

Comparative assessment of activation strategies for coal mine tailings as supplementary cementitious materials

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Abstract: The use of supplementary cementitious materials (SCMs) is an effective strategy for reducing cement consumption and associated CO₂ emissions. This study investigates the potential of coal mine tailings (CMT) obtained from a coal washing plant as an alternative SCM through selective activation methods. Three activation approaches were evaluated: mechanical activation, thermal activation at 600 and 800 °C, and pozzolan-assisted blending with fly ash and trass. Mortar specimens were prepared according to EN 196-1 with cement replacement levels of 5-20 wt.%, and compressive strength was determined after 28 and 90 days of curing. The results showed that mechanical activation reduced particle size but did not improve compressive strength. In contrast, thermal activation significantly enhanced the cementitious performance of CMT. The highest compressive strength, approximately 50 MPa, was achieved at 90 days with 10% CMT replacement after thermal activation at 800 °C, representing an approximately 100% increase compared with untreated CMT at the same replacement level. Pozzolan-assisted blending mainly improved long-term strength development. The binary blend containing 2.5% CMT and 2.5% trass achieved the highest 90-day strength among the pozzolan-assisted mixtures, reaching approximately 41 MPa and representing an increase of nearly 32% compared with the 5% untreated CMT mixture. Overall, the findings show that coal mine tailings can be transformed into effective SCMs through appropriate activation, particularly thermal treatment, providing a sustainable pathway to convert coal-processing wastes into value-added construction materials and support circular economy objectives.

Keywords: selective activation, coal mine tailings, supplementary cementitious materials

1. Introduction

Coal mining and coal preparation plants play a critical role in ensuring energy supply security; however, they also generate large volumes of fine-grained waste materials. Coal mine tailings (CMT) produced during beneficiation processes pose significant environmental challenges because of their high storage requirements, potential for acid mine drainage, heavy metal leaching, and dust emissions. These impacts make coal-derived wastes an important concern within sustainable mining and circular economy frameworks (Adiguzel et al., 2022; Acordi et al., 2023; Eker and Basetin, 2025; Adiguzel et al., 2026). At the same time, cement production contributes substantially to global CO₂ emissions and natural resource consumption. Therefore, valorizing such wastes in cementitious binder systems offers an opportunity to improve waste management and reduce the carbon footprint of the construction sector while supporting energy- and resource-efficient decision-making in coal processing systems (Zhang and Ling, 2020; Demir et al., 2024; Xu et al., 2025; Adiguzel Tuylu et al., 2026).

Coal-derived wastes constitute a heterogeneous group, including fly ash, coal gangue, slags, and washing tailings, with diverse mineralogical and chemical characteristics. Numerous studies have demonstrated that coal gangue and fly ash can be effectively utilized as supplementary cementitious

materials (SCMs) or as precursors in alkali-activated binders (Wang et al., 2024; Demir Şahin and Eker, 2024; Zhang et al., 2025). Nevertheless, the low initial pozzolanic activity and relatively inert crystalline structure of many coal-origin wastes limit their direct use as reactive binders, making physical and/or chemical activation necessary to enhance their binding potential.

Recent research highlights the effectiveness of mechanical, chemical (particularly alkali activation), thermal, and hybrid activation methods in enhancing the performance of coal-derived wastes in cementitious systems. Mechanical activation primarily refines particle size and increases specific surface area, which can promote pozzolanic reactivity and early-age strength development (Korkmaz and Kayıran, 2022; Bakil et al., 2025; Hu et al., 2024). However, its effectiveness depends heavily on waste-specific parameters, and excessive grinding may increase energy consumption without proportional performance gains (Wang et al., 2024; Xu et al., 2025).

Chemical activation studies have shown that the use of alkaline activators such as NaOH, sodium silicate, lime, or MgO promotes the formation of C-(A)-S-H and N-A-S-H type gels in coal gangue-slag-fly ash systems, leading to dense microstructures and compressive strengths exceeding 50-60 MPa at 28 days under optimized conditions (Guo et al., 2024; Kong et al., 2024; Xu et al., 2024). However, excessive alkali dosages may adversely affect hydration kinetics, reduce workability, and compromise long-term durability, underscoring the need for careful mixture design (Adesanya et al., 2021; Kumar et al., 2023).

Thermal activation, particularly in the temperature range of 600-800 °C, is widely reported as an effective approach for coal wastes containing kaolinite, illite, or smectite. Calcination within this range induces dehydroxylation and partial amorphization of crystalline phases, significantly enhancing pozzolanic reactivity and long-term mechanical performance (Zhang and Ling, 2020; Zhang et al., 2022; Zou et al., 2024). In contrast, temperatures above approximately 850 °C may cause recrystallization and vitrification, reducing reactivity and strength (Kong et al., 2024).

Hybrid activation strategies, combining mechanical, thermal, and chemical processes either sequentially or simultaneously, have recently gained attention due to their synergistic effects on hydration kinetics and microstructural development. Studies report that composite activation can yield activity indices exceeding 100% and outperform single activation methods in terms of strength and durability (Zhang et al., 2024; Li et al., 2025). Despite these promising findings, existing studies indicate that the binder performance of coal-derived wastes is highly sensitive to activation type and process parameters. Moreover, critical knowledge gaps remain regarding energy consumption, cost efficiency, environmental impacts, and long-term durability.

In particular, the literature reveals that: (i) systematic comparisons of different activation methods applied to the same coal waste source are limited; (ii) experimental studies focusing specifically on coal washing tailings are fewer than those on coal gangue or fly ash; (iii) synergistic effects arising from the combined use of activated coal wastes with other SCMs, such as fly ash or natural pozzolans, remain insufficiently explored; and (iv) standardized protocols addressing long-term durability, leaching behavior, and life-cycle performance are still lacking.

Motivated by these gaps, this study aims to evaluate the potential of fine-grained coal washing tailings from a single preparation plant as a supplementary cementitious material through selective activation approaches. The coal waste was subjected to (i) mechanical activation, (ii) pozzolanic-additive-assisted activation, and (iii) thermal activation. Cement mortars were prepared in accordance with EN 196-1 using 5-20 wt.% CMT replacement, and their compressive strength performance was assessed at 28 and 90 days. Additionally, selected mixtures incorporating fly ash and natural pozzolan were investigated to examine potential synergistic effects on early-age strength development. Through this approach, the study seeks to identify optimal activation-replacement combinations and provide practical guidance for designing sustainable, low-carbon cementitious binders based on coal washing tailings.

2. Materials and methods

2.1. Materials

2.1.1. Physical and chemical characteristics of CMTs

The coal mine tailings (CMT) used in the experimental study were collected from the waste disposal area of a privately operated coal company located in Uzunköprü, Edirne, Türkiye. This disposal area is

the chemical composition underlying these phase distributions, XRF analysis was conducted using a Rigaku ZSX Primus II spectrometer, and the results are summarized in Table 1.

