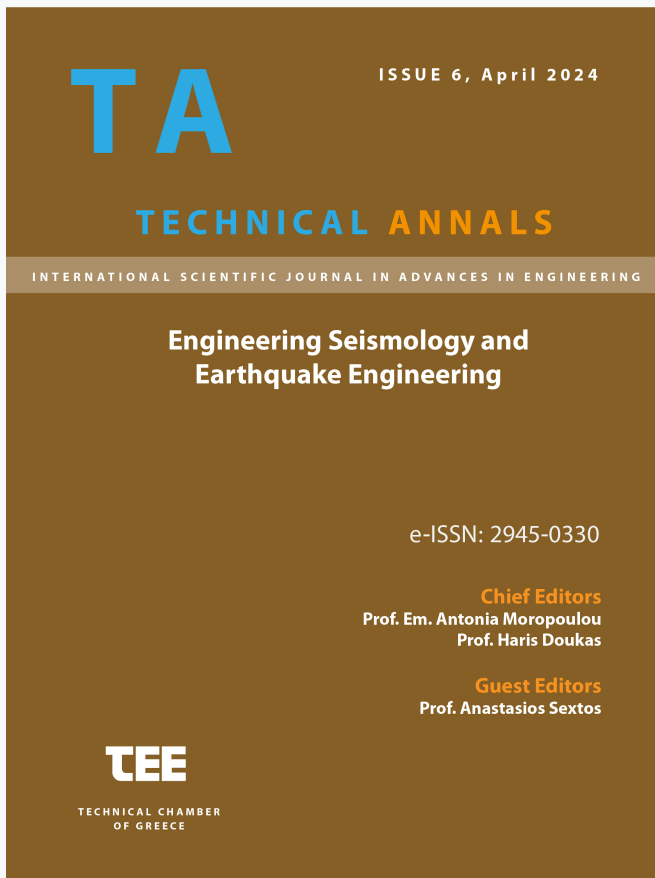


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Next Generation of Inorganic Composite Materials for Structural Strengthening: Development of Geopolymer Matrix

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Abstract. With the increasing need to strengthen seismically vulnerable structures, the use of composite materials, particularly textile-reinforced mortars (TRMs), has gained significant attention. In efforts to improve the mechanical properties of these materials while reducing their environmental impact, new alternatives are being explored. Geopolymer mortars, used as a matrix in composite materials, present a sustainable alternative to traditional cement-based mortars. The current study experimentally investigates the compressive and flexural strength of metakaolin-based geopolymer mortars, aiming to optimize their mix design. The properties of these geopolymer mortars are compared with commercially available cement-based mortars suitable for use as a matrix in TRMs. The study focuses on two key mix design parameters: the activator-to-precursor ratio and the sand gradation. Results indicate that the activator-to-precursor ratio significantly influences the strength and workability of the mortars, while the sand gradation primarily affects workability rather than strength. In comparison to the cement-based mortars studied, the geopolymer mortars demonstrated comparable, and in some cases superior, compressive and flexural strength.

Keywords: Geopolymers, Metakaolin, Composite materials, Structural strengthening.

1 Introduction

More than 88% of the Greek building stock was constructed before 2000, with nearly half of these buildings being constructed before 1970, according to the 2011 Building Census [1]. As a result, a significant portion of the existing buildings in Greece were designed and built using outdated construction methods, materials, and specifications, which do not meet modern standards for structural safety or environmental sustainability. This is particularly concerning given the high seismicity of Greece, where earthquakes pose a substantial threat to public safety. This combination of aging infrastructure and seismic risk underscores the urgent need for retrofitting and strengthening older buildings to ensure their safety and prolong their service life. As a result, the

demand for effective, sustainable strengthening methods has grown significantly, with the research community seeking to explore innovative materials and technologies that can provide enhanced performance while minimizing environmental impact.

