

Article

Ceramic Waste Powder-Derived Dry Activator for Sustainable One-Part Fly Ash-Based Geopolymer Mortars

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Abstract

Ceramic waste powder (CWP) is a silica-rich fired residue with limited reactivity but considerable potential as a precursor for solid alkaline activators. This study investigated the thermochemical conversion of CWP into a dry activator for a one-part fly ash-based geopolymer mortar. A Taguchi L9 design was used to examine the effects of SiO₂/Na₂O molar ratio (0.5–2.5), activation temperature (150–350 °C), and activation duration (1–3 h). The synthesised powders were characterized by FTIR, while their activation performance was evaluated through 28-day compressive strength testing, supported by the SEM-EDS analysis of selected mortars. Compressive strength ranged from 4.63 to 14.16 MPa, with the highest value obtained at a molar ratio of 0.5, 350 °C, and 3 h. The replicate-based S/N analysis showed the largest main-effect variations for activation temperature and molar ratio, whereas activation duration had a smaller influence. FTIR and microstructural observations indicated more pronounced changes in the principal Si-O-T spectral region and a denser geopolymer matrix in the better-performing systems. The results show that thermochemical processing can convert CWP into a functional dry activator, demonstrating the proof-of-concept feasibility of a powder-based route for one-part geopolymer mortar production.

Keywords: one-part geopolymer composites; dry alkaline activators; thermochemical synthesis; ceramic waste powder; compressive strength; sustainable construction



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1. Introduction

The construction industry is undergoing a significant shift toward low-carbon, resource-efficient binder systems as a response to the growing environmental burden associated with ordinary Portland cement (OPC). The production of OPC is responsible for approximately 8% of global CO₂ emissions, largely due to the calcination of limestone and the high thermal energy required during clinker formation [1]. In contrast, alkali-activated materials (AAMs) and geopolymers offer an attractive alternative due to their ability to utilise industrial by-products such as FA, slag, and silica-rich wastes while providing good mechanical and durability performance [2]. Recent studies have demonstrated that FA-based geopolymers can reduce embodied CO₂ by 40–80% compared to OPC, depending on the activator chemistry and curing regime [3,4]. Comparable reductions have also been

reported for slag- and other industrial by-product-based AAM systems, indicating that the bulk of the carbon saving derives primarily from the avoidance of clinker production rather than from the specific aluminosilicate precursor used. Despite these environmental advantages, the widespread adoption of geopolymer technology remains constrained by challenges associated with conventional activator systems.

Traditional two-part geopolymer systems rely on highly alkaline liquid activators, typically sodium hydroxide and sodium silicate solutions, which introduce several barriers to broader construction use. These liquids are corrosive, difficult to handle, sensitive to mixing sequence, and subject to storage and transport constraints. In addition, the industrial production of sodium silicate is itself energy-intensive, as it generally requires high-temperature (1200–1400 °C) processing of silica-rich raw materials [5]. These limitations have driven growing interest in one-part, or “just-add-water”, geopolymer systems, where solid activators are pre-mixed with the precursor and only water is added during mixing [6]. Such systems offer improved safety, simpler batching, and greater similarity to conventional cement-based construction practice. More broadly, geopolymer materials are being investigated for infrastructure applications including bridge components, precast elements, pavements and repair systems, although their field implementation remains considerably less mature than that of Portland-cement-based materials. Recent reviews of bridge and infrastructure applications emphasise that practical adoption depends not only on mechanical performance but also on durability, constructability, standardisation and reliable activator technology [7]. In this context, one-part systems are particularly attractive because the use of solid activators can simplify batching and potentially facilitate wider field implementation, although challenges related to reaction control, shrinkage, efflorescence and mix standardisation remain. Consequently, the development of sustainable and effective solid alkaline activators has become a key research priority for advancing practical one-part geopolymer technology.

A major challenge in one-part geopolymer development is the availability of low-cost and low-impact solid activators. To address this issue, a number of silica-rich waste materials have been investigated as alternative feedstocks for DA production [8], including rice husk ash [9,10], rice straw ash [11], silica fume [12], waste glass powder [13,14], red mud [15], and sewage sludge ash [16]. Previous studies have demonstrated that such wastes can be thermochemically or chemically transformed into sodium-silicate-rich activating powders capable of initiating geopolymerisation. For example, Tchakouté et al. [17,18] reported that rice husk ash can provide dissolved silica suitable for geopolymer activation, while Tong et al. [19] showed that silica dissolution can be strongly influenced by alkali concentration and temperature. Waste glass has also shown considerable promise due to its high amorphous silica content and reactivity in alkaline environments [20,21]. Previous waste-derived one-part activator studies have predominantly focused on silica-rich feedstocks with comparatively high amorphous or reactive fractions. These studies have demonstrated that waste-derived silica can partially or fully replace commercial sodium silicate, although the resulting activation efficiency depends strongly on feedstock chemistry, silica availability and the synthesis route. In contrast, comparatively little attention has been given to previously fired residues as feedstocks for dry activator synthesis.

CWP is one such residue. It is generated in large quantities from tile manufacturing, sanitaryware production, and demolition-related ceramic waste, and a substantial portion is landfilled despite its significant silica and alumina content. Ceramic manufacturing, installation and demolition generate substantial quantities of fired ceramic residues, much of which remain underutilised or landfilled despite its silica- and alumina-rich composition [22]. It is worth noting that within the alkali-activation literature, CWP has so far been explored almost exclusively in the opposite role to the one investigated here: as the

aluminosilicate precursor itself, activated using conventional liquid NaOH/sodium silicate solutions [23,24]. A recent review of ceramic-waste alkali activation reported compressive strengths of approximately 40–58 MPa for selected ceramic-waste mortars and substantially higher values when ceramic waste was combined with calcium-rich or conventional geopolymer precursors [25]. Similarly, CWP-slag geopolymer concrete optimised using conventional liquid NaOH/sodium-silicate activation reached approximately 58.9 MPa, while studies on sanitaryware-derived ceramic powders have demonstrated strong sensitivity to NaOH molarity [26]. These findings establish CWP as a potentially useful aluminosilicate precursor but also demonstrate its dependence on externally supplied alkaline and soluble silicate activators. Using CWP not as the geopolymer precursor but as the silica-bearing feedstock for synthesising, the solid activator therefore represents a fundamentally different application of this waste stream and, to the authors' knowledge, has not previously been investigated. In contrast to waste glass, however, ceramic waste originates from previously fired products and is therefore expected to exhibit lower intrinsic reactivity due to the formation of more stable mineral phases. This low reactivity makes CWP a challenging but attractive precursor for developing functional powder-based activators. If such a fired ceramic residue can be structurally modified through thermochemical treatment, it may provide a new route for producing waste-derived DA and the scope of one-part geopolymer technology could be extended beyond highly reactive glassy waste streams.

At present, the thermochemical reactivity of CWP-NaOH mixtures, the influence of synthesis parameters on the resulting activator quality, and the degree to which such activators can support geopolymerisation in FA-based mortars remain insufficiently understood. This question is also distinct from, and not a straightforward extension of, the authors' previous work on a waste-glass-derived DA [14]. Waste glass is typically dominated by an amorphous silica-rich network and is comparatively susceptible to alkaline attack; CWP, by contrast, originates from previously fired ceramic products and is therefore expected to contain thermally stabilised silicate/aluminosilicate constituents that may respond differently to alkaline thermochemical treatment than highly amorphous waste glass. The specific mineralogical assemblage of the CWP used here was not determined by XRD and is therefore not assumed in the present interpretation. Whether such a structurally stable, previously fired silica source can be converted into a functional DA at all, and if so, under what synthesis conditions is therefore an open question with implications beyond CWP itself: a positive result would indicate that the waste-derived DA approach generalizes beyond glassy precursors to the broader and largely untapped category of fired ceramic and construction-demolition wastes. In particular, the individual and combined effects of $\text{SiO}_2/\text{Na}_2\text{O}$ molar ratio (defined here as the molar ratio between the silica supplied by CWP and the Na_2O supplied by NaOH during DA synthesis), activation temperature, and activation duration on the engineering performance of CWP-derived one-part geopolymer systems have not been systematically established. Moreover, existing studies on waste-derived DAs rarely couple process optimisation with microstructural interpretation and comparative sustainability assessment within the same framework. Addressing these gaps is essential for evaluating whether CWP-derived DAs are merely chemically feasible or also practically relevant for sustainable binder development.