Table 1. Major oxide composition of CMT determined by XRF analysis (wt.%)

Major oxides	CO ₂	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	K ₂ O	Na ₂ O	TiO ₂	SO ₃
Content (%)	35.93%	33.89%	13.41%	5.56%	3.46%	3.31%	1.76%	0.92%	0.49%	0.29%

The XRF results indicate that the coal waste has high aluminosilicate content and low calcium. For compositional evaluation, the gaseous/volatile components (including CO₂ and SO₃) and loss-on-ignition contributions were excluded, and the remaining oxides were subsequently normalized to 100%. Based on this normalization, the composition is dominated by SiO₂ (~54%), Al₂O₃ (~21%), and Fe₂O₃ (~9%), yielding a combined (SiO₂ + Al₂O₃ + Fe₂O₃) content of approximately 84%, while CaO remains below 10% (~5.5%) and MgO contributes ~5.3%. The total alkali content (Na₂O + K₂O) is around 4.3%. These values are further supported by simple compositional indices, including a silica modulus (SM) of ~1.8, an alumina modulus (AM) of ~2.3, and a low basicity ratio ((CaO + MgO)/(SiO₂ + Al₂O₃)) of ~0.14, confirming a strongly acidic, Al-Si rich system. Chemically, this composition closely resembles a low-calcium ASTM C618 Class F fly ash or calcined clay-type material, which is not directly hydraulic but highly suitable for pozzolanic and alkali-activated binder systems. The presence of moderate CaO and MgO (~10-11% combined) suggests favorable conditions for hybrid binding mechanisms involving both N-A-S-H and C-(A)-S-H gel formation. The exceptionally high CO₂ content is consistent with the presence of magnesian calcite and residual organic matter identified by XRD, indicating that thermal treatment is expected to induce significant mass loss and porosity development, thereby enhancing reactivity. Overall, the XRF-XRD compositional characteristics demonstrate that the raw CMT_0 sample represents a Ca-poor, aluminosilicate-rich waste material with high potential for selective thermal and mechanical activation, pozzolanic use, alkali-activated/geopolymer binders, and paste backfill or geotechnical improvement applications. In this context, Fig. 2 shows the TGA-DSC curves of the raw CMT_0 sample to elucidate the temperature-dependent decomposition and transformation mechanisms. Fig. 2 illustrates the TGA-DSC thermal behavior of the raw CMT sample under an inert atmosphere. The initial mass loss observed between 25 and 150 °C is attributed to the removal of free and physically adsorbed water, accompanied by a weak endothermic effect on the DSC curve, indicating moisture evaporation.

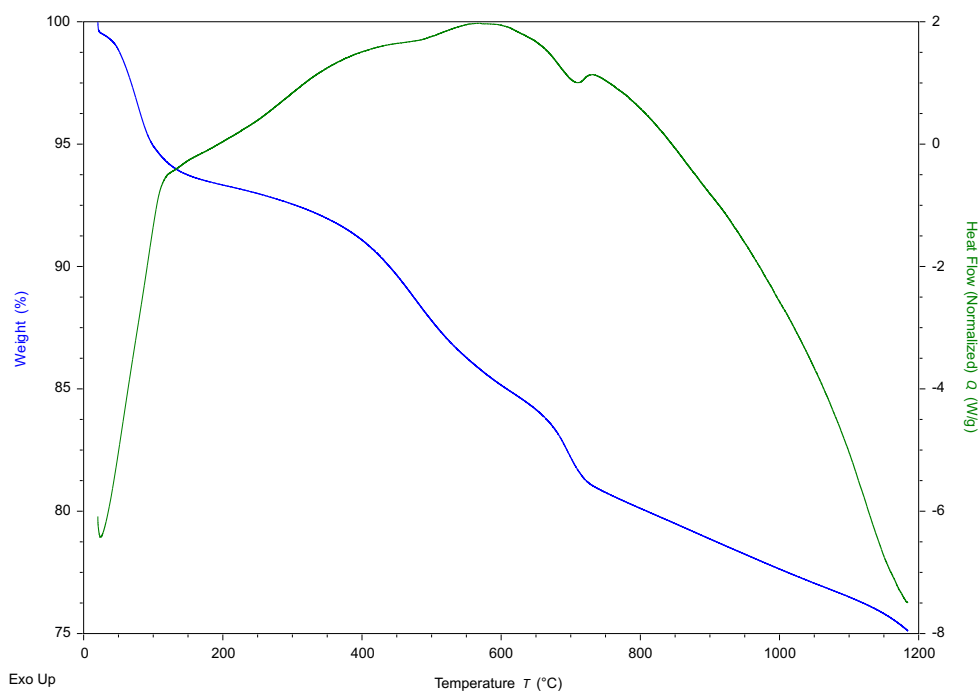


Fig. 2. Thermogravimetric (TGA) and differential scanning calorimetry (DSC) results of the raw CMT sample

This behavior confirms the presence of clay minerals with high water-retention capacity, such as smectite/montmorillonite. In the temperature range of 150-350 °C, a gradual increase in mass loss is detected, which is associated with the release of weakly bound interlayer water and the pyrolysis of residual organic matter inherent to coal waste. The absence of oxidative reactions under a nitrogen atmosphere supports this interpretation. The most critical transformation occurs between 350 and 650 °C, where a pronounced mass loss and a broad endothermic region are observed, corresponding to the dehydroxylation of clay minerals, including montmorillonite, kaolinite, and muscovite. This process leads to the formation of amorphous and highly reactive aluminosilicate phases, explaining the enhanced pozzolanic activity achieved through thermal activation at approximately 600 °C. A sharp mass loss in the 650-900 °C interval, together with a strong endothermic peak centered around 750-800 °C, is attributed to the decomposition of carbonate phases (CaCO_3 and MgCO_3) identified by XRF analysis, resulting in the release of CO_2 and the formation of CaO and MgO . This transformation accounts for the reduced CO_2 content in thermally treated samples (e.g., CMT_800T) and contributes to the development of a more reactive oxide-rich matrix suitable for alkali activation. Above 900 °C, mass loss becomes negligible, and the DSC signal exhibits broad fluctuations associated with structural rearrangement and the onset of sintering, indicating that further temperature increase may reduce reactivity due to crystallization and vitrification effects.

Thermal analysis provides insight into the temperature-dependent phase transformations and mass-loss mechanisms of the raw CMT sample; however, the microstructural characteristics governing its reactivity cannot be fully assessed solely through bulk thermal behavior. Therefore, scanning electron microscopy (SEM) was employed to examine the morphology, particle size distribution, and textural features of the untreated CMT_0 sample. The SEM observations aim to complement the TGA-DSC findings by revealing the physical structure of clay-rich agglomerates, particle packing, and surface characteristics that influence water retention, thermal decomposition, and subsequent activation behavior (Fig. 3). Fig. 3 presents SEM micrographs of the raw CMT_0 sample at 100 μm and 30 μm magnifications, revealing a heterogeneous and weakly bonded microstructure characteristic of untreated coal waste. At the 100 μm scale, the morphology is dominated by irregular, angular, and fractured particles, with a wide particle size distribution comprising coarse fragments (~20-60 μm) and finer fractions below 10 μm . This bimodal texture is consistent with the unground nature of the material and its composite origin, consisting of clay, silt, organic carbon, and carbonate phases.

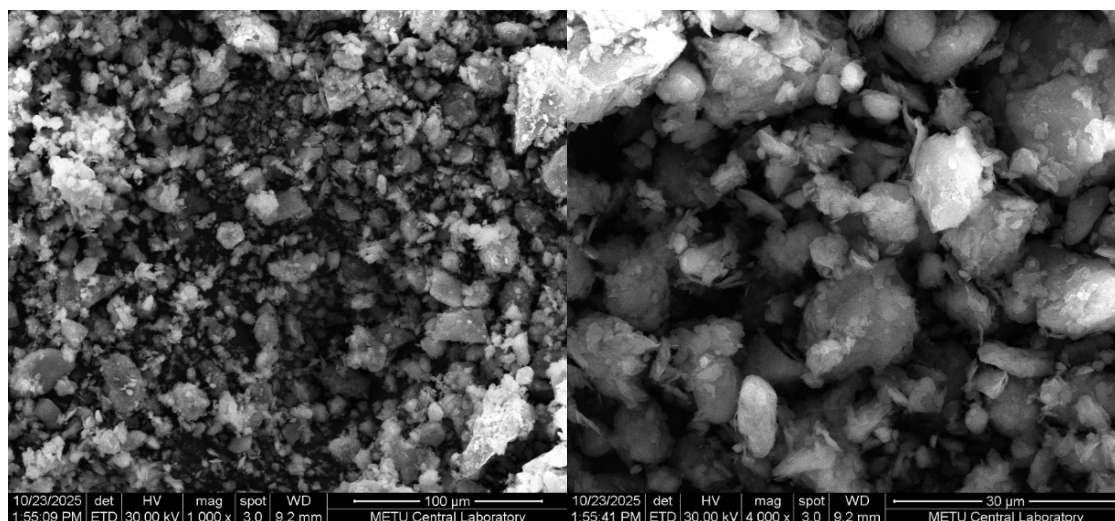


Fig. 3. SEM micrographs of the raw CMT_0 sample at different magnifications: (left) 100 μm and (right) 30 μm