Traditionally, one of the most widely used methods for strengthening existing buildings has been the application of externally bonded fibre-reinforced polymers (FRP). FRPs are known for their high strength-to-weight ratio and ability to enhance the structural integrity of buildings, particularly in seismic zones. However, while FRPs offer several advantages, their use is not without limitations, such as their vulnerability to environmental degradation over time, including UV radiation and moisture exposure. Recently, the use of textile-reinforced mortars (TRM) has been gaining popularity as an alternative or complement to FRPs in structural retrofitting. TRMs utilize an inorganic, usually cement-based, mortar matrix in combination with textile reinforcements, offering improved durability, fire resistance, and ease of application compared to traditional organic composites [2]. Despite the advantages of cement-based TRMs, the environmental impact of cement production—responsible for approximately 8% of global carbon dioxide emissions—has prompted a significant shift towards the search for more sustainable materials. In response to these environmental concerns, alkali-activated materials, also known as geopolymers, have emerged as a promising alternative to traditional cement-based binders [3]. Geopolymers are synthesized by activating aluminosilicate-rich materials, such as metakaolin, fly ash, and slags, with an alkaline solution, producing a highly durable and environmentally friendly binder. The use of geopolymer mortars has been expanding across various applications, including the repair and rehabilitation of structures, corrosion protection, and in environments subjected to high temperatures and aggressive chemical exposure [4-7].

The use of geopolymers in strengthening applications, such as reinforced concrete beams [8] and masonry [9-11], has shown promising results in comparison to conventional materials. The benefits of geopolymers include not only a reduced carbon footprint but also enhanced fire resistance, chemical durability, and mechanical properties such as high early strength. Despite these advantages, the widespread adoption of geopolymer mortars has been hindered by several challenges, primarily related to the lack of standardized mix design guidelines. The variability in the chemical composition of the precursor materials, such as metakaolin, fly ash, or slag, significantly influences the physical and mechanical properties of the geopolymer product, making it difficult to establish universal mix design standards [15].

Research into geopolymer mortars has intensified in recent years, driven by the need for sustainable construction materials and a growing understanding of their unique properties. One of the key factors influencing the properties of geopolymer mortars is the binder composition, particularly the activator-to-precursor ratio. Recent studies have demonstrated a clear correlation between activator content and the compressive and flexural strength of geopolymer mortars, with the optimal range of activator content leading to the highest mechanical performance [16-18]. The activator solution typically consists of alkali hydroxides or silicates, which are responsible for dissolving the aluminosilicate precursors and initiating the polymerization process. However, excessive activator content can lead to undesirable side effects, such as efflorescence, where salts

are deposited on the mortar's surface, which compromises both its aesthetic appearance and its mechanical properties [19].

Another important design consideration for geopolymer mortars is the type and gradation of sand used in the mixture. The gradation of the sand affects the mortar's consistency, packing density, and shrinkage behavior. Studies have shown that a well-graded sand mix, with a maximum nominal size of 1.18 mm, provides higher compressive and flexural strength compared to using either very fine or coarser sand fractions. The optimal sand gradation facilitates the formation of a dense, compact microstructure, contributing to enhanced strength and durability of the final mortar [20,21].

The aim of this study is to evaluate the mechanical properties of metakaolin-based geopolymer mortars and optimize the mix design for their use as a sustainable matrix in textile-reinforced mortars (TRM). The investigation focuses on optimizing key parameters, such as the activator-to-precursor ratio and sand gradation, and how these factors influence the strength, consistency, and workability of the geopolymer mortars. Additionally, the study benchmarks the performance of the optimized geopolymer mortars against commercial cement-based mortars commonly used in TRM applications. Through this study, the feasibility of geopolymer mortars as a sustainable and high-performance alternative to cement-based mortars for TRM applications will be assessed, with a focus on improving the longevity, safety, and environmental impact of retrofitted structures.

2 Experimental programme

In the current study, the mechanical properties of metakaolin-based geopolymer mortars were investigated to optimize the mix design for use as a matrix in textile-reinforced mortars. In the first phase of the investigation, the effect of the activator-to-precursor ratio was evaluated for five mortar mixtures (A1-A5). Based on the results from compressive and flexural strength tests, an optimal ratio was selected. In the second phase of the experimental program, the effect of limestone sand gradation on the strength and consistency of six geopolymer mortar mixtures (B1-B6) was examined. Finally, the geopolymer mortars were compared with various commercially available cement-based mortars used in strengthening applications (T1-T7).