Therefore, this study investigates the feasibility, optimisation, and performance of CWP-based DAs for FA-based one-part geopolymer mortars. A Taguchi design of experiments was employed to evaluate the effects of molar ratio, activation temperature, and activation duration on the performance of the synthesised DA. The resulting powders were assessed primarily through FTIR-based structural interpretation and 28-day compressive strength response, while selected mixes were further examined using SEM-EDS to provide supporting microstructural insight. In addition, a conventional two-part geopolymer

was used as a benchmark, and a simplified material-level sustainability and cost assessment was conducted to compare the environmental and economic implications of the proposed one-part system. By focusing on a fired ceramic residue rather than a highly reactive amorphous silica waste, the study aims to broaden the range of feasible feedstocks for DA production and provide a more demanding test case for waste-derived one-part geopolymer technology.

2. Materials and Methods

2.1. Raw Materials

Class F FA conforming to ASTM C618 [27], with a combined $\text{SiO}_2 + \text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}_3$ content exceeding 50 wt.%, was used as the principal aluminosilicate precursor. CWP, collected from Keane Ceramics, West Gosford, NSW, Australia, was used as the silica-bearing source for DA synthesis. The CWP was first crushed using a jaw crusher and then milled in a planetary ball mill for 1 h to obtain a powder finer than $75\text{ }\mu\text{m}$. River sand with a fineness modulus of 2.71 was used as fine aggregate. Potable tap water was used for specimen preparation.

The chemical compositions of FA and CWP were determined by X-ray fluorescence (XRF) and are presented in Table 1. FA mainly consisted of SiO_2 , Al_2O_3 , and Fe_2O_3 , while CWP was rich in SiO_2 with moderate Al_2O_3 content. Particle size distribution was measured using a Malvern Mastersizer 3000 (Worcestershire, United Kingdom), and representative morphologies of FA and CWP were examined using scanning electron microscopy (SEM) shown in Figures 1 and 2. FA exhibited predominantly spherical particles, whereas CWP showed angular and irregular particles produced by crushing of fired ceramic products.

Table 1. Chemical compositions of FA and CWP (%).

Materials	SiO_2	Al_2O_3	Fe_2O_3	CaO	SO_3	MgO	Na_2O	K_2O	LOI
FA	51.48	28.39	6.93	5.42	0.09	1.10	0.15	0.53	0.03
CWP	71.0	13.4	1.23	2.8	0.35	2.5	2.1	0.2	0.07

Note: All values in mass%, expressed on an oven-dry basis; LOI: loss on ignition at $1000\text{ }^\circ\text{C}$.

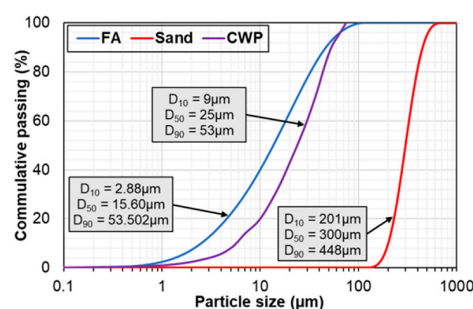


Figure 1. Particle size distributions of FA, CWP, and sand.

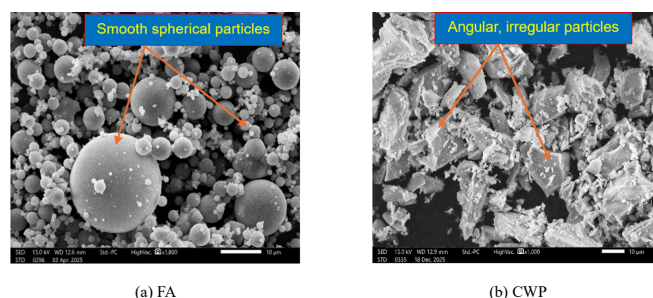


Figure 2. SEM micrographs of raw materials used in this study.

For benchmarking purposes, a conventional two-part geopolymer was also prepared using sodium hydroxide pellets (98% purity, Chem-Supply Pty Ltd., Gillman, Australia) and commercial sodium silicate solution (14.7% Na₂O, 29.4% SiO₂, and 55.9% water by mass, Sigma-Aldrich Australia, Bayswater, Australia).

2.2. Experimental Design

The experimental program was structured into five sequential phases to systematically address the research questions of this study. Phase I involved raw material characterisation to establish the physical and chemical suitability of CWP, FA, and sand for geopolymer production. Phase II focused on the synthesis of CWP-based DAs through thermochemical treatment under varying SiO₂/Na₂O molar ratios, activation temperatures, and activation durations, thereby addressing the question of whether CWP can act as a viable precursor for DA production. Phase III comprised the structural characterisation and statistical optimisation of the synthesised activators using FTIR, Taguchi signal-to-noise ratio analysis, and ANOVA to identify the dominant synthesis parameters. Phase IV evaluated the mechanical performance of one-part geopolymer mortars prepared with the synthesised activators, thereby assessing the engineering feasibility of the developed system. Phase V involved microstructural analysis and a comparative sustainability and cost assessment to interpret the observed strength response and evaluate the broader practical implications of the proposed DA approach.

The DA synthesis was designed using a Taguchi L9 orthogonal array with three factors at three levels [28] to evaluate the influence of three parameters: molar ratio (A), activation temperature (B), and activation duration (C). The selected levels are given in Table 2. The Taguchi approach was adopted to minimise the number of experiments while still enabling systematic assessment of the main synthesis variables. Since only three factors were considered, the fourth column of the standard L9 array remained unassigned and was used only to represent the residual/unassigned contribution in the descriptive main-effect analysis.

Table 2. Selected design parameters and their levels.

Parameters		Levels		
		1	2	3
A	Molar ratio	0.5	1.5	2.5
B	Activation temperature (°C)	150	250	350
C	Activation duration (h)	1	2	3

The selected molar ratio range (0.5–2.5), activation temperature (150–350 °C), and activation duration (1–3 h) were chosen based on previous studies on waste-derived dry activators and preliminary feasibility considerations [13,14,16,29–32]. These ranges were intended to cover alkali-rich to silica-rich compositions and low to moderately high thermal treatment conditions. Table 3 shows the L9 orthogonal array used in the study.

Table 3. L9 orthogonal array used for experimental design.

Experiment No.	E1	E2	E3	E4	E5	E6	E7	E8	E9
Molar ratio	0.5	0.5	0.5	1.5	1.5	1.5	2.5	2.5	2.5
Temperature (°C)	150	250	350	150	250	350	150	250	350
Time (h)	1	2	3	2	3	1	3	1	2

2.3. Synthesis of CWP-Based Dry Activator

The DAs were synthesised by thermochemical treatment of CWP with NaOH [33], following approaches adapted from previous studies on waste-derived DAs [13,14,29]. CWP

was used as the silica-bearing feedstock, while NaOH supplied the alkaline component required for thermochemical modification of the CWP. FA was excluded from the DA synthesis stage because it was subsequently used as the principal aluminosilicate precursor in the one-part mortar. For each experimental run, 150 g of solid mixture was prepared according to the target molar ratio, calculated from the measured SiO_2 content of CWP (71 wt.%) and the stoichiometric Na_2O equivalent of NaOH. The powders were dry mixed thoroughly and transferred into covered ceramic crucibles. The theoretically weighed masses for 150 g of CWP-derived dry activator are given in Table 4 considering 98% NaOH purity.

Table 4. Theoretical weighed masses for 150 g of CWP-derived dry activator.

Molar Ratio	CWP (g)	NaOH (g)
0.5	51.21	98.79
1.5	91.29	58.71
2.5	108.24	41.76

Heat treatment was carried out in a programmable muffle furnace at the selected temperature (150, 250, or 350 °C) for the selected duration (1, 2, or 3 h), using a heating rate of 10 °C/min. After cooling to room temperature, the reacted material was lightly crushed, ground, and sieved to below 75 μm , then stored in airtight containers prior to use. In this study, the term DA refers to the thermochemically treated CWP-NaOH powder that was subsequently used to activate the one-part geopolymer system. The methodology of development of CWP-based DA through thermochemical synthesis is shown in Figure 3.