The prevalence of sharp-edged particles reflects mechanical breakage during coal preparation processes, a feature typically preserved in non-thermally treated materials. Several particles show dull, rough, and darkened surface textures, indicating organic carbon residues. The EDS analysis was performed over the entire area of the right-side SEM image (30 μm) shown in Fig. 3. The analysis revealed a relatively high carbon content of approximately 23% (Fig. 4), supporting the presence of organic carbon residues observed in the SEM images.

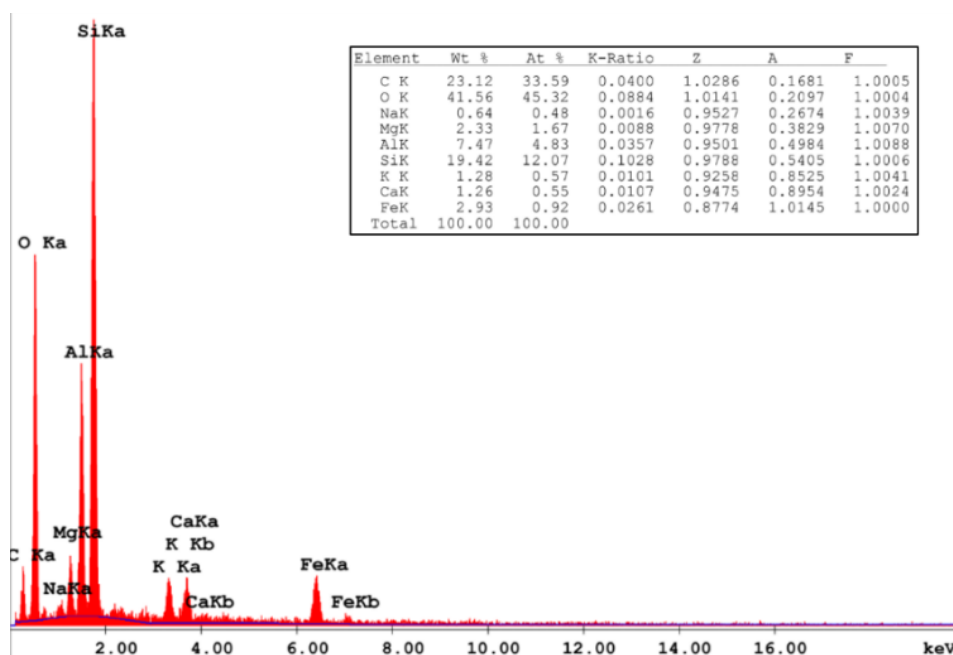


Fig. 4. SEM-EDS characterization of raw coal mine tailings (CMT)

Interparticle spaces are partially filled by a soft, fine-grained matrix attributed to clay-silt phases, representing the typical morphology of kaolinite- and montmorillonite-rich assemblages prior to thermal activation. At the higher magnification (30 μm), angular particles in the 20-40 μm range remain prominent, while the surrounding amorphous-looking matrix appears loosely packed and non-compact, confirming the absence of dehydroxylation and structural reorganization at this stage. Minor microcracks observed between particles are attributed to mechanical fragmentation rather than thermal stress. Overall, the observed microstructure indicates a low-cohesion and weakly reactive raw material, suggesting that significant morphological transformation and enhanced reactivity are expected following thermal activation in the 600-800 $^{\circ}\text{C}$ range.

2.1.2. Mixing materials

In this study, CEN standard reference sand, CEM I 42.5R Portland cement, fly ash (FA), trass, and mixing water were used as constituent materials in accordance with EN 196-1. The CEN reference sand is a rounded natural siliceous sand with a SiO_2 content exceeding 98% and a moisture content below 0.2%. The sand was supplied in pre-packed bags of 1350 ± 5 g with a standardized particle size distribution compliant with the relevant standards.

The performance of cement in mortar and concrete systems is governed not only by its chemical composition but also by its physical and mechanical properties. Therefore, the physical characteristics of the CEM I 42.5R cement used in this study were determined in accordance with EN 196, and the results are presented in Table 2.

Table 2. Physical properties of the CEM I 42.5R cement used in this study

Cement type	CEM I 42.5R
Specific gravity (g/cm^3)	3.12
Blaine specific surface area (cm^2/g)	350
Residue on 200 μm sieve (%)	0
Residue on 90 μm sieve (%)	2.5

The chemical compositions of the cement as well as those of the fly ash and trass used as cement replacement materials are summarized in Table 3. As shown by the chemical analysis, both trass and fly ash are rich in SiO_2 , Al_2O_3 , and Fe_2O_3 , indicating pronounced pozzolanic characteristics, while their

CaO contents are considerably lower than those of Portland cement. This compositional feature suggests that these materials can contribute to strength development in cementitious systems through secondary hydration reactions.

A comparison of the particle size distributions of cement, trass, fly ash, and coal mine tailings is presented in Fig. 5.

Table 3. Chemical compositions of cement, fly ash (FA), and trass (wt.%)

Component	SiO ₂	Fe ₂ O ₃	Al ₂ O ₃	K ₂ O	Na ₂ O	CaO	MgO	Cl ⁻	SO ₃	*LOI
Trass	56.28	7.90	16.08	1.21	2.03	6.03	3.79	0.0399	0.1	5.72
Fly ash	54.57	8.38	23.51	1.57	1.19	3.26	1.90	0.0191	0.72	2.5
Cement	19.22	3.49	4.59	0.85	0.28	63.63	0.92	-	3.21	-

*LOI: loss on ignition

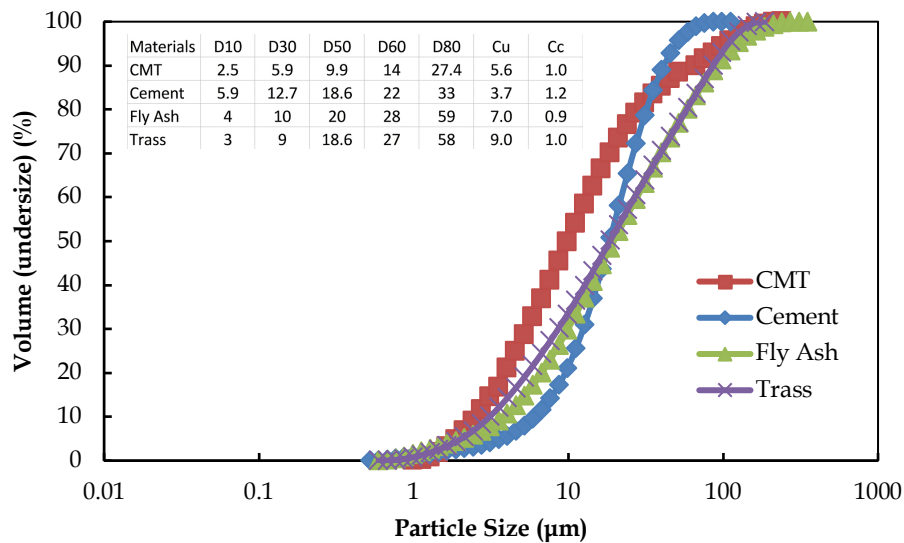


Fig. 5. Cumulative particle size distribution curves of CMT, trass, cement, and fly ash (FA)

Particle size distribution is a key parameter directly influencing specific surface area, water demand, reaction kinetics, and the workability and mechanical performance of the hardened product. Fine-grained materials, owing to their higher specific surface area, are generally more reactive and can enhance binding properties; however, excessive fineness may increase water demand and adversely affect workability. Coarser particles, on the other hand, contribute to the formation of a skeletal structure, playing a decisive role in strength development and pore structure. Therefore, the appropriate combination of materials with different particle size ranges is critical for achieving optimal packing density, mechanical performance, and workability. Mixing water conforming to EN 196-1, with a pH value between 6 and 8 and low chloride and sulfate contents, was used throughout the experimental program. The water quality was considered suitable and not detrimental to cement setting or strength development.

