Review

Mine Tailings-Based Geopolymers: Durability, Microstructure, Thermal and Leaching Properties

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Abstract: The mining sector generates a substantial quantity of stone waste and tailings, which constitutes an environmental risk. The most prevalent method for disposing of this industrial waste is dumping, which contributes to soil deterioration and water contamination while acquiring precious land. It can be recycled using a number of processes, such as the promising geopolymerization technique, which transforms waste into value. This study reviews current developments in the manufacturing of mine tailings-based geopolymer composites from industrial waste as a possible sustainable building material. This paper also gives in-depth studies on the characteristics and behaviors of mine tailings composites used in geopolymer manufacturing, including durability, microstructure, thermal and leaching properties. This review also identifies knowledge gaps that must be filled in order to advance mine tailings composites for geopolymers.

Keywords: Mine Tailings; Industrial Waste; Geopolymer; Durability; Microstructure; Thermal; Leaching

1. Introduction

Mine tailings accumulate in tailings ponds and mine waste landfills, and the challenge of sustainable disposal of these wastes is becoming considerably more critical [1, 2]. This is due to the metallurgical and mining sectors' increasing production volumes, as well as the lack of an acceptable means for disposing of the waste created by these industries, on the one hand. On the other hand, it can be explained by the increasing stringency of ecological regulations in the majority of wealthy countries throughout the world. Lead and mercury, radioactive materials, and other mine tailings-related toxins are actively released into the environment as a result of the buildup of tailings, biota, polluting soils, air , and water , and causing cancer in humans . Pollutants from food processing and feed waste harm valuable farms and natural ecosystems . The functioning of tailing dams increases the likelihood of man-made catastrophes occurring [3, 4].

Furthermore, from the standpoint of rational natural resource management, mine tailings should be seen as a mineral source that has been extracted from the earth's subsurface, transported, and underutilized. The tailings can comprise trace amounts of target material as well as previously unclaimed elements that can be restored via more effective mining procedures [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 result, tailings have the potential to serve as an alternative source for meeting a wide range of construction and industry requirements [12-14].

A prospective trend in mine tailings use appears to be the use of mine tailings as geopolymers and precursors of alkali-activated materials or aggregates [15-17]. Materials composed mostly of amorphous sodium aluminum-silicate hydrate are called geopolymers [18]. They are primarily solids formed by 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

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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 potential of utilizing other forms of aluminosilicate waste, and the versatility of geopolymer characteristics as a general-purpose construction adhesion [20-24]. Recently, there has been a considerable increase in understanding among a diverse group of specialists in the management of tails in common methods . Over a dozen articles have been published detailing the efforts made to increase our understanding of the geopolymerization processes of tails in order to govern the properties of geopolymers for applications such as pollutant 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 factors make it more difficult to manage mine tailings directly in order to obtain geopolymers that meet ecological safety criteria in respect of impurity content while also achieving the essential complex functional characteristics for the manufactured product [45, 46].

As a result, tackling the issues associated with the use of mine tailings-geopolymer composites is especially useful, both in terms of limiting the negative impact on the environment and the prospect of growing the resource base of fabricated mineral raw materials. It is greatly beneficial to solve the problems linked with the use of mine tailings-geopolymer composites. This review begins with a discussion of some of the physicochemical and ecological issues surrounding the utilization of mine tailings-geopolymer composites. Mine tailings-geopolymer composites are discussed in length in this paper, which is both a generalization and a thorough investigation of the link between their structural, mechanical, and thermal capabilities, as well as their durability and other substantial aspects. Apart from the useful features of the formation of the characteristics of mine tailings-geopolymer composites, we discuss comprehensively the well-known cases of its utilization in promising 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

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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.