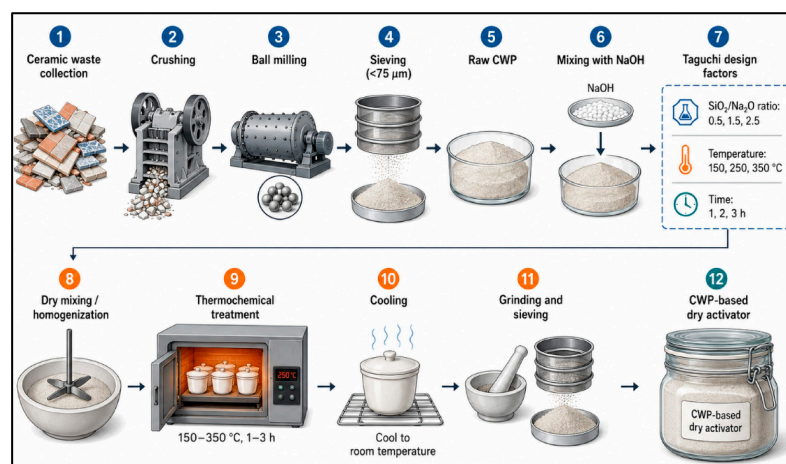


Figure 3. Methodology of development of CWP-based DA via thermochemical synthesis.

2.4. Preparation of One-Part Geopolymer Mortar

Nine one-part geopolymer mortars were produced using the nine synthesised DAs (E1–E9). The FA-to-sand ratio was fixed at 1:1.5 by mass, the total binder-to-sand ratio was 1:1.275, while the FA-to-DA ratio was maintained at 85:15. The water-to-binder ratio was fixed at 0.34. The corresponding Na_2O -equivalent dosage on an FA basis ranged from 3.73% to 8.82%. The selected range intentionally covered relatively alkali-rich, intermediate and lower-alkali conditions, allowing the sensitivity of the CWP-derived activator to alkali availability to be examined [2,34,35]. This proportion provides balanced alkali content and favourable workability at the selected water-to-binder ratio ($w/b = 0.34$). The Na_2O -equivalent content supplied by NaOH was calculated stoichiometrically using the actual NaOH mass and purity. For molar ratios of 0.5, 1.5 and 2.5, the corresponding Na_2O -equivalent fractions in the dry activator were approximately 50.0%, 29.7% and

21.1%, respectively. At a FA:DA ratio of 85:15, these corresponded to Na₂O dosages of approximately 8.82%, 5.24% and 3.73% relative to the FA mass, as summarised in Table 5. The mix proportions adopted for geopolymer mortar are summarised in Table 6.

Table 5. Calculated Na₂O content on FA basis for different molar ratios.

Run	E1	E2	E3	E4	E5	E6	E7	E8	E9
Na ₂ O in DA (%)	50.01	50.01	50.01	29.72	29.72	29.72	21.14	21.14	21.14
Na ₂ O on FA (%)	8.82	8.82	8.82	5.24	5.24	5.24	3.73	3.73	3.73

Table 6. Mix proportions of the one-part and two-part geopolymer mortars (kg/m³).

Mix	Fly Ash	Dry Activator	Sand	Water	NaOH	Na ₂ SiO ₃
One-part	425	75	637.5	170	-	-
Two-part	500	-	750	-	71.43	178.57

The dry materials, including FA and DA, were first mixed for 2 min, followed by addition of sand for 1 min. Water was then gradually introduced and mixing continued for 3 min until a uniform mortar was obtained [36]. Approximately 300 g of mortar was prepared for each mix, sufficient to cast six cylindrical specimens of 25 mm diameter × 25 mm height in accordance with ASTM C579 method A [37].

Immediately after casting, the moulds were covered with plastic film and left at room temperature for 24 h. The specimens were then heat-cured at 60 °C for 24 h and subsequently stored under sealed conditions at 23 ± 2 °C until testing at 28 days. This curing regime was adopted to provide consistent and comparable conditions for screening the effectiveness of the synthesised DAs. The complete methodology is shown in Figure 4.



Figure 4. Methodology of one-part geopolymer mortar preparation.

2.5. Preparation of Conventional Two-Part Geopolymer Mortar

A conventional two-part FA-based geopolymer mortar was prepared as a practical benchmark. The liquid activator consisted of a nominal 10 M NaOH solution and commercial sodium silicate solution at a sodium-silicate-to-NaOH-solution mass ratio of 2.5. The activator-to-binder ratio was fixed at 0.50. At a binder content of 500 kg/m³, the total liquid activator content was therefore 250 kg/m³, comprising 71.43 kg/m³ NaOH solution and 178.57 kg/m³ sodium silicate solution. No additional mixing water was added.

For compositional accounting, the nominal 10 M NaOH solution was evaluated using a density of 1.33 g/cm³ at 20 °C, consistent with supplier data for 10 M NaOH solution.

Accordingly, the 71.43 kg/m³ NaOH solution contained approximately 21.48 kg/m³ NaOH and 49.95 kg/m³ water. The NaOH content corresponds to an approximately 16.64 kg/m³ Na₂O-equivalent. The commercial sodium silicate solution contained 14.7% Na₂O, 29.4% SiO₂ and 55.9% water by mass and therefore contributed 26.25 kg/m³ Na₂O, 52.50 kg/m³ SiO₂ and 99.82 kg/m³ water. Consequently, the complete liquid activator contained approximately 42.89 kg/m³ total Na₂O-equivalent, 52.50 kg/m³ SiO₂ and 149.77 kg/m³ water, corresponding to an activator-water-to-binder ratio of approximately 0.30. The complete mass and compositional balance are summarised in Table 7. The benchmark was used as a practical formulation-level reference rather than as an iso-water, iso-alkali or iso-silicate mechanistic control.

Table 7. Mass and compositional balance of the liquid activator used in the two-part benchmark.

Parameter	NaOH Solution	Sodium Silicate Solution	Total Activator
Solution dosage (kg/m ³)	71.43	178.57	250
NaOH (kg/m ³)	21.48	-	21.48
Na ₂ O-eq (kg/m ³)	16.64	26.25	42.89
SiO ₂ (kg/m ³)	-	52.50	52.50
Water (kg/m ³)	49.95	99.82	149.77
Na ₂ O-eq/binder (%)	3.33	5.25	8.58
SiO ₂ /binder (%)	-	10.50	10.50
Water/binder	-	-	0.30

2.6. Testing Methods

The 28-day compressive strength of the one-part and two-part geopolymers was determined using a 100 kN universal testing machine following ASTM C579 method A [37]. Six specimens were tested for each mix, and the average compressive strength together with standard deviation was reported.

The synthesised DAs were characterised using Fourier transform infrared spectroscopy (FTIR) to examine the structural changes caused by thermochemical treatment. The FTIR interpretation was used as a comparative indicator of structural modification among the activators rather than as definitive phase identification. Selected geopolymer mortars representing the best-performing, intermediate, and lowest-performing systems were further examined by SEM coupled with energy-dispersive X-ray spectroscopy (EDS) to evaluate matrix morphology and local gel chemistry.

2.7. Statistical Analysis

Mean compressive strength was retained as the primary engineering response, while the larger-the-better signal-to-noise ratio (S/N) was used as a complementary Taguchi screening metric. Unlike comparison based only on mean strength, the replicate-based S/N ratio incorporates both response magnitude and within-run variability and therefore provides a more robust basis for ranking the experimental conditions [38]. For each experimental run, the S/N ratio was calculated from the six individual compressive-strength measurements using Equation (1):

$$\eta_i = -10 \log_{10} \left(\frac{1}{n_r} \sum_{r=1}^{n_r} \frac{1}{y_{ir}^2} \right) \quad (1)$$

where η_i is the signal-to-noise ratio for experimental run i , n_r is the number of replicate specimens for each run ($n_r = 6$), and y_{ir} is the compressive strength of replicate r in experimental run i . The mean S/N ratio at each level of each synthesis factor was then calculated using Equation (2):

For factor-level S/N:

$$\eta_{k,j}^- = \frac{1}{N_j} \sum_{i \in (k,j)} \eta_i \quad (2)$$

where $\eta_{k,j}^-$ is the mean S/N ratio at level j of factor k , and $N_j = 3$ is the number of L9 experimental runs occurring at each factor level. The grand mean of the S/N ratios was calculated using Equation (3):

$$\eta^- = \frac{1}{N} \sum_{i=1}^N \eta_i \quad (3)$$

where $N = 9$ is the total number of experimental runs.