Table 4 presents the pozzolanic activity values of fly ash and trass. The higher activity index and k_p value of trass than fly ash indicate that trass can contribute more to binder performance when used as a cement replacement material. The k_p factor represents the degree to which a pozzolan can deliver mechanical performance equivalent to that of cement at a given replacement level and is commonly evaluated based on compressive strength results. In addition, the significantly higher reactive silica content of trass further supports its superior pozzolanic reactivity and helps explain the enhanced strength development observed in trass-containing mixtures.

2.2. Methodology

This study adopted an experimental methodology to enhance the binding performance of coal mine tailings (CMT), based on similar investigations reported in the literature (Fig. 6). The experimental

program consisted of four main stages: (i) preparation of reference materials, (ii) activation processes, (iii) mortar preparation, and (iv) evaluation of mechanical performance. All mortar preparation, curing, and mechanical testing procedures were conducted in accordance with EN 196-1.

Table 4. Pozzolanic activity values of fly ash and trass

Type of pozzolan	Activity values (MPa)	*k _p	Reactive silica (%)
Trass	8.46	6.0	25.25
Fly ash	6.05	4.8	0.3

*k_p: pozzolanic activity coefficient based on compressive strength

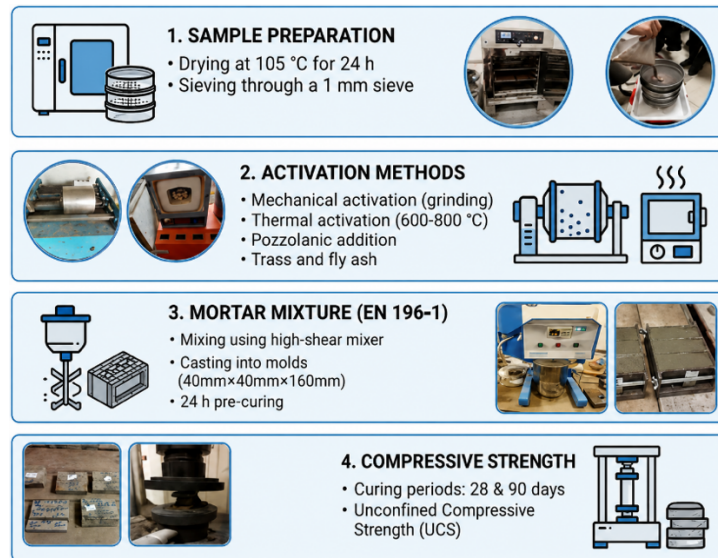


Fig. 6. Flowchart of the experimental methodology adopted in this study

2.2.1. Activation methods

2.2.1.1. Mechanical activation

For mechanical activation, CMT samples were ground in 500 g batches using a ball mill. Grinding durations of 30, 60, 120, 150, and 180 min were applied. The grinding process was carried out in a ball mill with a 20 cm diameter grinding bowl, operating at 60 rpm. A mixture of grinding balls with four different diameters was used, occupying approximately one-third of the mill volume. After grinding, particle size distributions were determined using a Malvern Mastersizer 3000 laser diffraction analyzer with a dry dispersion unit. The obtained distributions were evaluated using the coefficient of uniformity (Cu) and coefficient of curvature (Cc), as defined in ASTM D2487, and samples exhibiting optimal liberation characteristics were selected for mortar production.

2.2.1.2. Pozzolan-assisted chemical activation

In the pozzolan-assisted activation stage, fly ash (FA) and trass were incorporated to enhance the binding performance of CMT. Pozzolans were evaluated in binary and ternary combinations together with CMT as partial cement replacement materials. Replacement ratios were determined from the compressive strength results at 28 and 90 days for reference mixtures, and the total replacement level was fixed at 5 wt.% for both curing ages. Accordingly, mortar mixtures containing 2.5% CMT + 2.5% trass, 2.5% CMT + 2.5% FA, and 2.5% CMT + 1.25% trass + 1.25% FA were prepared and subjected to mechanical testing.

2.2.1.3. Thermal activation

Separate CMT samples were heated from room temperature to the target activation temperature (600 or 800 °C) in a metallurgical muffle furnace at a constant heating rate of 10 °C/min. Once the target

temperature was reached, the samples were held at that temperature for 1 h. The thermal activation process was carried out in ambient air. After thermal treatment, the samples were allowed to cool naturally inside the furnace to room temperature (furnace cooling) to avoid thermal shock and ensure phase stability. The thermally activated samples were then used to prepare and cast mortar. The effect of thermal activation was evaluated based on compressive strength results obtained after 28 and 90 days of curing.

2.2.2. Mortar preparation and curing

Prior to use, CMT samples were oven-dried at 105 °C for 24 h and sieved through a 1 mm mesh. Mortar mixtures were prepared with cement replacement levels of 5%, 10%, 15%, and 20% by mass. For each mixture, 1350 g of CEN standard sand, 220.5 g of mixing water, and 4.5 g of superplasticizer were used. Mortars were mixed in accordance with the mixing procedure specified in EN 196-1, using low- and high-speed mixing stages, and cast into 40 × 40 × 160 mm prismatic molds. After casting, the specimens were stored in molds for 24 h at 20 ± 2 °C and 90% relative humidity. The demolded specimens were then cured in a water curing tank for 28 and 90 days. At the end of the curing periods, uniaxial compressive strength tests were performed on the specimens. For each mixture, three prismatic specimens were prepared and tested, and the reported compressive strength values represent the average of three measurements. The variability of the results was evaluated using standard deviation.

3. Results and discussion

This section presents and discusses the compressive strength results obtained under three activation approaches: mechanical activation, pozzolan-assisted chemical activation, and thermal activation.

3.2. Effect of mechanical activation

The reactivity of pozzolanic materials in cementitious systems, and consequently strength development, is closely related to particle fineness, as finer particles provide a larger specific surface area that can accelerate pozzolanic reactions. Following mechanical grinding of coal mine tailings (CMT), the particle size characteristics and distribution curve classifications determined according to ASTM D2487 are summarized in Table 5.