2.1 Materials

For the preparation of the geopolymer mortars a metakaolin with a content of >92% in aluminium and silicon oxides and particle size distribution of $d_{10} = 2 \mu\text{m}$, $d_{50} = 30 \mu\text{m}$, $d_{90} = 100 \mu\text{m}$ was used. The activator comprised a potassium silicate solution with molar ratio between silicon and potassium oxide equal to 1.68 and total dry weight equal to 45%. Crushed limestone sand with maximum particle size 1 mm divided in 2 gradations, 0.5-1 mm and < 0.5 mm, was used as filler. 6 mm-long polypropylene fibres were also added at a volume fraction of 1% in each geopolymer mortar. A commercial cement-based mortar (T1) was experimentally investigated and consisted of a cement-based dry mix with fine sand of maximum particle size 1.3 mm and polypropylene fibres. The cement-based mortar was mixed with water according to the manufacture's

provisions. The examined geopolymer-based mortars are still under development; however, both metakaolin and the potassium silicate solution, the primary components, are commercially available.

2.2 Mix design optimisation

The mix design of the geopolymer mortars in the first optimisation phase is presented in Table 1. The activator to metakaolin (A:M) weight ratios chosen ranged between 1.2:1 and 2:1. Ratios below this range resulted in dense mixtures that were difficult to apply, while ratios exceeding this range led to highly fluid mixtures, which were deemed unsuitable for the current study. During this phase, the sand gradation ratio was kept constant. For the mixtures in the second phase, presented in Table 2, the gradation of the sand (F:P ratio) was investigated. The two sand gradations used were fine sand with particle sizes of 0.5-1 mm (F) and powdered sand with particle sizes of < 0.5 mm (P). In this phase, the A:M ratio was maintained at 1.2:1, as it was identified as optimal in the first optimization phase. To ensure adequate workability of the mortars, the total amount of sand was adjusted accordingly.

Table 1. Mix design of geopolymer mortars of the first optimisation phase.

Mortar ID	A:M ratio	Potassium silicate	Weight ratio		
			Metakaolin	Sand 0.5-1 mm	Sand < 0.5 mm
A1	1.2:1	1.2	1	1	2
A2	1.3:1	1.3	1	1	2
A3	1.5:1	1.5	1	1	2
A4	1.7:1	1.7	1	1	2
A5	2:1	2	1	1	2

Table 2. Mix design of geopolymer mortars of the second optimisation phase.

Mortar ID	F:P ratio	Potassium silicate	Weight ratio		
			Metakaolin	Sand 0.5-1 mm	Sand < 0.5 mm
B1	1:1	1.2	1	1.25	1.25
B2	1:1.5	1.2	1	1	1.5
B3	1:2	1.2	1	0.75	1.5
B4	1:2.5	1.2	1	0.6	1.5
B5	1.5:1	1.2	1	1.42	0.95
B6	2:1	1.2	1	1.77	0.88

2.3 Mortar preparation

A mechanical mixer was employed for the preparation of all mortars. For the geopolymer mortars, metakaolin and sand were initially dry-mixed for 1 minute. Subsequently, the alkali solution, to which polypropylene fibers had been added, was gradually incorporated into the dry ingredients. The mixture was then blended at medium

speed for 2–3 minutes until homogenized, followed by an additional 5 minutes of mixing at high speed. The preparation of the cement-based mortar (T1) followed a similar procedure. The cement-based binder was combined with water, which was added gradually at medium speed for 5 minutes, followed by mixing at high speed for a further 5 minutes. After mixing, all mortars were poured into molds in two layers to form prisms with dimensions of 40×40×160 mm, and subsequently vibrated to ensure proper compaction. The prisms were removed from the molds after 2 days and cured under ambient room conditions for a total of 28 days prior to testing. Representative prisms of both the cement-based and geopolymer mortars are depicted in Fig 1. To evaluate the flexural strength, each prism was subjected to 3-point bending, after which each half was tested under monotonic compression using a 40 mm cube, in accordance with EN 1015-11 [22].



