

## GREEN ALTERNATIVES IN CEMENT PRODUCTION: ASSESSING CALCINED CLAY IN ECO-FRIENDLY BINDER SYSTEMS

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### **Abstract**

*The cement industry contributes significantly to global  $CO_2$  emissions, driving the need for sustainable alternatives in cement production. Calcined clay has emerged as a potential replacement for clinker in cement formulations. This study examines calcined clays sourced from different regions of central Albania for its suitability in cement production. The research includes chemical composition analysis, pozzolanic activity, and compressive strength testing of cement samples containing calcined clay. The results show that some of the clays exhibit pozzolanic properties, allowing for clinker replacement of up to 10%. Clays with kaolinite content above 40% in LC3 formulations can reach strengths comparable to CEM I 42.5R (>50 MPa), but the majority of local clays show lower reactivity and performance. However, large-scale adoption faces challenges due to high investment costs and economic feasibility, therefore practical clinker substitution rates are estimated at around 6–10%, balancing technical and economic considerations. Further research is needed to optimize the calcination process and evaluate the long-term performance of these regional clays.*

**Key words:** Sustainable alternatives; calcined clay; clinker; compressive strength; pozzolanic activity.

### **Përbledhje**

*Industria e çimentos kontribuon ndjeshëm në emetimet globale të  $CO_2$ , duke nxitur nevojën për alternativa të qëndrueshme në prodhimin e çimentos. Argjilat e kalcinuara shfaqen si një zëvendësues i mundshëm për klinkerin në përzierjet e çimentos. Ky studim shqyrton argjila të kalcinuara të marra nga*

*rajone të ndryshme të Shqipërisë për përshtatshmërinë e tyre në prodhimin e çimentos. Kërkimi përfshin analizat e përbërjes kimike, aktivitetin pucolanik dhe rezistencën në shtypje të mostrave të çimentos që përmbytjë argjilë të kalcinuar. Rezultatet tregojnë se disa nga këto argjila shfaqin veti pucolanike, duke lejuar zëvendësimin e klinkerit deri në 10%. Argjilat me përmbytje kaoline mbi 40% në formulimet LC<sup>3</sup> mund të arrijnë qëndrueshmëri të krahasueshme me CEM I 42.5R (>50 MPa), por shumica e argjilave vendase tregojnë reaktivitet dhe performance më të ulët. Gjithashtu, adoptimi në shkallë të gjërë përballet me sfida për shkak të kostove të larta të investimit dhe fizibilitetit ekonomik; prandaj, normat praktike të zëvendësimit të klinkerit vlerësohen rreth 6-10%, duke balancuar konsideratat teknike dhe ekonomike. Kërkime të mëtejshme janë të nevojshme për të optimizuar procesin e kalcinimit dhe për të vlerësuar performancën afatgjatë të këtyre argjilave rajonale.*

**Fjalë kyçë:** *Alternativa të qëndrueshme; argjilë e kalcinuar; klinker; rezistencë në shtypjes; aktivitet pucolanik.*

## **Introduction**

Cement production is a major contributor to global greenhouse gas emissions, accounting for a significant share of the industry's impact on climate change. Portland cement (OPC) production depends heavily on clinker production, which is created by heating limestone at high temperatures in rotary kilns. This energy-intensive process leads to substantial CO<sub>2</sub> emissions, primarily from the decarbonization of limestone and the burning of fossil fuels for heating (Schneider et al., 2011). As a result, there is growing pressure on the cement sector to adopt more sustainable practices to reduce its environmental footprint. One promising approach is the incorporation of supplementary cementitious materials (SCMs), which can partially replace clinker in the cement mixture (Lothenbach et al., 2011). The pozzolanic qualities of those materials, such as fly ash, slag, and calcined clays, allow them to react with calcium hydroxide in the presence of water to produce more calcium silicate hydrate (C-S-H) gel, which increases the strength of concrete. The potential of calcined clays, especially those high in kaolinite, to reduce the environmental impact of cement production while maintaining or improving the mechanical performance of concrete has drawn increasing attention (Alujas et al., 2015).