Table 5. Percent finer than 150 µm and coefficients of curvature (Cc) and uniformity (Cu)

-150 µm	D ₁₀	D ₃₀	D ₅₀	D ₆₀	D ₈₀	Cu	Cc	Distribution curve
0 min.	2.7	5.3	9.8	12.7	30	4.7	0.82	Even graded
30 min.	2.13	4.5	8.7	12.7	42	4.7	0.76	Even graded
60 min.	2.13	4.5	8.7	12.7	42	6	0.76	Medium graded
90 min.	2.1	4	6.7	8.7	17	6	0.92	Medium graded
120 min.	2	4.3	8.7	12.7	24	6.35	0.73	Medium graded
150 min.	2	3.7	6	8.5	18.7	4.25	0.8	Even graded
180 min.	2	3.7	6	9	24.3	4.5	0.76	Even graded

As shown in Table 5, characteristic particle sizes (D₁₀, D₃₀, D₅₀, D₆₀, and D₈₀) systematically decrease with increasing grinding time up to 90 min, indicating significant particle refinement and a reduced proportion of coarse fractions. In particular, the reduction of D₅₀ to 6.7 µm and D₈₀ to 17 µm confirms that after 90 min of grinding, approximately 80% of the material reached substantially finer sizes. However, further grinding beyond 90 min (120-180 min) did not result in a notable additional fineness gain, and minor fluctuations in particle size parameters were observed.

This behavior can be attributed to the dominance of ultra-fine particles at prolonged grinding durations, which promotes particle agglomeration driven by van der Waals forces and electrostatic interactions. Although excessive grinding increases the nominal specific surface area, agglomerate formation may limit access to reactive surfaces and reduce the material's effective fineness. Bakil et al. (2025) also reported similar agglomeration behavior during prolonged mechanical activation of coal-derived waste. Moreover, ultra-fine and agglomerated particles may increase water demand because of

their increased surface area. This increased water demand can adversely affect the hydration process and potentially limit strength development. Accordingly, a grinding duration of 90 min was selected as an optimal balance between particle refinement and energy efficiency, as it maximizes fineness development while avoiding the adverse effects associated with excessive fineness, particle agglomeration, and inefficient water utilization.

Mechanically activated CMT ground for 90 min (CMT_MA) was then incorporated into mortar mixtures as a partial cement replacement at 5%, 10%, 15%, and 20%. Fig. 7 presents the uniaxial compressive strength (UCS) results after 28 and 90 days of curing, along with the corresponding results for untreated CMT (CMT_0). As shown in Fig. 7, mechanical activation did not improve compressive strength at either early or later curing ages. On the contrary, mortars incorporating untreated CMT (CMT_0) consistently exhibited higher UCS values than those containing mechanically activated CMT (CMT_MA) across all replacement ratios and curing durations. At 28 days, the mixture with 5% CMT_0 achieved a UCS of approximately 26 MPa, whereas the corresponding CMT_MA mixture reached only about 16-17 MPa, corresponding to a strength reduction of nearly 35% due to mechanical activation. A similar trend was observed at 10% replacement, where UCS values decreased from approximately 24 MPa (CMT_0) to 15 MPa (CMT_MA). Similarly, compressive strength decreased by approximately 37.5%. Although this reduction was not evident at the 15% replacement ratio, a comparable decrease was observed at the 20% replacement ratio at 28 days of curing.

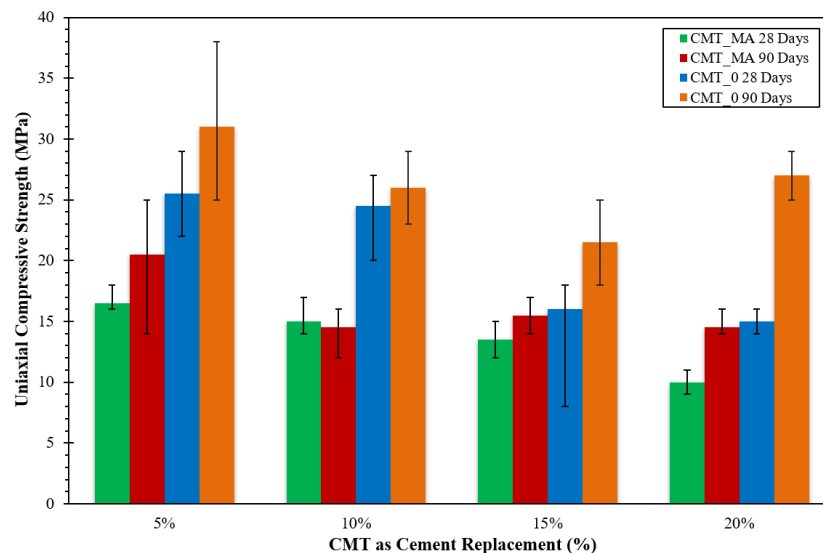


Fig. 7. Compressive strength results of mechanically activated CMT (CMT_MA, 90 min grinding) and untreated CMT (CMT_0) after 28 and 90 days of curing

At 90 days of curing, although all mixtures showed higher strength than their 28-day counterparts, untreated CMT mixtures maintained a clear strength advantage. In particular, at 5% replacement, the UCS of the CMT_0 mixture increased to approximately 31 MPa, while the CMT_MA mixture reached only about 21 MPa. In other words, the application of mechanical activation at the 5% replacement level resulted in an approximately 32% reduction in compressive strength. A similar trend was also observed at higher replacement levels. After 90 days of curing, mechanical activation reduced compressive strength from 26 MPa to 15 MPa at the 10% replacement level, from 21 MPa to 15 MPa at the 15% replacement level, and from 27 MPa to 14 MPa at the 20% replacement level. Overall, the untreated CMT mixtures exhibited higher strength performance than their mechanically activated counterparts. Consistent with the short-term results, these findings indicate that mechanical activation did not significantly contribute to long-term strength development.

This behavior can be attributed to excessive fineness and particle agglomeration induced by mechanical grinding, which likely increased water demand and restricted effective hydration, thereby offsetting the potential benefits of increased surface area. Similar findings have been reported in the literature, where excessive mechanical activation of coal-derived wastes was shown to limit strength

development due to agglomeration effects and unfavorable water-binder interactions (Zhang and Ling, 2020; Bakil et al., 2025).

As cement replacement increased from 5% to 20%, compressive strength gradually decreased in both CMT_0 and CMT_MA mixtures. For instance, the UCS of CMT_0 mixtures decreased from approximately 26–31 MPa at 5% replacement to around 15–27 MPa at 20% replacement, depending on curing age. This trend can be explained by the dilution effect associated with reduced active cement content in the binder system. At higher replacement levels (15–20%), disruption of paste continuity and weakening of the load-bearing skeleton further contributed to strength loss.

3.3. Effect of pozzolan-assisted chemical activation

Fig. 8 presents the compressive strength results of mortar mixtures prepared using untreated coal mine tailings (CMT_0) combined with natural and artificial pozzolanic materials at 28 and 90 days.

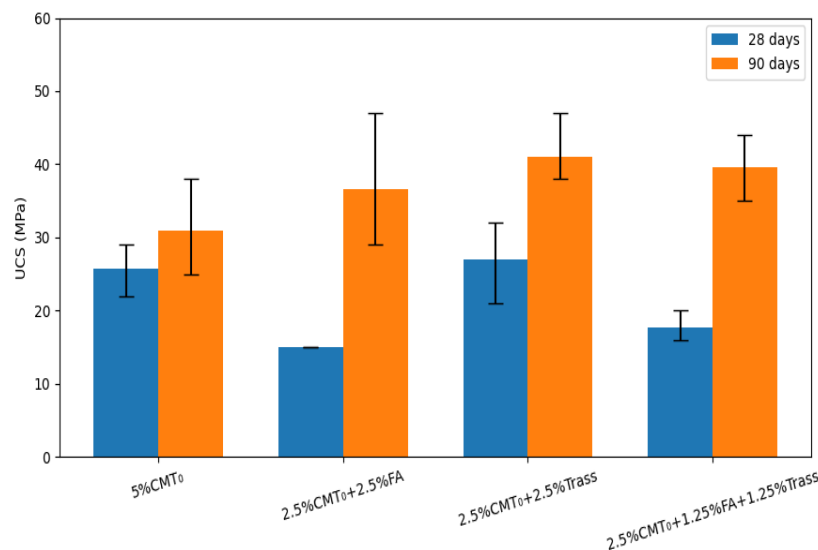


Fig. 8. Compressive strength results of mortars incorporating untreated CMT (CMT_0) and pozzolanic materials at 28 and 90 days of curing