Fig. 1. Typical prism of (a) cement-based and (b) geopolymer mortar.

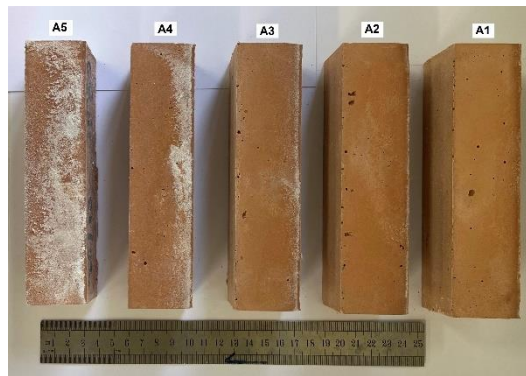
3 Results and discussion

3.1 Mix design optimisation

The results of the compressive and flexural strength of both optimisation phases are presented in Table 3. Based on the results of the first phase, mortar A1 exhibited the highest strength, with an A:M ratio of 1.2:1. Its flexural and compressive strengths were measured at 6.6 MPa and 38.3 MPa, respectively. Increasing the A:M ratio led to a progressive decrease in strength, with mortar A5 showing values of 3 MPa in flexure and 18.3 MPa in compression. The increase in activator content also improved workability, leading to a more fluid and less stable consistency in the mortars. However, excessive activator content caused efflorescence in mortars A3, A4, and A5 (as shown in Fig. 2) and led to a decrease in strength. The efflorescence was more pronounced with higher activator content and progressed over time. As a result, mortar A1 exhibited the best strength, and its A:M ratio of 1.2:1 was selected as the optimal mix.

Table 3. Strength results from the mix design optimisation.

Mortar ID	Flexural strength (MPa)	Compressive strength (MPa)
A1	6.61	38.27
A2	6.00	34.38
A3	4.93	26.14
A4	3.81	22.05
A5	3.04	18.32
B1	6.08	35.55
B2	5.30	35.00
B3	6.09	31.18
B4	5.17	29.35
B5	5.16	33.52
B6	5.02	33.47

**Fig. 2.** Mortars of the first optimisation phase with efflorescence.

As for the mortars in the second optimization phase, since the A:M ratio was already optimal, no signs of efflorescence were observed. The flexural and compressive strength ranged from 5 MPa to 6.1 MPa and from 29.3 MPa to 35.5 MPa, respectively. In this case, the effect of the sand gradation ratio (P:F) was not as significant as the A:M ratio. On the contrary, the total amount of sand, as well as its gradation, seemed to have a greater impact on the consistency of the geopolymer mortars. When more powdered sand (< 0.5 mm) was used, as in mortars B2, B3, and B4, the mortars were denser. In contrast, mortars B5 and B6, which contained more fine sand (0.5-1 mm), were more watery, granular, and had a tendency to segregate from the geopolymer paste. This is a deterrent for applications as a matrix in composite materials, as the mortar needs to adequately impregnate the fiber textile; thus, a well-graded mix is preferred. Based on both strength and consistency, mortar B3, with a F:P ratio of 1:2, was selected as the optimal mix. Mortar B1, which had similar flexural strength and slightly higher compressive strength compared to B3, contained more fine sand and could be considered a good alternative. However, B3 was chosen as the optimal due to its better consistency.

3.2 Comparison between cement-based and geopolymer mortars

Table 4. Strength results from cement-based mortars.

Mortar ID	Flexural strength (MPa)	Compressive strength (MPa)
T1	4.25	21.51
T2 [23]	9.80	39.20
T3 [24]	3.28	8.56
T4 [24]	4.24	30.61
T5 [25]	3.50	20.00
T6 [25]	8.30	40.00
T7 [25]	4.30	30.00