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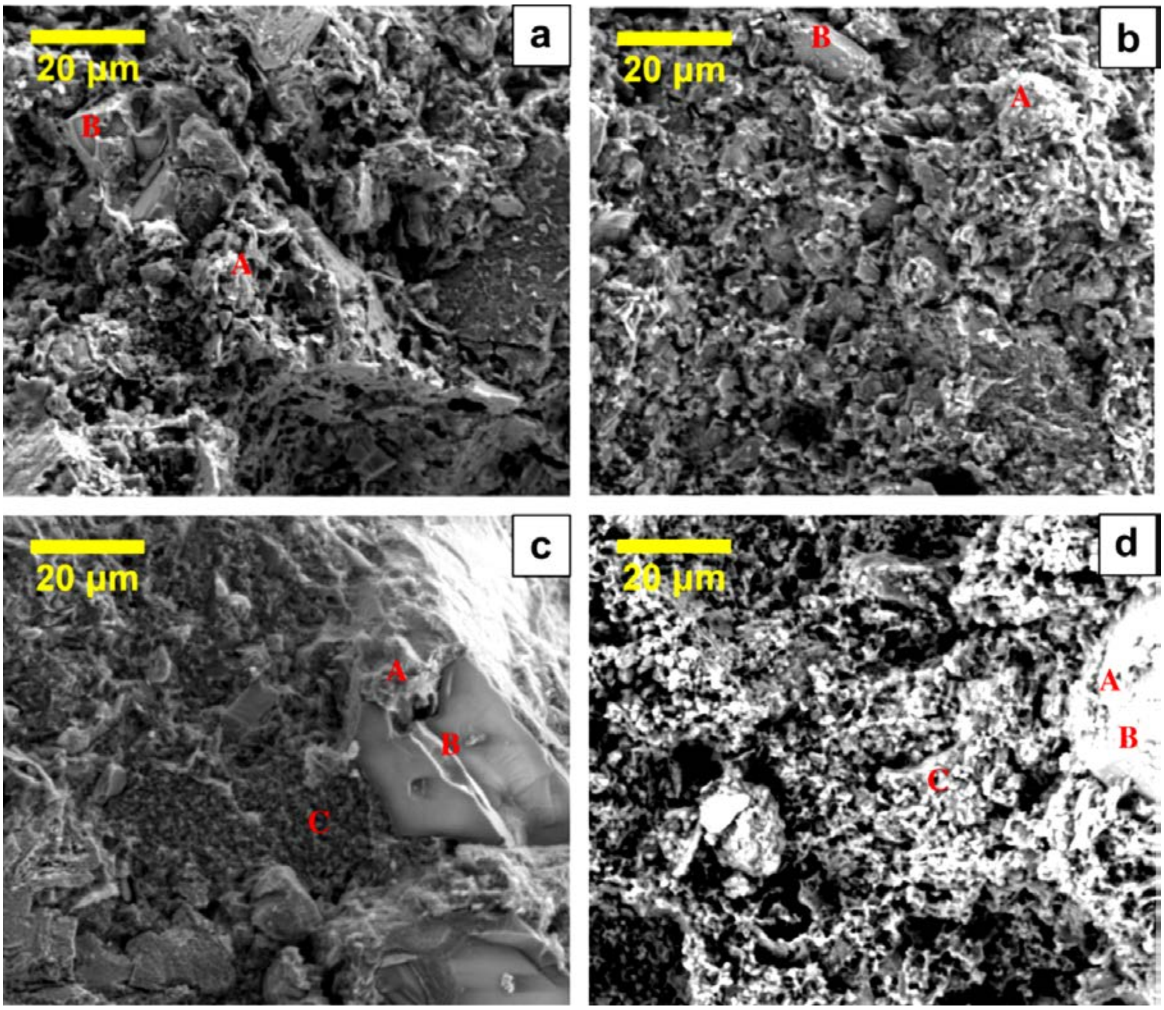

Fig. 1. SEM image of geopolymer brick samples made at 15 molarity NaOH, 16% water content, and cured for 7 days at 90 ºC: (a) 0% cement kiln dust, (b) 5% cement kiln dust, (c) 10% cement kiln dust, and (d) 10% cement kiln dust and after immersion in water for 7 days. (a and c indicate the binder stage,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

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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.