A descriptive analysis of variance (ANOVA) was subsequently performed on the S/N response to estimate the relative main-effect contribution of molar ratio, activation temperature and activation duration. The total sum of squares was calculated using Equation (4):

$$SS_T = \sum_{i=1}^N (\eta_i - \bar{\eta})^2 \quad (4)$$

The sum of squares associated with each factor k was determined from the corresponding factor-level S/N means using Equation (5):

$$SS_k = \sum_{j=1}^L N_j (\bar{\eta}_{k,j} - \bar{\eta})^2 \quad (5)$$

where $L = 3$ is the number of levels for each factor. The residual/unassigned sum of squares was calculated as:

$$SS_R = SS_T - \sum_{k=1}^K SS_k \quad (6)$$

where $K = 3$ is the number of investigated synthesis factors. The mean square for each factor was obtained using Equation (7):

$$MS_k = \frac{SS_k}{df_k} \quad (7)$$

where $df_k = L - 1 = 2$ for each three-level factor. The relative contribution of each factor to the total S/N variation was expressed using Equation (8):

$$P_k = \frac{SS_k}{SS_T} \times 100 \quad (8)$$

where P_k is the percentage contribution of factor k .

Because the L9 design provides only two residual degrees of freedom and the residual/unassigned component does not represent an independently replicated synthesis-level estimate of experimental error, F-statistics and formal significance testing were not applied. The ANOVA was therefore interpreted as a descriptive main-effect decomposition rather than inferential statistical evidence. Accordingly, the statistical analysis was used to rank the relative influence of the investigated synthesis parameters within the experimental design space rather than to establish universal parameter significance.

2.8. Sustainability and Economical Analysis

A comparative material-level cost and CO₂ assessment was conducted for the conventional two-part geopolymer and the CWP-based one-part geopolymer. The calculations were normalised to 1 m³ of geopolymer mortar and included the upstream burdens associated with the constituent materials and transportation. CWP was treated as a waste-derived material under a cut-off allocation approach; therefore, no upstream production burden was assigned to the incoming ceramic waste. Electricity consumption and associated costs for CWP crushing, ball milling, sieving, thermochemical furnace activation and specimen cur-

ing were not included. Accordingly, the assessment represents a simplified material-level comparison rather than a complete cradle-to-gate life-cycle assessment, and the reported reductions may be lower when the full processing energy of dry-activator production is considered.

3. Results and Discussion

3.1. Comparative FTIR Characteristics of the Synthesised Dry Activators

Figure 5 presents the FTIR spectra of the nine CWP-derived dry activators synthesised under different $\text{SiO}_2/\text{Na}_2\text{O}$ molar ratios, activation temperatures and activation durations. To facilitate direct comparison, the spectra are presented in the principal fingerprint region and are individually normalised and vertically offset; therefore, differences in absolute transmittance intensity are not used for quantitative comparison. The interpretation instead focuses on experimentally observed band positions, spectral shape and the presence of features within defined wavenumber regions. FTIR is used here as a comparative structural characterisation technique rather than for definitive identification or quantification of individual reaction phases.

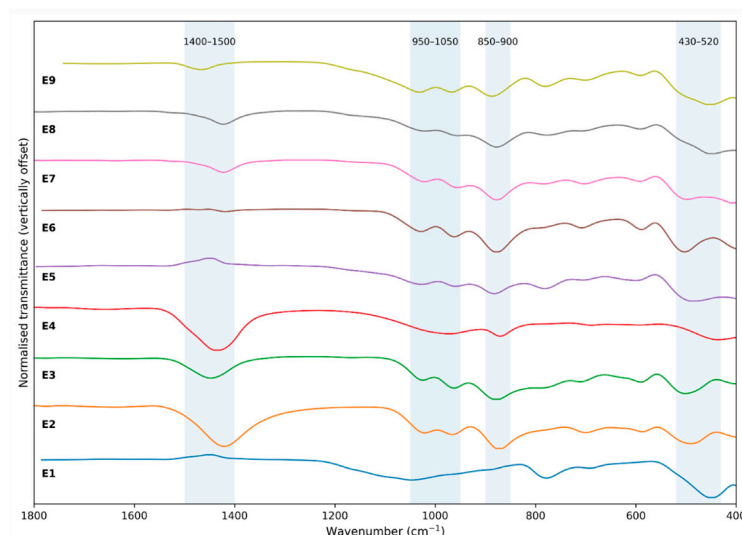


Figure 5. FTIR spectra of the synthesised CWP-based DAs (E1–E9).

A Si–O–T-related spectral feature was observed in every synthesised activator within approximately $950\text{--}1050\text{ cm}^{-1}$, directly confirming that this characteristic silicate region was retained across E1–E9 [39,40]. Importantly, however, its position was not identical among the activators. E1 exhibited a broad principal minimum at approximately 1047 cm^{-1} , whereas E2 and E3 showed features at approximately 1021 and 1025 cm^{-1} , respectively. E2 and E3 additionally exhibited secondary features at approximately 966 and 963 cm^{-1} . The raw spectra confirm these positions directly. The difference between E1 and E2–E3 therefore represents a change in band position and spectral definition rather than the presence versus absence of a Si–O-containing structure.

For the intermediate-molar-ratio activators, E4 displayed its dominant feature at approximately 972 cm^{-1} , while E5 and E6 showed principal features at approximately 1027 and 1028 cm^{-1} together with additional features near 959 and 962 cm^{-1} , respectively. This relatively broad range demonstrates that thermochemical treatment produced measurable differences in the local vibrational environment of the silica-bearing CWP–NaOH powders. However, these shifts should not be interpreted independently as evidence for the formation of a specific sodium-silicate phase or as a quantitative measure of reaction extent.

The higher-molar-ratio activators E7–E9 exhibited closely grouped principal features at approximately 1022, 1020 and 1032 cm^{-1} , respectively, accompanied by secondary bands near 956–968 cm^{-1} . Features were also consistently observed around 878–887 cm^{-1} . Specifically, the corresponding lower-wavenumber features occurred at 877.80 cm^{-1} for E7, 877.74 cm^{-1} for E8 and 887.41 cm^{-1} for E9. The relatively close spectral positions within E7–E9 indicate that changing activation temperature and duration within this molar-ratio group modified the spectra less markedly than the broader differences observed between some of the different compositional groups. This observation is qualitative and is not used as a measure of statistical significance.

A second notable region occurred at approximately 1400–1500 cm^{-1} . A distinct feature in this interval was detected for E2 (1421 cm^{-1}), E3 (1448 cm^{-1}), E4 (1435 cm^{-1}), E6 (approximately 1419 and 1472 cm^{-1}), E7 (1423 cm^{-1}), E8 (1423 cm^{-1}) and E9 (1467 cm^{-1}), whereas a clearly resolved feature in this interval was not identified by the instrument peak search for E1 or E5. This region is commonly associated with carbonate-related vibrations in alkali-containing materials and may reflect carbonation of Na-bearing species during cooling, handling or storage [14]. Because carbonation was not independently quantified, these bands are described as carbonate-related rather than being assigned to a specific carbonate phase. Additional spectral features were observed between approximately 850 and 900 cm^{-1} and below approximately 520 cm^{-1} across most activators. The latter low-wavenumber region was represented by minima between approximately 437 and 503 cm^{-1} among E1–E9 and is consistent with bending vibrations associated with silicate/aluminosilicate environments. Nevertheless, these features are treated as supporting structural observations only.

Overall, examination of the individual raw spectra demonstrates that the principal Si–O–T-related region is present in all nine CWP-derived activators, while its position and definition vary with synthesis condition. The FTIR results therefore support differences in the local silicate environment following thermochemical treatment but do not independently establish sodium–silicate formation, N–A–S–H formation, phase abundance or degree of polymerisation. The FTIR observations are consequently interpreted together with the compressive-strength and SEM-EDS results, while definitive identification of the phases produced during dry-activator synthesis would require complementary techniques such as XRD and/or dissolution-based analysis.