Fig. 8 illustrates the UCS results of mortars containing untreated coal mine tailings (CMT_0) either alone or in binary and ternary combinations with fly ash (FA) and trass at curing ages of 28 and 90 days. At 28 days, the mixture with 5% CMT_0 achieved a UCS of approximately 26 MPa, higher than that of the binary and ternary mixtures containing pozzolanic additives. Specifically, the UCS values of the mixtures containing 2.5% CMT_0 + 2.5% FA and 2.5% CMT_0 + 1.25% FA + 1.25% trass were approximately 18 MPa, indicating that pozzolan incorporation did not enhance early-age strength under the investigated conditions. Among the pozzolan-containing mixtures, the binary blend of 2.5% CMT_0 + 2.5% trass showed the highest 28-day UCS, reaching approximately 27 MPa, indicating that trass contributed more effectively to early-age reactions than fly ash. These results are consistent with the pozzolanic activity values of the pozzolans used in the binary and ternary systems, as previously presented in Table 4. Indeed, the higher pozzolanic activity of trass supports the strength results obtained.

After 90 days of curing, all mixtures showed a pronounced increase in compressive strength, reflecting continued cement hydration and secondary pozzolanic reactions. Notably, mixtures incorporating pozzolanic additives outperformed the mixture containing only untreated coal mine tailings (CMT_0) at the same total replacement level. While the UCS of the 5% CMT_0 mixture increased to approximately 31 MPa, the binary mixture containing 2.5% CMT_0 + 2.5% trass achieved the highest UCS value of about 41 MPa, corresponding to an increase of nearly 32% compared to the CMT_0 only mixture. This superior performance can be attributed to the higher content of reactive amorphous silica in trass, which actively participates in sustained secondary pozzolanic reactions with calcium hydroxide

released during cement hydration, promoting the formation of additional C-(A)-S-H gel and a denser microstructure.

The ternary blend (2.5% CMT_0 + 1.25% fly ash + 1.25% trass) followed closely, achieving a UCS of approximately 39–40 MPa, indicating a synergistic contribution of the two pozzolanic materials. In contrast, the mixture containing 2.5% CMT_0 + 2.5% fly ash reached a UCS of around 36 MPa, reflecting the comparatively lower long-term contribution of fly ash under the applied curing conditions. This behavior is associated with the delayed pozzolanic reactivity of fly ash and its reduced availability of reactive silica at later ages, resulting in less pronounced formation of secondary hydration products than with trass.

The superior performance of trass containing mixtures can be attributed to the higher pozzolanic activity and reactive silica content of trass, which promote sustained secondary hydration reactions and contribute to the formation of strength-giving C-(A)-S-H type gels. Similar trends have been reported in the literature, where natural pozzolans rich in reactive silica were shown to significantly enhance late-age strength in blended cementitious systems (Zhang and Ling, 2020; Kong et al., 2024).

Overall, Fig. 8 shows that untreated CMT_0 can be used as a partial cement replacement without compromising early-age strength at low replacement levels. However, combining it with natural pozzolans, particularly trass, yields substantially superior mechanical performance at later curing ages. In this context, binary and ternary pozzolanic blends offer a viable, sustainable strategy to reduce cement consumption while maintaining or enhancing mechanical performance.

3.4. Effect of thermal activation

Fig. 9 presents the SEM images of the CMT samples thermally activated at 600 °C at two different magnifications. Fig. 10 shows the SEM-EDS analysis results of the CMT sample thermally activated at 600 °C. The SEM images show a marked reduction in dull, coarse, and dark-colored particles compared with the raw CMT sample. This observation indicates that thermal activation substantially removed organic residues. This is also consistent with the decrease in carbon content determined by EDS analysis; the C content decreased from approximately 23% in the raw CMT sample to around 10% after thermal activation at 600 °C (Fig. 9). The TGA-DSC results indicate that the mass loss occurring during heating may partly be associated with the thermal decomposition of residual organic matter.

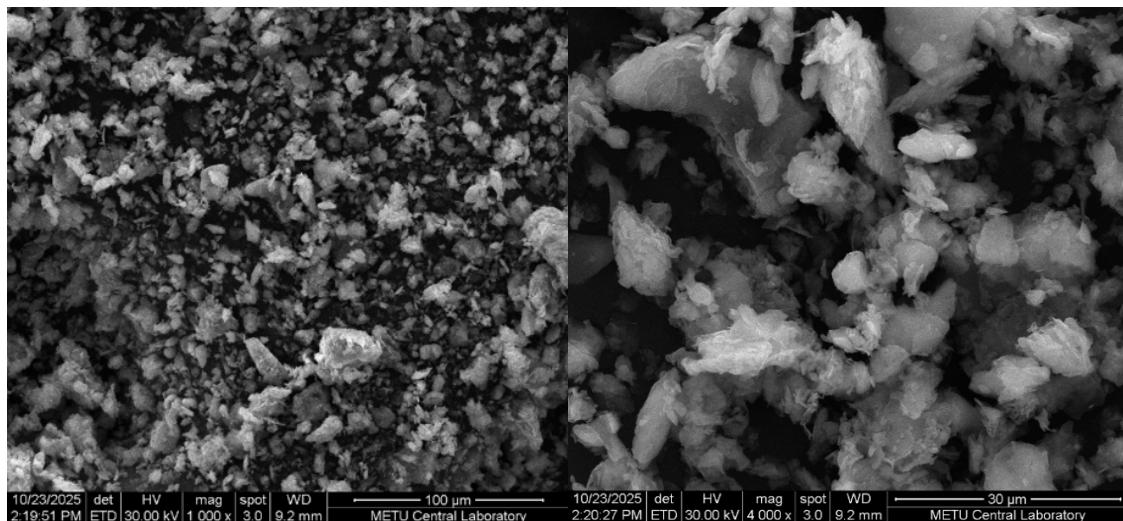


Fig. 9. SEM micrographs of the raw CMT_600 sample at different magnifications: (left) 100 μm and (right) 30 μm

The SEM images also reveal smaller granular particles, some of which appear locally agglomerated. In particular, finer and more granular particles in the range of 5–20 μm became more evident. This suggests that thermal activation at 600 °C caused fragmentation and disintegration of the CMT particles. These morphological changes may be associated with the initial breakdown of the crystalline structure of clay minerals, dehydroxylation, and the resulting shrinkage behavior. This structural transformation may indicate enhanced pozzolanic reactivity of the material. In some regions, small particles formed

granular clusters. This can be attributed to the partial shrinkage and adhesion of the clay-silt phases induced by thermal treatment. Compared with the SEM images of the raw CMT sample, more continuous and finer interparticle voids were observed. This pore structure indicates the development of secondary microporosity resulting from the combustion of the organic phase and the shrinkage of clay minerals. Such microstructural changes have been reported to increase pozzolanic reactivity. In addition, unlike the distinctly layered and rough morphology observed in the raw CMT sample, the particle surfaces appeared duller. This may be interpreted as a morphological indicator of dehydroxylation in clay minerals.

Fig. 11 shows the SEM images of the CMT samples thermally activated at 800 °C at two different magnifications. Fig. 12 shows the SEM-EDS analysis results of the CMT sample thermally activated at 800 °C.