For comparison purposes, several commercial cement-based mortars used in strengthening applications were investigated and their strength is presented in Table 4. Mortar T1 is a common mortar used by the authors in strengthening applications, while mortars T2-T7 were used as a matrix in TRMs in other studies from literature [23-25]. It is evident that both optimised geopolymer mortars (A1 and B3) exhibited higher strength than mortar T1. Specifically, mortar A1 had higher flexural and compressive strength by 55.6% and 77.9%, respectively. Likewise, mortar B3 had 43.4% and 44%, higher flexural and compressive strength, respectively. Mortars T2-T7 had a flexural and compressive strength that ranged between 3.3-9.8 MPa and 8.5-40 MPa, respectively. It is noticeable that there is a significant variation in the results due to the different compositions of the cement-based mortars, with some also incorporating additive polymers to enhance strength (T2 and T4). When comparing the cement-based mortars with the geopolymer mortars A1 and B3, it is evident that the latter exhibited similar, and in some cases, higher strength. As shown in Fig 3, the optimal geopolymer mortars from both phases (A1 and B3) were able to achieve strengths comparable to the investigated cement-based mortars, surpassing their mean values for both flexural and compressive strength, which were 5.38 MPa and 27.1 MPa, respectively. Therefore, based on these preliminary results, geopolymer mortars could be a promising alternative to cement-based mortars in TRMs.

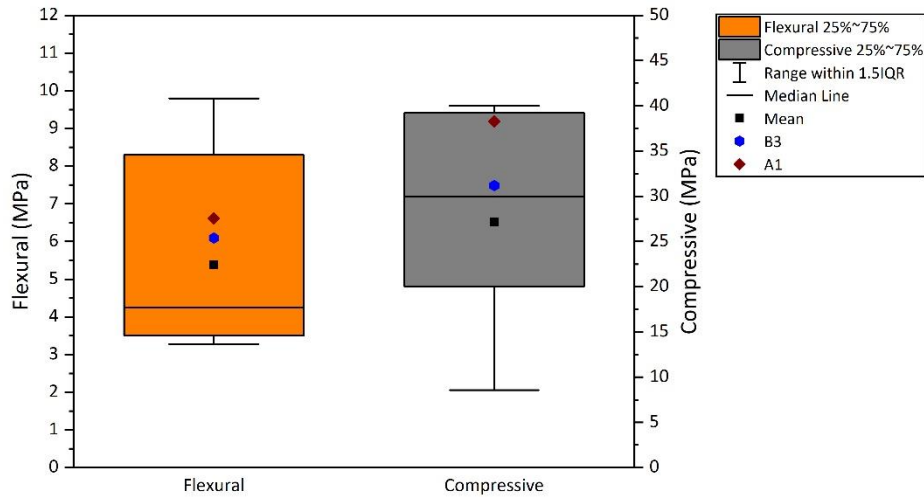


Fig. 3. Comparison between geopolymer and cement-based mortar strength.

4 Conclusions

In the current study the mechanical properties of geopolymer mortars with the intend to be used as a matrix in composite materials as an alternative to cement-based mortars were investigated. Two optimisation phases were conducted investigating the effect of the activator to precursor ratio and the gradation of sand ratio on the strength and consistency of the mortars. In the end a comparison of the optimal geopolymer mortars with various cement-based ones was carried out. Based on the results the following conclusions were drawn:

- The activator to precursor ratio (A:M) played a significant role on the strength of the geopolymer mortars, while the sand gradation ratio (F:P) affected more their consistency and workability.
- The experimental programme resulted in the selection of the optimal A:M and F:P ratios equal to 1.2:1 and 1:2, respectively, based on the raw materials used.
- The flexural and compressive strength of the optimal geopolymer mortar, which was measured equal to 6.1 MPa and 31.2 MPa, respectively, had similar strength to cement-based mortars.

Based on the above conclusions, it is evident that geopolymer mortars could be a promising sustainable alternative to cement-based mortars in TRMs. However, further investigation on the mechanical properties of the optimal mortar as well as its compatibility with various textiles should be carried out to evaluate its performance and find an optimal solution for strengthening applications.

It is also important to consider the cost implications. Cement-based mortars are priced at approximately 1 euro per kilogram, whereas geopolymer mixtures are estimated to cost around 1.5 euros per kilogram. However, it should be noted that geopolymer pricing is based on small-scale production, and bulk costs could potentially be

significantly lower, which may make geopolymers more economically competitive in the future.

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