3.2. Compressive Strength of One-Part Geopolymer Mortars

The 28-day compressive strengths of the one-part geopolymer mortars are presented in Figure 6. Table 8 shows the mean compressive-strength response at each factor level. The strength varied from 4.63 MPa to 14.16 MPa (Table 9), confirming that the synthesis condition of the DA had a strong influence on mortar performance. Among all mixes, E3 achieved the highest strength (14.16 MPa), followed by E2 (11.51 MPa), whereas E4 gave the lowest value (4.63 MPa). The intermediate group (E6–E9) showed strengths between 7.05 and 8.12 MPa, while E1 and E5 achieved 5.79 and 5.50 MPa, respectively. The best-performing mix was therefore 206% stronger than the weakest mix, showing that the activator synthesis parameters had a substantial practical impact on geopolymerisation efficiency. This overall response is consistent with the purpose of Taguchi optimisation, where the objective is to identify synthesis conditions that maximise compressive strength while keeping variability under control. A recent paper published follows the same structure by first reporting the raw compressive-strength range and then using S/N analysis to identify the dominant synthesis variables [32]. It should be noted that the adopted mortar was relatively paste-rich, with a sand-to-total-binder ratio of 1.275. The moderate strength obtained despite this relatively low aggregate fraction indicates that performance

was controlled primarily by the reactivity of the CWP-derived activator rather than by aggregate dilution. Accordingly, the present formulation should be regarded as a controlled proof-of-concept system for evaluating DA synthesis parameters rather than an optimised engineering mortar mix.

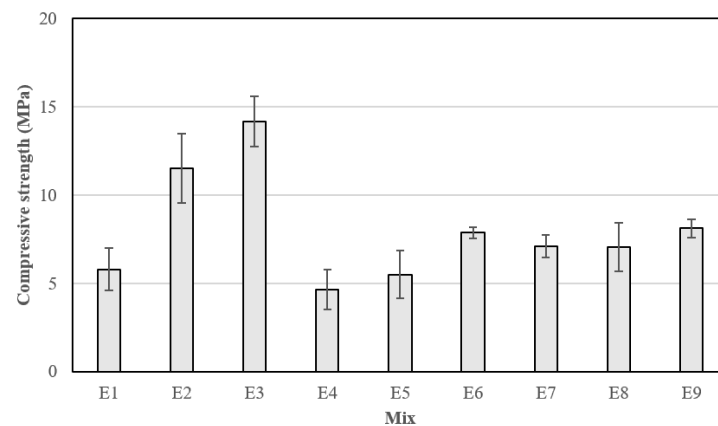


Figure 6. Compressive strength of tested geopolymer mixes.

Table 8. Mean compressive-strength response at each factor level.

Factor	Level 1	Level 2	Level 3	Range (MPa)
Molar ratio	10.49	6	7.42	4.49
Temperature	5.84	8.02	10.05	4.21
Duration	6.9	8.09	8.92	2.02

Note: Each level mean was calculated as the arithmetic mean of the mean compressive strengths of the three L9 experimental runs containing that factor level.

Table 9. Compressive strength, standard deviation and signal-to-noise ratios of the experiments.

Mix	E1	E2	E3	E4	E5	E6	E7	E8	E9
f'_c	5.79	11.51	14.16	4.63	5.50	7.86	7.10	7.05	8.12
SD	1.19	1.98	1.41	1.12	1.34	0.31	0.62	1.36	0.51
S/N	14.77	20.88	22.91	12.72	13.99	17.89	16.94	16.47	18.14

Note: Compressive strength (f'_c) is the average of 6 samples for each mix in MPa. SD is standard deviation in MPa; S/N is the signal-to-noise ratio in dB.

3.2.1. Influence of Dry Activator Synthesis Parameters

The $\text{SiO}_2/\text{Na}_2\text{O}$ molar ratio strongly influenced the compressive strength of the one-part geopolymer mortars. The mortars prepared using activators synthesised at the lowest molar ratio (0.5) produced the highest overall strength, with E2 and E3 reaching 11.51 MPa and 14.16 MPa, respectively. By contrast, the intermediate molar ratio (1.5) resulted in the lowest average group performance, while the highest molar ratio (2.5) produced moderate but more stable strengths. The mean strengths at molar ratios 0.5, 1.5, and 2.5 were 10.49, 6.00, and 7.42 MPa, respectively. This trend indicates that alkali-rich activators were more effective in promoting the reaction of the FA-based one-part system. A lower ratio corresponds to a higher relative Na_2O content, which likely improved the release of soluble alkali species after water addition and enhanced the dissolution of reactive aluminosilicate phases from FA. Greater dissolution would then support the formation of a more continuous sodium aluminosilicate gel. Previous studies have reported that sufficient alkali availability is essential for activating aluminosilicate precursors and accelerating the geo-polymerisation reaction [41,42]. In contrast, at higher molar ratios, the lower

relative alkali availability may have limited the reaction of FA and reduced the extent of geopolymer gel development. Similar trends have been reported in previous studies, where excessive silica relative to alkali resulted in reduced reaction kinetics and lower mechanical strength [43]. Comparable behaviour has been reported in one-part geopolymer systems, where the solid activator dosage and silicate modulus were identified as key variables controlling dissolution and final strength. Although the highest individual strength in this study occurred at the lowest molar ratio, the high-ratio mixes (E7–E9) still developed moderate strengths, indicating that structural modification of the DA was not completely suppressed. This suggests that the CWP-derived activator remained functional over the whole design space but was most effective when sufficient alkali was available during synthesis and subsequent activation.

Activation temperature had a major influence on strength development. The highest-performing mix, E3, was synthesised at 350 °C, and the other mixes prepared at this temperature (E6 and E9) also showed moderate-to-high strengths of 7.86 and 8.12 MPa. By comparison, several of the mortars prepared using activators synthesised at 150 °C showed lower strengths, including E4 (4.63 MPa) and E1 (5.79 MPa). At lower temperatures, the CWP-NaOH interaction may have been less extensive, resulting in a less effective activator after water addition. The mean strengths at 150, 250, and 350 °C were 5.84, 8.02, and 10.05 MPa, respectively. The corresponding SNR trend indicates that increasing activation temperature consistently improved the average performance of the DA. Higher thermal input likely enhanced the interaction between NaOH and the silica-bearing phases in CWP, leading to more pronounced changes in the silicate environment and a powder that produced a stronger mortar response. This interpretation is also consistent with the FTIR results, where the better-performing mixes exhibited more distinct development in the main silicate band region. Similar temperature-driven improvements have been reported in recent studies on thermally or alkali-fused one-part activators, where stronger structural modification of the silica source was associated with improved mechanical performance [14,44,45]. However, the temperature effect should not be interpreted independently of composition. The results indicate that high temperature alone was not sufficient to guarantee high strength; rather, it became most effective when combined with a favourable alkali–silica balance. Therefore, temperature acted as a strong secondary driver that amplified the effect of the selected molar ratio.

Compared with molar ratio and activation temperature, activation duration had the weakest influence on the compressive strength of the mortars. Nevertheless, a gradual increase in mean performance was still observed across the selected time range. The average strengths for 1, 2, and 3 h were 6.90, 8.09, and 8.92 MPa, respectively. This suggests that extending the thermochemical treatment promoted some additional structural modification of the CWP-based activator, but the improvement remained modest compared with the stronger effects of composition and temperature. The best-performing mix (E3) was produced at 3 h, but the overall trend does not suggest that reaction time was the dominant control parameter. Rather, the relatively small change in performance implies that the main thermochemical interaction between CWP and NaOH likely occurred within the lower end of the investigated time window, and that longer treatment mainly refined rather than fundamentally changed the activator structure. This behaviour is consistent with the logic of several recent one-part activator studies, where time was found to have a secondary effect once sufficient alkali and temperature were available [14,46]. From a practical perspective, this is favourable because it suggests that prolonged heat treatment may not be necessary to produce a functional CWP-derived DA. In other words, the system appears more sensitive to what is synthesised than to how long it is held once the essential reaction has been initiated.

3.2.2. Taguchi Optimisation and Descriptive ANOVA

To quantify the influence of the synthesis variables, the compressive strength results were further analysed using the larger-the-better signal-to-noise ratio. The S/N value ranged from 12.72 dB to 22.91 dB, with the highest value recorded for E3, indicating the strongest replicate-based S/N response among the conditions investigated. The mean S/N plots are shown in Figure 7, and the corresponding level means are summarised in Table 10.