Calcination is a method where clays are heated to temperatures among 500°C and 800°C, altering their mineral shape and activating their pozzolanic traits. All through this method, kaolinite, a common clay mineral, is transformed into metakaolin, a reactive compound which can improve concrete strength (Jaskulski et al., 2020). The effectiveness of calcined clays depends on the calcination temperature and the mineral composition of the raw material (Alujas et al., 2015). Recent research has demonstrated that even low-quality kaolinitic clays, once thought to be unsuitable for industrial use, can show considerable pozzolanic activity when subjected to proper calcination (Alujas et al., 2015). The mixture of calcined clays with limestone, referred to as Limestone Calcined Clay Cement (LC<sup>3</sup>), is considered as an alternative to conventional cement (Antoni et al., 2012). Additionally, LC<sup>3</sup> has demonstrated superior long-term durability compared to ordinary Portland cement (OPC), particularly in aggressive environments such as those containing sulfates and chlorides (Dhandapani et al., 2018).

Albania, located in the Balkans, has several clay deposits, particularly in the central areas around Fushë Krujë, Durrës, and Elbasan. These regions hold significant potential for sourcing calcined clay due to their close proximity to cement plants and abundant natural clay reserves. However, the suitability of these clays for cement production depends on factors such as their chemical composition, particularly the kaolinite content, and their performance when used in cement mixtures (Pinheiro et al., 2024). Previous research indicates that kaolinite-rich clays with Al<sub>2</sub>O<sub>3</sub> levels exceeding 17% demonstrate considerable pozzolanic reactivity after calcination. The chemical composition, especially the silica (SiO<sub>2</sub>) to alumina (Al<sub>2</sub>O<sub>3</sub>) ratio, plays a crucial role in the performance of calcined clay within cement-based systems.

**Figure 1.** Clay quarries in Albania.



Metakaolin, an amorphous aluminosilicate produced through the calcination of kaolinite-rich clays (typically containing 30–40% kaolinite), contributes to secondary hydration reactions, improving both strength and durability of concrete (Scrivener et al., 2018; Alujas et al., 2015).

The widespread use of LC<sup>3</sup> technology faces many challenges. One extensive task lies in developing lower-priced calcination techniques while keeping strict first-rate standards to gain the most pozzolanic reactivity of the clays. Additionally, the supply of kaolinitic clays, which might be vital for LC<sup>3</sup> manufacturing, is a restricting factor. In areas wherein those clays aren't conveniently reachable, huge-scale production may become economically not viable (Joseph et al., 2016). Scaling up LC<sup>3</sup> production also requires substantial upgrades in infrastructure and policy reforms. Analyzing the chemical composition of the clays and assessing the compressive strength of cement mortars mixed with calcined clay are the main focuses of this research. For reference, performance comparison is also made with Greek clay (S.1), which is known for its pozzolanic reactivity. The findings are intended to determine important areas for further study and development, as well as the feasibility of using regional Albanian clays for the production of sustainable cement.

## 1 Methodology

### 1.1 Materials and methods

This section describes the experimental study of the chemical composition, pozzolanic activity, compressive strength as well as its long-term prediction for different calcined clay samples. The main purpose is to determine the mechanical strength of the samples formed with the above-mentioned materials and their comparison with ordinary Portland cement, in order to evaluate potential suitability in cement applications.

The research included the collection and analysis of clay specimens from five distinct regions in central Albania. To facilitate comparative analysis, an additional clay specimen sourced from Greece (S.1) was incorporated, owing to its extensively documented high kaolinite concentration and pozzolanic properties. The selection of sampling sites was carried out upon their geographical proximity to cement manufacturing facilities (within a radius of

150 km) and the historical prevalence of substantial clay reserves in these regions.