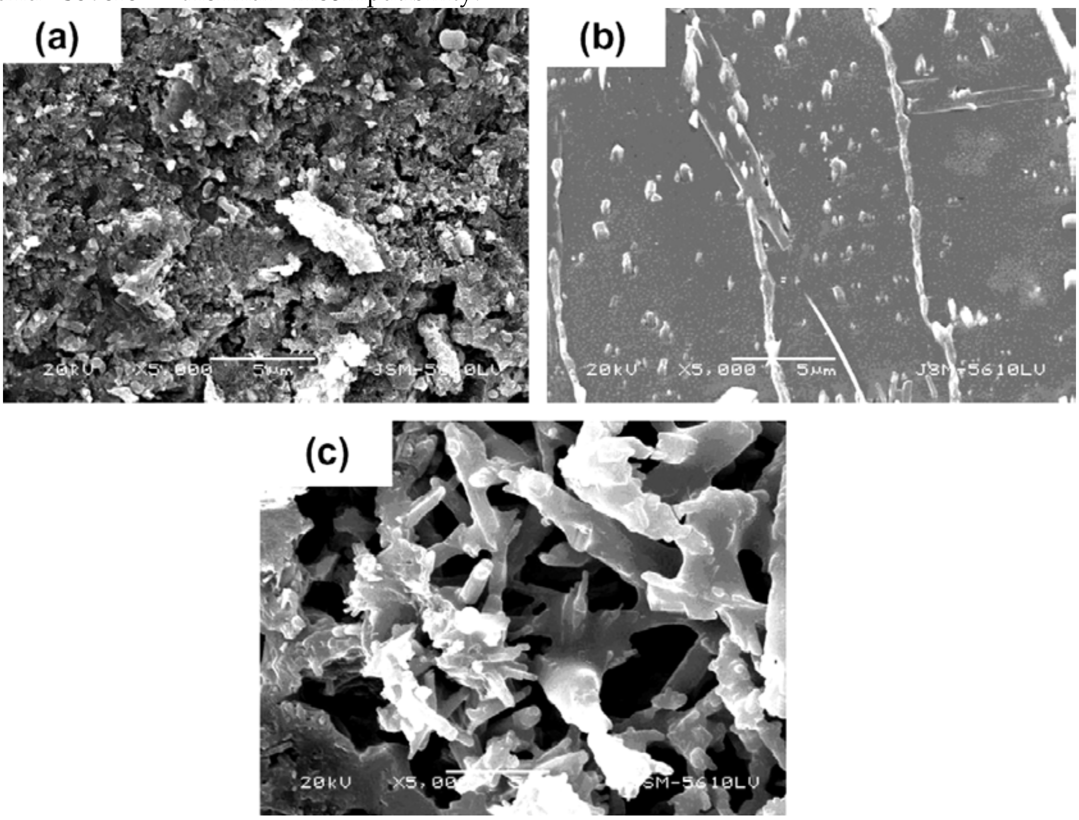

Fig. 2. SEM microanalysis of the geopolymer specimen: (a) ambient temperature; (b) at 900 ºC; and (c) at 1050 ºC [86].

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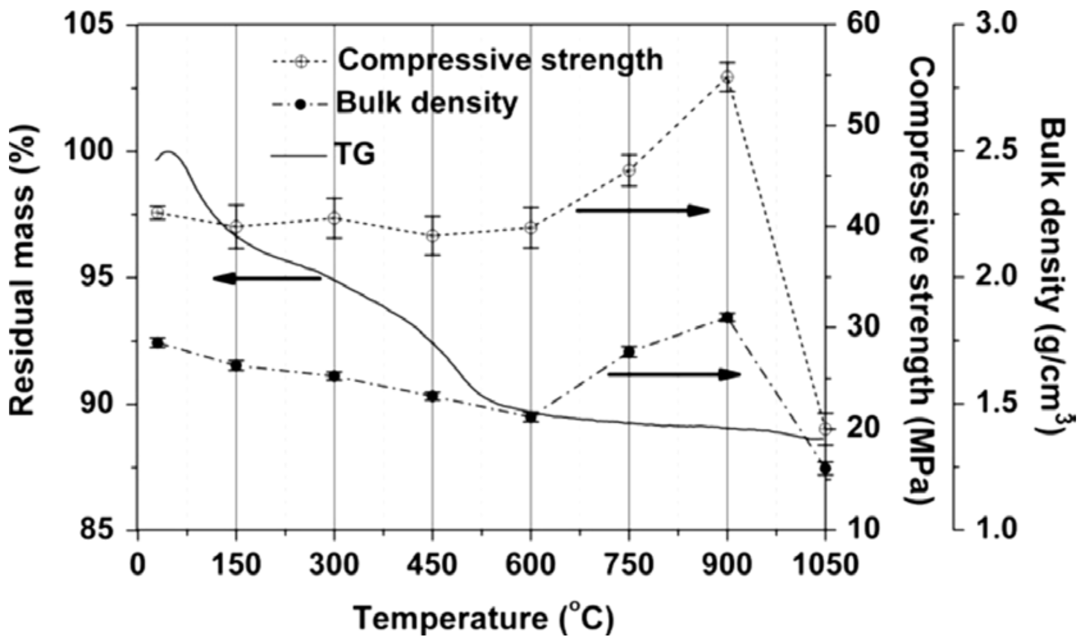

Fig. 3. Compressive strength, residual mass, and bulk density of the geopolymer specimen at high temperatures [86].