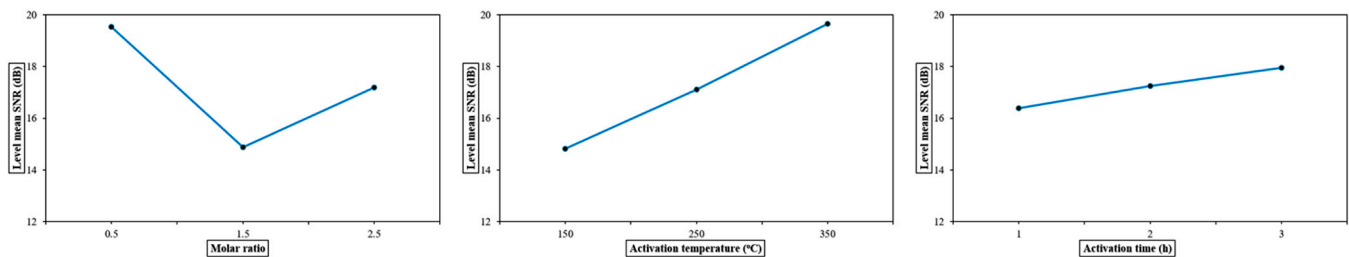


Figure 7. Main-effects plot of the larger-the-better S/N values.

Table 10. Level mean S/N of each design parameter at different levels.

Levels		1	2	3	Max.	Min.	Δ SNR	Rank
Mean SNRs	S/N _{Ms}	19.52	14.87	17.18	19.52	14.87	4.65	2
	S/N _T	14.81	17.11	19.65	19.65	14.81	4.84	1
	S/N _t	16.38	17.25	17.95	17.95	16.38	1.57	3

The S/N trends indicate that the optimum combination for compressive strength was A1B3C3, corresponding to molar ratio = 0.5, activation temperature = 350 °C, and activation duration = 3 h. This optimum is consistent with the actual best-performing experimental run (E3). The near-equal Δ SNR values for molar ratio and temperature indicate that both parameters exerted strong and comparable control over the final response, whereas duration had a much smaller influence. This style of optimisation, where the main effects plot visually ranks the variables and the optimum level is identified directly from the highest S/N, closely follows the structure used in recent studies [14,32].

The ANOVA results are presented in Table 11. Activation temperature accounted for the largest proportion of the variation in S/N response (41.61%), followed closely by the molar ratio (38.49%). Activation duration contributed only 4.40%, while the remaining 15.50% was associated with the residual component. These results are consistent with the factor-level S/N analysis, indicating that temperature and molar ratio produced the strongest main-effect variations within the investigated design space.

Table 11. Percentage contribution of each design parameter.

Parameter	DOF	SS	MS	Contribution (%)
Molar ratio	2	32.48	16.24	38.49
Temperature	2	35.12	17.56	41.61
Duration	2	3.71	1.86	4.40
Residual	2	13.08	6.54	15.50
Total	8	84.39	-	100

Overall, the replicate-based S/N analysis and descriptive ANOVA consistently indicate that activation temperature and molar ratio produced the largest main-effect variations within the investigated design space, whereas activation duration had a comparatively

smaller influence. Given the resolution of the L9 design, these results are restricted to main-effect screening and are not interpreted as evidence of factor interactions.

3.3. Microstructural and Elemental Characteristics of the Geopolymer Mortar

3.3.1. SEM Observations

Representative SEM micrographs (Figure 8) of the mortars produced using E3, E6, and E4 showed clear differences in matrix quality. The best-performing mix, E3, exhibited a relatively dense and continuous matrix, with most FA particles well embedded in the geopolymer gel and only limited micro-porosity. E6 showed a more heterogeneous structure, with visible pores and partially reacted FA particles, indicating an intermediate degree of reaction. The poorest microstructural quality was observed in E4, where the matrix appeared loose and porous, with several unreacted or weakly reacted particles and less effective particle-matrix bonding.

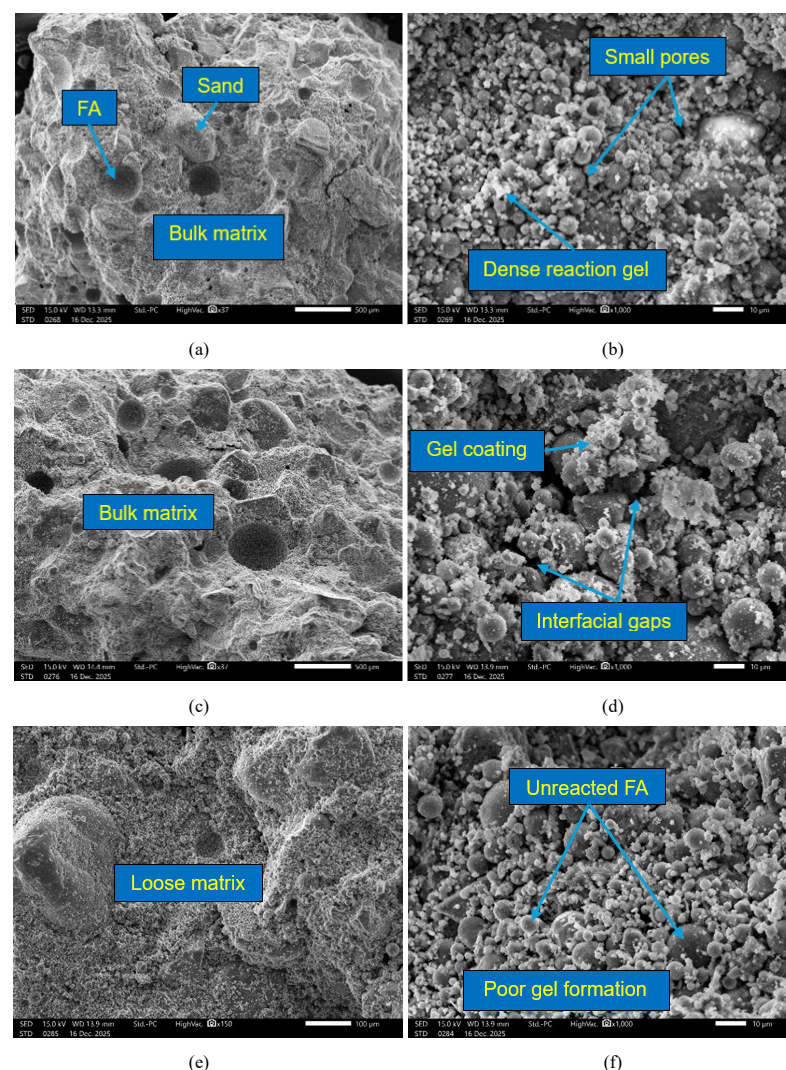


Figure 8. SEM micrographs of geopolymer mortar prepared with CWP-based dry activators: (a,b) E3 (optimal), (c,d) E6 (intermediate), and (e,f) E4 (lowest-performing).

These observations are consistent with the performance trends of the mortars and indicate that the chemistry of the CWP-derived DA strongly influenced matrix continuity and pore development. Similar dense-versus-porous contrasts are commonly reported in one-part geopolymer studies, where better-performing activators promote more extensive

dissolution and gel formation, while weaker systems retain more unreacted particles and interfacial gaps [47,48].

3.3.2. Energy Dispersive X-Ray Spectroscopy (EDS)

EDS was employed to evaluate the elemental composition of geopolymer gel regions in mixes E3, E6, and E4. Multiple point analyses were conducted on representative gel areas, and the resulting atomic percentages were used to determine Si/Al and Na/Al ratios (shown in Table 12), which are key indicators of geopolymer gel chemistry. The optimised mix E3 exhibited an average Si/Al ratio of 1.80 ± 0.16 and Na/Al ratio of 1.01 ± 0.09 . These local EDS ratios indicate comparatively greater Na incorporation in the analysed binder regions than in E6 and E4 [49]. Together with the denser morphology observed by SEM, this is consistent with more extensive alkali activation [50]; however, EDS alone cannot establish the identity, quantity or degree of polymerisation of the reaction products. In contrast, E6 shows a similar Si/Al ratio (1.84) but a markedly lower Na/Al ratio (0.67), indicating reduced alkali incorporation within the gel structure. The lower local Na/Al ratio is consistent with reduced Na incorporation in the analysed regions and coincides with the more heterogeneous morphology observed by SEM.

Table 12. Average geopolymer gel chemistry obtained from EDS point analysis.