Central Albania, with particular emphasis on the Fushë-Krujë area (3 different samples from this region), possesses a longstanding tradition of clay extraction for brick and cement manufacturing.



**Figure 2.** Sampling locations of clays (S.2–S.8) in Albania.

The parameters for the selection of clay samples were as follows:

1. Geographical Considerations: Clay deposits needed to be located within a 150 km radius of active cement factories to reduce transportation costs and improve industrial scalability.
2. Chemical Properties: A loss on ignition (LOI) <11% and alumina ( $\text{Al}_2\text{O}_3$ ) content >17% were used as preliminary benchmarks.

3. Volume Availability: Priority was given to deposits with sufficient clay reserves to enable sustainable industrial-scale extraction.



**Figure 3.** Some clay samples taken for analysis.

## 1.2 Sample Preparation

Proper sample preparation is crucial to ensure consistency and reliability in testing. The following steps were followed for the preparation of the clay samples in this study:

1. Drying and Crushing: To eliminate moisture content that could interfere with subsequent processes, the raw clay samples were dried in a Controls 10-D1396 thermostat at 105°C for 2.5 hours. After drying, the samples were crushed with a PY 175 jaw crusher to reduce the particle size to below 10 mm, ensuring uniformity for the grinding stage.
2. Grinding: Following the crushing step, the clay samples were finely ground using a GJ-100-1 ring mill to achieve the desired particle size for calcination. The fine grinding of the samples increases the surface area, which is essential for enhancing their reactivity during calcination and pozzolanic activity tests. The particle size distribution (PSD) of the samples was determined using a Malvern Mastersizer Scirocco (2015). PSD analysis is a crucial step, as the reactivity of pozzolanic materials is often linked to their particle size, with finer materials generally exhibiting higher reactivity (International Organization for Standardization, 2008).

The aim of the preparation process was to ensure that all clay samples were uniform in composition and met the necessary physical requirements for the calcination and analytical stages of the study.



**Figure 4.** a). Jaw crusher PY 175 and, b). Ring mill GJ-100-1.

### 1.3 Chemical Composition Analysis (XRF)

The oxide composition of the clay samples (CaO, SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, Fe<sub>2</sub>O<sub>3</sub>) was determined by X-ray Fluorescence (XRF) using a Cubix PANalytical spectrometer (2005 model). The emitted fluorescent X-rays were measured to quantify the main oxides, providing data for assessing the clay's suitability as a supplementary cementitious material (SCM).

### 1.4 Loss on Ignition Analysis of the Clay Samples, Kaolinite Content & Pozzolanic Activity Determination

Loss on Ignition (LOI) measures the weight loss of a clay sample when heated, indicating the removal of water and volatile compounds. It also verifies the effectiveness of calcination and the clay's suitability for cement applications. Samples were heated in a Nabertherm muffle furnace at 950°C for 1 hour, and weight loss was used to assess dehydration and volatilization. Kaolinite content is important for evaluating the clay's potential to form reactive metakaolin for pozzolanic activity. It was determined using a thermogravimetric method: clay samples were heated in a muffle furnace at 200°C, 400°C, and 600°C, and weight loss at each stage was used to calculate kaolinite content using (Eq.1). The kaolinite content was measured using thermogravimetric method, where clay samples were heated in a muffle furnace to approximately 800°C for 1 hour to remove impurities.

$$w\ t\ %\ k\ a\ o\ l\ i\ n\ i\ t\ e = \frac{Wt\ 400^{\circ}\text{C} - Wt\ 600^{\circ}\text{C}}{Wt\ 200^{\circ}\text{C} - Wt\ i} \times 7.17 \times 100 \quad (\text{Eq.1})$$