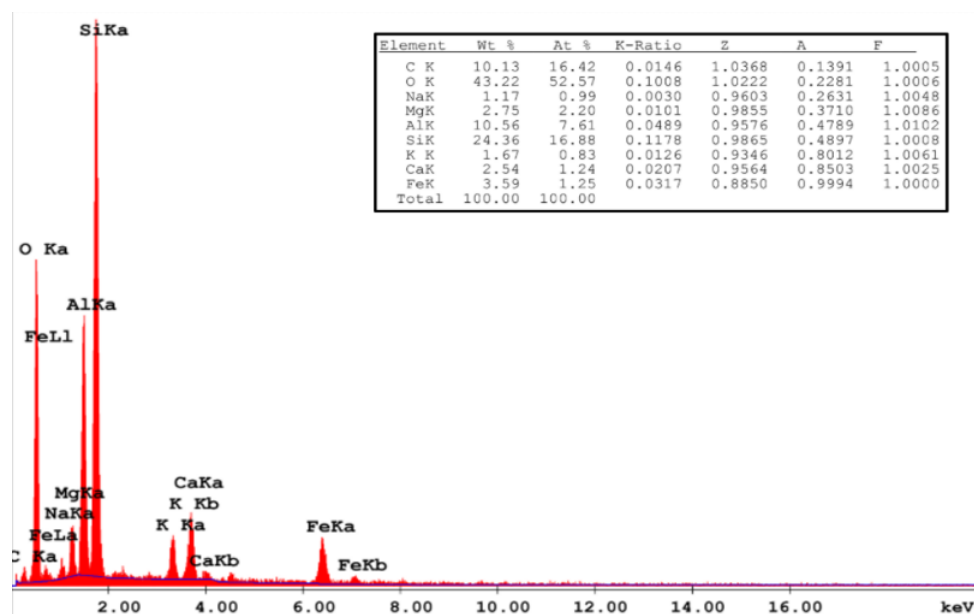


Fig. 10. SEM-EDS characterization of 600 °C thermally activated coal mine tailings (CMT)

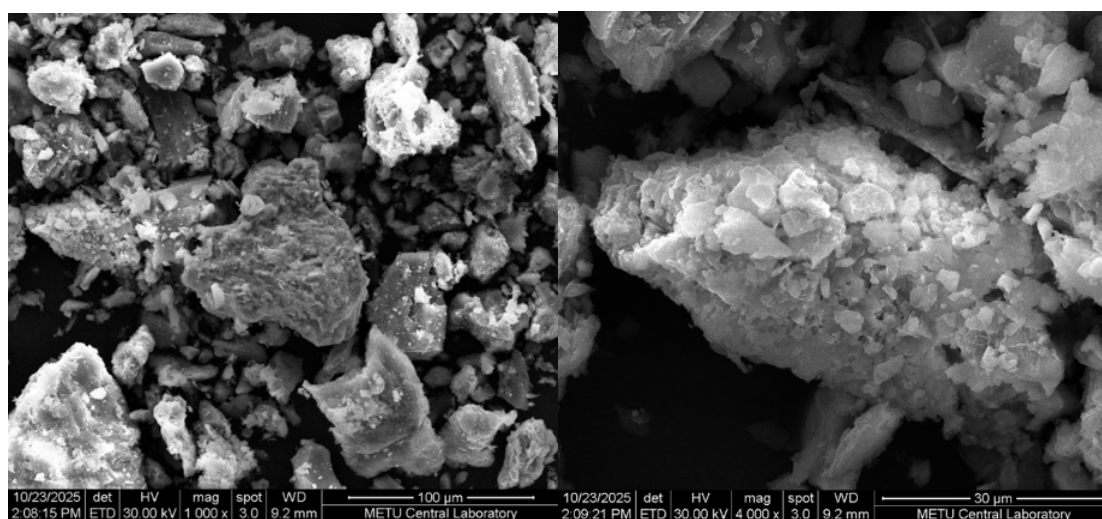


Fig. 11. SEM micrographs of the raw CMT_800 sample at different magnifications: (left) 100 μm and (right) 30 μm

Compared with the raw CMT and the CMT sample activated at 600 °C, the particles in the 800 °C-activated sample exhibited a more rounded geometry. Moreover, the particle surfaces showed brighter, more compact microtopography. This morphological change suggests the onset of partial sintering in the clay phase.

The SEM images indicate that residues associated with the organic phase were largely eliminated.

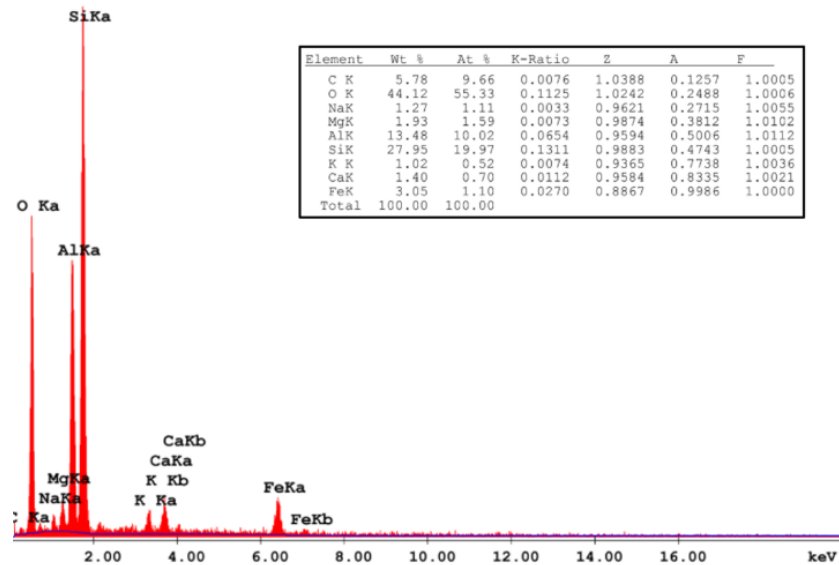


Fig. 12. SEM-EDS characterization of 800 °C thermally activated coal mine tailings (CMT)

This observation is further supported by the EDS results, which show that the C content of the CMT sample thermally activated at 800 °C decreased to approximately 5%, indicating substantial removal of organic matter (Fig. 12). The TGA-DSC results indicate that the mass loss occurring during heating may partly be associated with the thermal decomposition of residual organic matter. In addition, the interparticle voids became denser and more closed compared with those observed in the sample activated at 600 °C. This finding is consistent with the onset of sintering and indicates that the particles coalesced into larger, more compact masses.

Fig. 13 presents the compressive strength results of mortar specimens prepared with untreated coal mine tailings (CMT_0) and thermally activated CMT at 600 °C and 800 °C at 28 and 90 days of curing. Fig. 13 compares the UCS values of mortars incorporating CMT_0 and thermally activated CMT at 600 °C and 800 °C at 28 and 90 days of curing. Overall, thermal activation markedly influenced CMT binding performance, with its effectiveness strongly dependent on both activation temperature and curing duration.