Mix	No. of EDS Points	Si/Al (Mean $\pm$ SD)	Na/Al (Mean $\pm$ SD)
E3	6	1.80 ± 0.16	1.01 ± 0.09
E6	6	1.84 ± 0.33	0.67 ± 0.08
E4	6	2.36 ± 0.68	0.71 ± 0.42

The comparatively high and variable Si/Al ratio, together with the lower Na/Al ratio, indicates greater local compositional heterogeneity in the analysed regions, consistent with the weaker mortar response [50].

Overall, the EDS results suggest an association between local Na incorporation and the observed mortar performance; however, these point measurements should be regarded as semi-quantitative supporting evidence rather than definitive identification of geopolymer gel chemistry.

3.4. Comparison Between Conventional Two-Part and CWP-Based One-Part Geopolymers

The activators in the two systems differ fundamentally in physical form and composition, comparison based solely on total activator mass is not appropriate. The 75 kg/m^3 activator used in the one-part system is a dry CWP-NaOH powder, whereas the 250 kg/m^3 activator used in the two-part benchmark is a liquid solution containing a substantial water fraction. The two systems were therefore compared on a common chemical basis using total binder content, actual NaOH solids, Na_2O -equivalent content, activator-derived SiO_2 , $\text{SiO}_2/\text{Na}_2\text{O}$ molar ratio and water content, as summarised in Table 13.

As shown in Table 13, the apparent difference in total activator dosage does not directly represent the difference in alkali content. The two-part system contained 250 kg/m^3 of liquid activator, but approximately 149.77 kg/m^3 of this mass was water. When expressed on a chemically comparable basis, the total Na_2O -equivalent contents were 37.51 and 42.89 kg/m^3 for the one-part and two-part systems, respectively, corresponding to 7.50% and 8.58% of total binder. However, the systems differed substantially in activator-derived SiO_2 content, $\text{SiO}_2/\text{Na}_2\text{O}$ ratio and water content. Accordingly, the comparison is used only to provide formulation-level performance context and is not interpreted as chemically equivalent or mechanistic control.

Table 13. Chemical and compositional basis for comparison of the CWP-based one-part and conventional two-part geopolymers mortars.

Parameter	CWP-Based One Part (E3)	Conventional Two-Part
Total binder (kg/m ³)	500	500
FA (kg/m ³)	425	500
Dry activator (kg/m ³)	75	-
Total liquid activator (kg/m ³)	-	250
NaOH solids (kg/m ³)	49.4	21.48
Na ₂ O-eq (kg/m ³)	37.51	42.89
Na ₂ O eq/binder (%)	7.50	8.58
Activator-derived SiO ₂ (kg/m ³)	18.18	52.50
SiO ₂ /Na ₂ O molar ratio	0.50	1.26
Total water (kg/m ³)	170	149.77
Water/binder ratio	0.34	0.30
Sand (kg/m ³)	637.5	750
Compressive strength (MPa)	14.16	18

Note: For the one-part E3 formulation, Na₂O-equivalent and activator-derived SiO₂ were calculated from the 75 kg/m³ CWP-NaOH dry activator using the experimentally adopted SiO₂/Na₂O molar ratio of 0.5 and 98% NaOH purity. For the two-part benchmark, the values include contributions from both the nominal 10 M NaOH solution and commercial sodium silicate solution.

Under these respective formulation conditions, the conventional two-part FA-based geopolymer achieved a 28-day compressive strength of 18 MPa, compared with 14.16 MPa for the best-performing CWP-based one-part mortar. Thus, the one-part system reached approximately 79% of the strength of the benchmark two-part system. This indicates that the CWP-derived DA was effective in initiating geopolymerisation, although the liquid-activated reference still delivered higher strength under the same curing period.

The higher strength observed for the two-part benchmark may reflect differences in activator chemistry, water availability and soluble alkali/silicate content; however, because these parameters were not matched between the two formulations, the present comparison cannot isolate the individual cause of the strength difference [51]. In the one-part system, the activating species must first dissolve out of the thermochemically synthesised powder after water addition, which slows the overall reaction pathway. A similar early-versus-later strength contrast between commercial/solution activators and waste-derived solid activators has also been noted in recent one-part studies [39,52].

Despite this strength gap, the one-part system offered practical handling advantages and showed potential material-level sustainability benefits associated primarily with eliminating commercial sodium silicate. Fořt et al. [53] highlights the strong sustainability potential of waste-derived activators but also notes the need for more standardised methods and application-focused studies. Accordingly, the present comparison supports proof-of-concept feasibility of the CWP-derived one-part system, but does not establish application-specific suitability, handling safety, storage stability or overall environmental superiority. The conventional two-part geopolymer was included as a practical formulation-level benchmark rather than a chemically equivalent control. The two systems differ in activator form, water contribution, alkali availability and silicate supply and therefore should not be interpreted as iso-water, iso-alkali or iso-silicate formulations. Accordingly, the comparison of 28-day compressive strength is intended only to provide practical context for the performance of the CWP-derived one-part system, rather than to establish a mechanistic superiority or inferiority between the two activation routes.

Although the compressive strength achieved in this study was moderate, it is sufficient to demonstrate the feasibility of using CWP as a DA precursor for one-part geopolymer mortars. The primary objective was not to produce a high-strength binder, but to assess whether a comparatively less reactive ceramic waste stream could be transformed into a

functional DA capable of initiating one-part geopolymerisation. The lower strength relative to the conventional two-part geopolymer benchmark is expected, since one-part systems rely on the gradual dissolution of solid activator species after water addition, while two-part systems provide immediately available soluble alkaline and silicate species. Accordingly, the present results should be interpreted as proof-of-concept evidence that CWP-derived DA can activate an FA-based one-part mortar. Establishing application-specific suitability will require further evaluation of durability, shrinkage, efflorescence, flexural behaviour, long-term strength, ambient curing and scale-up performance.

4. Sustainability and Cost Analysis

4.1. Methodology

A comparative cost and material-level CO₂ analysis was conducted for the conventional two-part geopolymer and the CWP-based one-part geopolymer developed in this study. All calculations were normalised to 1 m³ of geopolymer mortar based on the mix proportions used in the experimental program. The analysis was intended to provide a material-level screening comparison rather than a complete cradle-to-gate life-cycle assessment [3,54].

The system boundary covered raw material production, chemical activators, and transportation to the mixing location. CWP was treated as a waste-derived material under the cut-off allocation principle (ISO 14044) [55], and therefore no raw material burden was assigned to the incoming ceramic waste itself. Unit cost and CO₂ values for FA, sand, NaOH, and sodium silicate were adopted from published Australian or Australia-relevant sources. A transportation distance of 50 km was assumed for all materials. It should be noted that the present analysis did not include the electricity demand associated with crushing, ball milling, sieving, and thermochemical furnace activation of CWP. Therefore, the reported reductions should be interpreted as simplified material-level estimates within the adopted system boundary.

Table 14 summarises the unit costs and CO₂ emission factors used in the analysis. The adopted values were selected from published Australian or Australia-relevant sources wherever possible to ensure consistency with the regional context of the study.

Table 14. Input values for cost and CO₂ analysis.

Material	Unit Cost (AUD/kg)	Emission (kg CO ₂ e/kg)	Reference
FA	0.08	0.02	[56]
CWP	0	0	ISO14044
Sand	0.10	0.005	[57]
NaOH	0.90	1.12	[58,59]
Na ₂ SiO ₃	1.10	0.70	

The total CO₂ emissions and material cost per cubic meter were calculated using Equations (9) and (10).

$$E_T \text{ (kgCO}_2\text{/m}^3\text{)} = \sum_i (m_i \cdot EF_i) + \sum_i \left(\frac{m_i}{1000} \cdot d_i \cdot f_{trans-CO_2} \right) \quad (9)$$

$$C_T \text{ (AUD/m}^3\text{)} = \sum_i (m_i \cdot c_i) + \sum_i \left(\frac{m_i}{1000} \cdot d_i \cdot f_{trans-cost} \right) \quad (10)$$

where (m_i = mass of material i (kg/m³), c_i = unit cost (AUD/kg), EF_i = unit CO₂ emissions (kg/kg), d_i = transport distance (km), $f_{trans-cost}$ = 0.14 AUD/t.km, and $f_{trans-CO_2}$ = 0.102 CO₂/t.km, corresponding to Australian road freight conditions).