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.

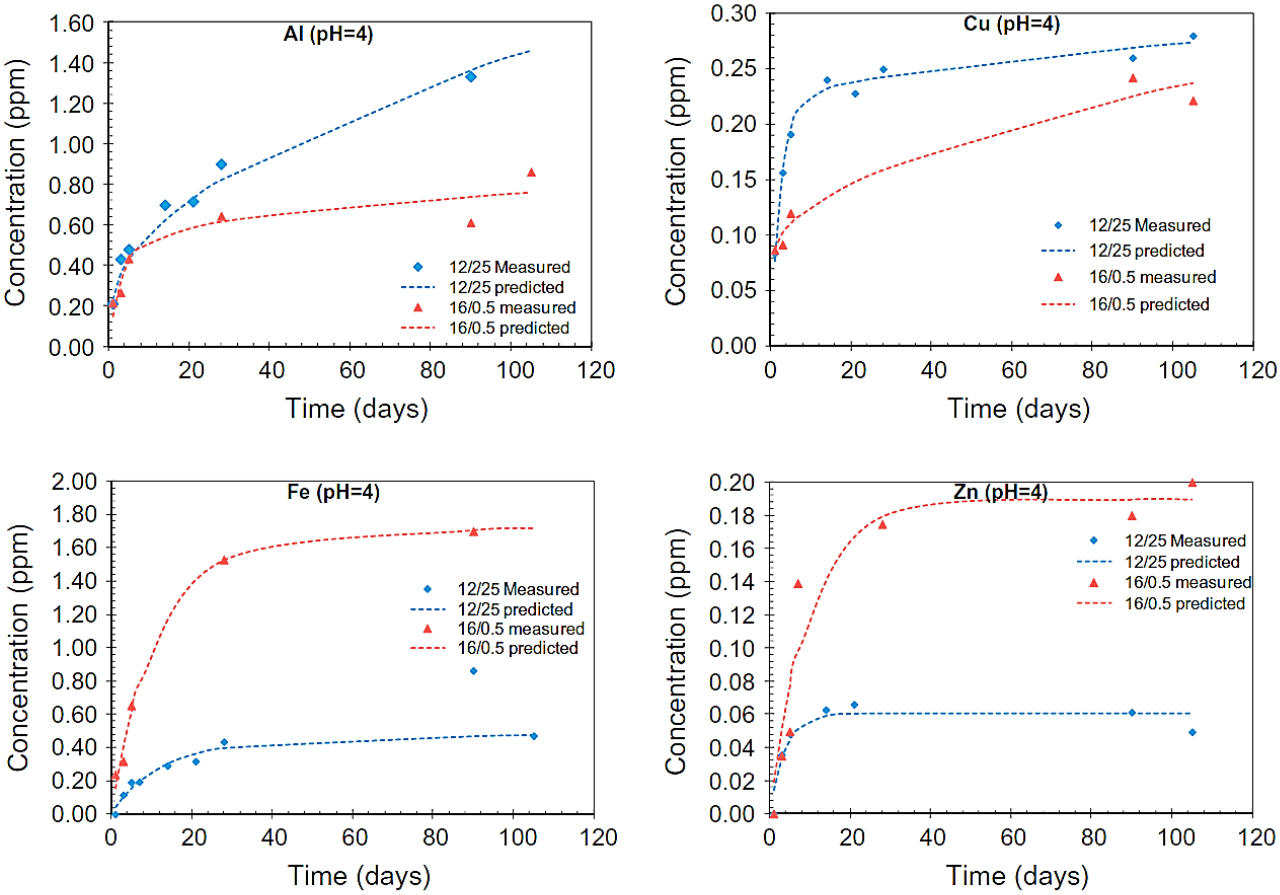

Fig. 4. Measured and predicted concentrations of heavy metals at pH = 4 a by first-order reaction/diffusion model (FRDM) [48].

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.

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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 con-

sumed 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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