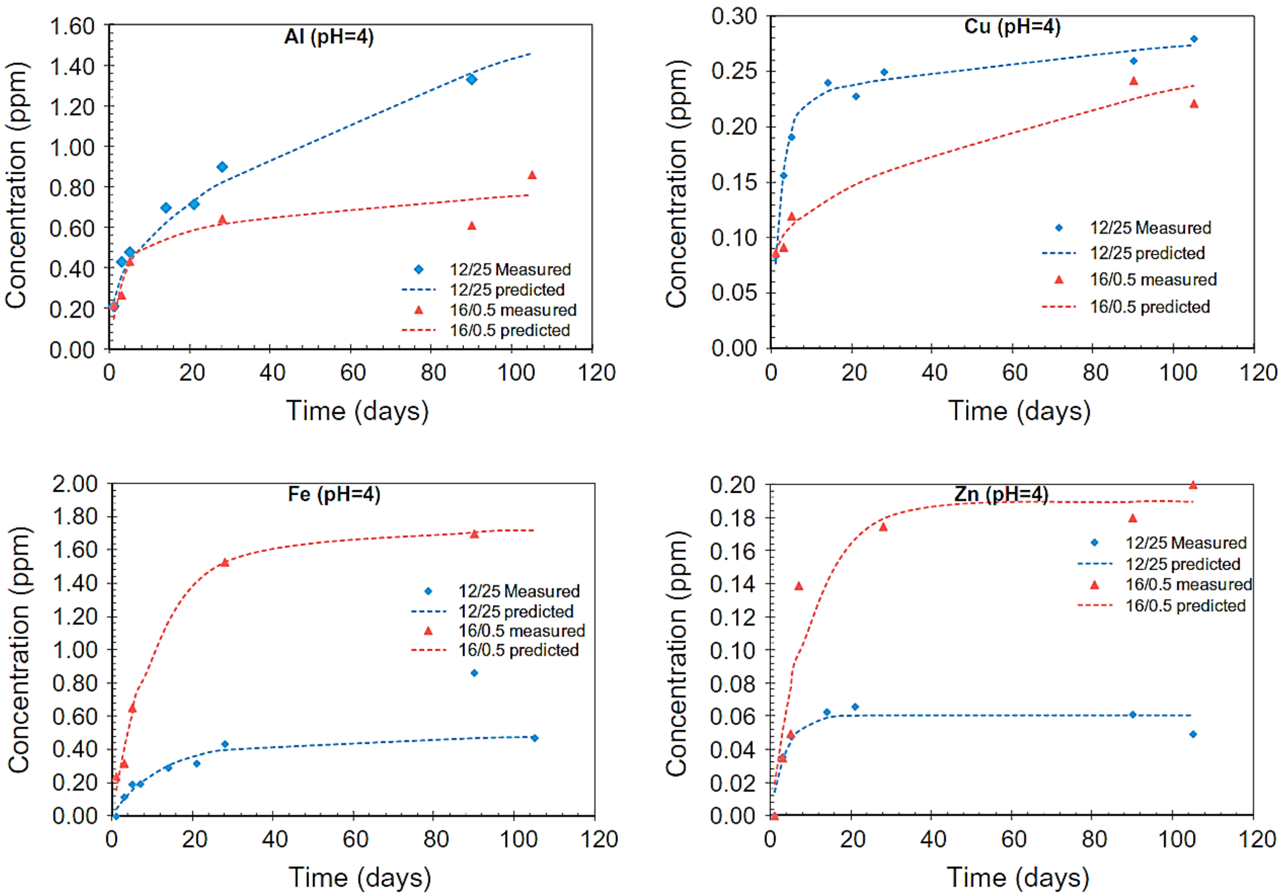


Fig. 4. Measured and predicted concentrations of heavy metals at pH = 4 a by first-order reaction/diffusion model (FRDM) [48].

## 6. Conclusions

The key annotations for this paper review are as follows:

1. According to the investigation, geopolymers seem to be attractive options for recovering mine waste and generating sustainable building and construction materials, mine paste backfills, and stabilizing materials for hazardous element immobilization. This strategy not only provides for a reduction in the carbon footprint associated with typical cementitious materials but also avoids the substantial ecological contamination produced by mine waste buildup.
2. Mine tailings are often composed of a highly crystalline matrix, which results in minimal interaction throughout geopolymerization and, consequently, a product with low mechanical characteristics. Incorporating extra elements with increased interaction into mine tailings-geopolymer composites may efficiently tune and enhance the characteristics of the geopolymers. Furthermore, since the majority of the additives utilized for this function are industrial by-products, their usage has the additional benefit of reducing the amount of waste produced.
3. When compared to low-Ca-comprising additions, high-Ca-comprising elements have a more favorable impact on the geopolymer's overall strength and durability. This is induced by the production of extra CSH gels, which strengthen the matrix as a result of its co-existence with NASH, which improves the matrix density.
4. Supplemental materials, especially those with a lot of calcium, tend to be better at making geopolymer characteristics.
5. The minerals that form mine tailings are identified by their varying chemical reactivity to alkali. The interactions of the precursors' metal components in alkaline conditions affect the structure and characteristics of the geopolymer's aluminosilicate framework. Many times, the alkaline reactivity of mine tailings is extremely low, which is the best thing when mine tailings are used to make geopolymers.
6. No classification strategy for mine tailings is in place that is based on its interaction. Recent research findings, like employing the topological technique to assess glass interaction, can be utilized to categorize and classify these materials, hence

encouraging their usage in geopolymerization applications.

## 7. Recommendations

The following are the main recommendations for future investigations:

1. The high silica concentration of mine tailings raises the molar proportion of $SiO_2$ /$Al_2O_3$ in mine tailings-geopolymer composites, impairing the process of geopolymerization. A solution to this difficulty can be found by including additional precursors, like metakaolin or scattered aluminum oxide, into the mix. A preliminary classification of tailings-based on the characteristics of their mineralogical and chemical compositions is recommended.
2. Because of the low interaction of native metal trichlorides, the presence of beneficial components ingrained in the minerals initially processed, and the risk of toxic contamination by leaching components, utilizing tailings for geopolymer preparation is prohibitively expensive and time-consuming from an economic and production standpoint. Aspects like the geographic closeness of the mining and processing enterprises to the mine tailings customers as well as the regions where finished geopolymer products are consumed should be taken into consideration when conducting a feasibility study for its application in geopolymer composites.
3. Pre-treatment of mine tailings can be utilized to boost their interaction. Therefore, further investigation is recommended in this regard.

## Conflicts of interest/Competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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