4.2. Material-Level CO₂ and Cost Results

The calculated material-level CO₂ emissions and material costs are presented in Table 15. The conventional two-part geopolymers exhibited a total CO₂ of 170.21 kg CO₂/m³ and a material cost of 340.91 AUD/m³, whereas the CWP-based one-part geopolymer showed corresponding values of 72.43 kg CO₂/m³ and 149.86 AUD/m³. Within the adopted material-level calculation boundary, the one-part system showed approximately 57.4% lower calculated CO₂ emissions and 56% lower material cost than the conventional two-part benchmark. These percentages exclude the electricity and associated costs required for CWP crushing, ball milling, sieving and thermochemical activation and should therefore not be interpreted as complete cradle-to-gate reductions. Inclusion of these processing burdens would decrease the net environmental and economic benefit.

Table 15. Material-level CO₂ emissions and cost per m³ of geopolymer mortar.

Material	Cost (AUD/m ³)		Material-Level CO ₂ (kg CO ₂ e/m ³)	
	Two-Part	One-Part	Two-Part	One-Part
FA	40	34	10	8.5
NaOH	19.33	44.15	24.06	54.94
Na ₂ SiO ₃	196.43	-	125	-
CWP	-	-	-	-
Sand	75	63.75	3.75	3.19
Transport	10.15	7.96	7.4	5.8
Total	340.91	149.86	170.21	72.43

The main reason for this difference is the elimination of commercial sodium silicate from the one-part formulation. In the conventional two-part system, sodium silicate alone contributed 196.43 AUD/m³ to cost and 125 kg CO₂/m³ to material-level emissions, making it the dominant contributor in both categories. Sodium hydroxide was the second major contributor. In contrast, the one-part system replaced sodium silicate with CWP as a waste-derived silica source, which substantially lowered the activator-related burden. It is important to distinguish between NaOH solution dosage and actual NaOH solids when interpreting the constituent contributions. Although the two-part benchmark contained 71.43 kg/m³ of NaOH solution, this corresponded to only approximately 21.48 kg/m³ of NaOH solids. In contrast, the alkali-rich one-part E3 formulation contained approximately 49.40 kg/m³ of NaOH within the 75 kg/m³ DA. Consequently, the NaOH-related cost and CO₂ contribution of the one-part formulation were higher despite its lower total activator mass. The material-level benefit of the one-part system therefore arose from eliminating commercial sodium silicate rather than from reducing NaOH consumption.

From a cost perspective, the one-part geopolymer reduced the total material cost by 191.05 AUD/m³ relative to the two-part benchmark. From an environmental perspective, the CO₂ was reduced by 97.78 kg CO₂/m³. Within the present screening boundary, these results indicate the potential material-level benefit of replacing commercial sodium silicate with a CWP-derived dry activator; however, confirmation of the overall environmental advantage requires inclusion of process-specific energy consumption in a full life-cycle assessment.

4.3. Contribution of Individual Constituents

Figure 9 shows the contribution of each constituent to total cost and CO₂. In the conventional two-part geopolymer, commercial sodium silicate was the dominant contributor, accounting for approximately 57.6% of total material cost and 73.4% of calculated CO₂ emissions. In contrast, the one-part system eliminated commercial sodium silicate, shifting the burden primarily to sand in terms of cost (42.5%) and NaOH in terms of CO₂ emissions (75.9%). This redistribution indicates that the lower calculated material-level CO₂ burden

of the one-part system arises mainly from avoiding commercial sodium silicate, while NaOH remains the principal emissions contributor within the CWP-derived formulation. This result is consistent with the wider literature on geopolymer sustainability, where commercial alkali activators, particularly sodium silicate, are often reported as the dominant contributors to both emissions and cost [60,61].

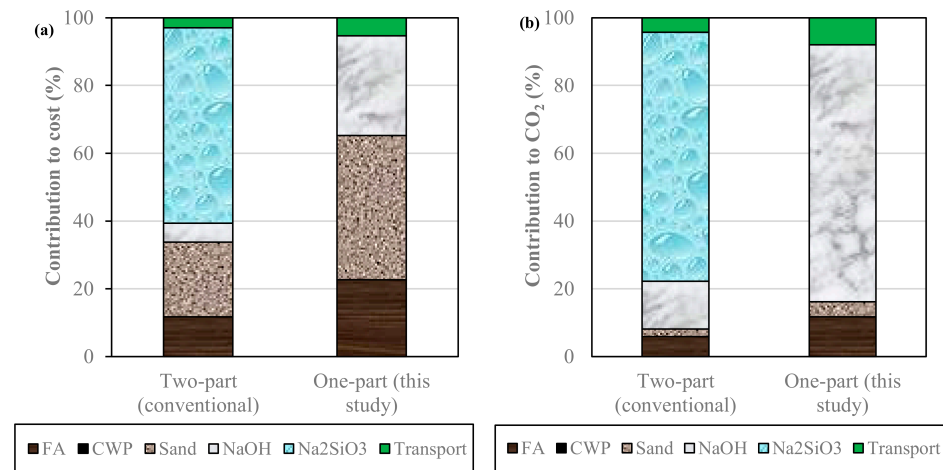


Figure 9. Contribution of individual constituent to (a) material cost and (b) CO₂ of the conventional two-part and CWP-based one-part geopolymer mortar.

Because process-specific electricity consumption was not measured during laboratory-scale DA synthesis, a break-even sensitivity was therefore used to indicate the magnitude of the omitted burden. Based on the calculated material-level differences, additional DA-processing burdens would have to exceed approximately 97.8 kg CO₂e/m³ or 191.1 AUD/m³ of mortar-equivalent to about 1.30 kg CO₂e/kg DA or 2.55 AUD/kg DA to eliminate the calculated advantage of the one-part formulation within the present comparison.

4.4. Study Limitations

Some limitations should be considered when interpreting the present results. First, the Taguchi L9 design was intended for efficient screening of the three synthesis variables and provided limited resolution for interaction effects and inferential statistical analysis. The identified optimum condition, A1B3C3, corresponded to the best-performing formulation within the L9 experimental matrix and was not independently validated through a separate confirmation synthesis; therefore, it should be interpreted as the optimum within the investigated design space rather than a universally validated optimum. Second, FTIR and SEM-EDS provide comparative structural, morphological and elemental information but do not constitute definitive phase identification; XRD and complementary dissolution measurements are required to establish the reaction products and mechanisms more rigorously. Third, performance evaluation was primarily based on 28-day compressive strength, while durability, shrinkage, efflorescence, flexural response, long-term strength and ambient-curing performance were not investigated. In addition, the two-part system served as a practical formulation benchmark rather than an iso-alkali or iso-silicate mechanistic control. Finally, the sustainability and cost assessment excluded energy associated with CWP processing, thermochemical activation and curing and therefore represents a simplified material-level comparison rather than a complete life-cycle assessment.

5. Conclusions

- This study demonstrates the proof-of-concept feasibility of converting fired ceramic waste powder into a functional dry activator for one-part fly ash geopolymer mortar

through thermochemical treatment with NaOH. The experimental and statistical results show that the effectiveness of the resulting activator is governed primarily by alkali–silica balance and activation temperature, whereas activation duration plays a secondary role within the investigated range. The best-performing formulation confirmed that CWP can support one-part activation, although the moderate strength achieved indicates that the present system remains a screening-stage binder rather than an application-optimised mortar.

- FTIR, SEM and EDS collectively provided comparative evidence that the better-performing systems were associated with greater modification of the silicate environment, a denser matrix and higher local Na incorporation; however, these observations are not interpreted as definitive phase identification. The comparison with the two-part system should likewise be regarded as a practical formulation benchmark rather than a chemically equivalent control.
- The simplified material-level assessment indicated approximately 57% lower calculated CO₂ emissions and 56% lower material cost for the CWP-based one-part system relative to the conventional two-part benchmark, primarily due to the elimination of commercial sodium silicate. These estimates exclude the energy and associated costs of CWP crushing, ball milling, sieving, thermochemical furnace activation and specimen curing and therefore should not be interpreted as complete cradle-to-gate life-cycle reductions.

Overall, the results establish proof-of-concept feasibility for using CWP as a silica-bearing feedstock in thermochemically synthesised dry activators for one-part FA-based mortar. Application-specific use, however, requires further optimisation and validation of mechanical performance, durability and ambient-curing behaviour. Future work should also include definitive phase characterisation, process-specific energy measurements and full life cycle and techno-economic assessment, together with investigation of hybrid waste-derived activator systems.

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