At 28 days, CMT activated at 600 °C exhibited higher UCS values than untreated CMT_0 at low to moderate replacement ratios. Specifically, at 5% replacement, the UCS of the CMT_600T mixture

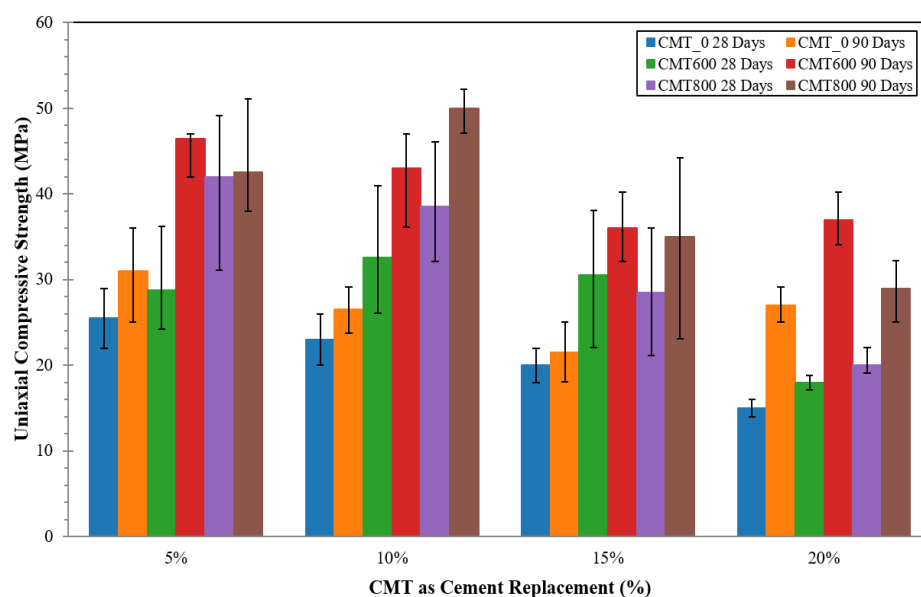


Fig. 13. Effect of thermal activation temperature (600 °C and 800 °C) on the compressive strength of mortars containing CMT as a cement replacement

reached approximately 29 MPa, compared to about 26 MPa for the untreated CMT_0 mixture. An enhancement was observed at 10% replacement, where CMT_600T achieved around 32 MPa, while CMT_0 remained close to 23 MPa. A similar improvement was also observed at the 15% replacement level. At this replacement ratio, the CMT_600T sample reached a compressive strength of approximately 30 MPa, whereas the CMT_0 sample achieved only around 20 MPa. At higher replacement levels, although strength increased in samples thermally activated at 600 °C after 28 days of curing, the improvement remained limited. In contrast, after 28 days of curing, CMT samples thermally activated at 800 °C showed higher UCS values than untreated CMT samples, particularly at lower replacement levels. Specifically, the UCS of CMT_800T reached about 42 MPa at 5% replacement and about 39 MPa at 10% replacement. In contrast, CMT thermally activated at 800 °C showed a decreasing strength trend with increasing replacement level; the UCS value declined to about 20 MPa at 20% replacement.

At 90 days of curing, the beneficial effect of thermal activation became more pronounced, particularly for mixtures incorporating thermally treated coal mine tailings. This pronounced long-term strength gain is directly associated with the thermal transformations identified by TGA-DSC and XRD analyses. The TGA-DSC results indicate significant dehydroxylation of clay-bearing phases at elevated temperatures, breaking down hydroxyl-containing crystalline structures. Consistently, XRD patterns reveal a reduction in crystalline peak intensities and partial amorphization, confirming the formation of highly reactive amorphous aluminosilicate phases. CMT activated at 800 °C achieved the highest UCS at a 10% replacement level, reaching approximately 50 MPa, corresponding to an increase of nearly 100% compared to the untreated CMT mixture at the same replacement ratio. However, in general, CMT thermally activated at 600 °C showed better compressive strength at longer curing ages. This indicates that high-temperature activation may adversely affect late-age reactivity, likely because excessive sintering and partial recrystallization reduce the availability of reactive amorphous phases.

These thermally induced structural changes enhance the pozzolanic reactivity of CMT by increasing the availability of reactive silica and alumina, thereby promoting sustained secondary pozzolanic reactions and prolonged C-(A)-S-H gel formation during extended curing periods. Consequently, the cementitious matrix becomes progressively densified, resulting in superior long-term strength development for CMT activated at 800 °C. In comparison, CMT activated at 600 °C exhibited a more balanced strength evolution, providing consistently higher UCS values than untreated CMT_0 at both curing ages. For example, at a 20% replacement level, the UCS of CMT_600T reached approximately 37 MPa, compared to about 27 MPa for CMT_0, indicating that moderate thermal activation effectively enhances reactivity while limiting excessive phase transformation.

Untreated CMT_0 specimens exhibited lower compressive strength values than thermally activated counterparts at both curing ages, confirming that thermal activation significantly improves the suitability of CMT as a cement replacement material. Nevertheless, all mixtures showed a general reduction in UCS at the 20% replacement level, highlighting the dominance of the dilution effect at high substitution ratios and indicating that the optimal replacement level should be limited.

These findings are consistent with previous studies reporting that thermal activation in the range of 600-800 °C enhances the pozzolanic reactivity of clay-rich coal wastes through dehydroxylation and partial amorphization, while excessively high temperatures may induce recrystallization and compromise early-age performance (Zhang and Ling, 2020; Zou et al., 2024). Overall, Fig. 13 shows that thermal activation substantially improves the mechanical performance of CMT; however, its effectiveness depends strongly on both the activation temperature and the replacement level.

From a sustainability perspective, it is important to consider the energy demand associated with high-temperature activation, particularly at 800 °C. Activation at 600 °C offers a more favorable performance-energy trade-off, delivering substantial improvements in both early- and late-age strength with lower energy consumption. Accordingly, thermal activation at 600 °C combined with moderate replacement levels (5-10%) emerges as a more practical and sustainable option for large-scale applications, while 800 °C activation should be selectively employed when enhanced long-term strength is a primary design objective.

4. Conclusions

This study evaluated the feasibility of utilizing coal mine tailings (CMT) as a supplementary cementitious material through selective activation strategies, including mechanical grinding, pozzolan-

assisted blending, and thermal activation. The findings show that CMT effectiveness is strongly governed by the activation method, curing age, and replacement level, highlighting the importance of a tailored activation–replacement strategy to optimize performance.

Mechanical activation through grinding successfully refined particle size but did not translate into improved mechanical performance. Mortars containing untreated CMT consistently outperformed those incorporating mechanically activated CMT, indicating that excessive fineness led to particle agglomeration and increased water demand, which limited effective hydration and strength development. Accordingly, mechanical activation under the applied conditions cannot be considered a viable route for enhancing CMT performance.

In contrast, pozzolan-assisted blending proved to be an effective strategy, particularly at later curing ages. Binary and ternary mixtures incorporating trass and fly ash exhibited clear synergistic effects, with trass playing a dominant role in long-term strength development due to its higher pozzolanic reactivity. While the binary mixture containing trass achieved the highest UCS value of about 41 MPa, corresponding to an increase of nearly 32%, the ternary blend containing trass and fly ash achieved UCS value of about 39–40 MPa, corresponding to an increase of nearly 25% compared to the CMT₀ only mixture. These results confirm that combining CMT with suitable natural or industrial pozzolans can significantly enhance its contribution as a cement replacement material.

Among the investigated approaches, thermal activation produced the most pronounced performance improvement. Activation at 600 °C provided a balanced enhancement of both early- and late-age strength at low to moderate replacement levels. At 28 days, mortars containing 600 °C thermally activated CMT at 10% and 15% replacement levels showed strength increases of 35% and 50%, respectively, compared with untreated CMT mixtures. At 90 days, strength increased by 50% and 65% at 5% and 10% replacement levels, respectively. However, strength reductions observed at higher replacement levels across all mixtures emphasize the dominance of dilution effects and indicate that excessive substitution should be avoided. From a practical and sustainability perspective, the results suggest that thermal activation at 600 °C combined with moderate replacement ratios (5–10%) offers a favorable balance between mechanical performance, energy demand, and industrial feasibility, making it a promising option for large-scale implementation.

Overall, this study demonstrates that coal washing tailings can be effectively valorized as a partial cement replacement when appropriate activation and blending strategies are employed. Future research should extend beyond strength-based assessment to include durability performance, leaching behavior, and comprehensive energy and CO₂ footprint analyses to support environmentally responsible and industrially viable utilization of CMT in cementitious systems.

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