The pozzolanic activity test evaluates the ability of calcined clay to react with calcium hydroxide (Ca(OH)<sub>2</sub>) in the presence of water, forming secondary

hydration products, such as calcium silicate hydrate (C-S-H), which contribute to the strength and durability of cement-based materials. The pozzolanic activity test, conducted on the clay samples according to the S SH EN 196-5:2011 standard, aimed to evaluate the consumption of calcium hydroxide  $[\text{Ca}(\text{OH})_2]$  released from silicate reactions and its following uptake by the aluminates in metakaolin. The consumption of  $\text{Ca}(\text{OH})_2$  was monitored over time, and the reduction in its concentration indicated the pozzolanic activity of the clay. Clay samples were dried, ground, and calcined at  $800^\circ\text{C}$  for 1 hour, inducing kaolinite dihydroxylation and forming reactive metakaolin (Tironi et al., 2012). Clays with loss on ignition (LOI)  $>11\%$  were excluded from calcination due to reduced alumina ( $\text{Al}_2\text{O}_3$ ) and increased calcium oxide (CaO) and magnesium oxide (MgO), which negatively affect pozzolanic performance and cement durability (Zunino et al., 2022; Kakali et al., 2001). Other clays, despite not meeting the  $\text{Al}_2\text{O}_3 >17\%$  criterion, were calcined because of their proximity to the cement facility.

### 1.5 Mortar Sample Preparation and Compressive Strength Testing Procedure

The materials used in the mix consist of 520 gr of CEM I 42.5R, 10 gr of gypsum, 170 gr of limestone, and 300 gr of calcined clay in order to form 1000 gr of LC<sup>3</sup> cement. Each ratio for the preparation of the three samples (prisms) should consist of  $450 \pm 2$  g cement,  $1350 \pm 5$  g sand, and  $225 \pm 1$  g water (European Committee for Standardization, 2016).

Cement, water ( $\pm 1$  g and  $\pm 1$  ml precision), and sand were mixed following a standard sequence: initial cement–water mixing, gradual sand addition, and high-speed mixing with one pause to scrape the bowl. Fresh mortar was cast into  $40 \times 40 \times 160$  mm prism molds in two layers, each compacted with 60 strikes. Molds were covered, stored in a humidity cabinet, then cured in water at  $20 \pm 1^\circ\text{C}$ . Specimens for 24 h tests were demolded within 20 minutes before testing (European Committee for Standardization, 2016).

Prisms were first fractured in bending, and the half-prisms were then loaded in compression. Each specimen was aligned within  $\pm 0.5$  mm and tested at a loading rate of  $2400 \pm 200$  N/s after a 100 kN preload. Strength was calculated as load divided by area, expressed in MPa (European Committee for Standardization, 2016).



**Figure 5.** a). Mixer, b). Compressive strength machine, c). Prisms (40 mm x 40 mm x 160 mm).

## 2 Results

### 2.1 X-ray Fluorescence (XRF) Results of Sample Oxide Composition

The results for the oxide composition of the clay samples, loss on ignition and kaolin content are shown in Table 1, and a comparison of the main oxides for all samples is presented graphically in Fig.6.

**Table 1.** XRF analysis results for several clay samples.

Sample	SiO <sub>2</sub> [%]	Al <sub>2</sub> O <sub>3</sub> [%]	Fe <sub>2</sub> O <sub>3</sub> [%]	CaO [%]	MgO [%]	SO <sub>3</sub> [%]	K <sub>2</sub> O [%]	Na <sub>2</sub> O [%]	L.O.I [%]	Kaolinite [%]
S.1	54.5	20.5	5.9	2.0	1.9	0.3	2.8	1.4	8.1	30.5
S.2	55.8	13.9	7.4	4.2	3.2	0.25	2.3	1.3	9.1	19.8
S.3	75.0	7.9	4.3	2.5	2.2	0.45	1.5	1.0	2.5	10.7

S.4	65.2	13.4	6.5	2.7	1.2	0.24	1.3	1.2	6.4	22.7
S.5	43.7	12.3	6.8	11.2	6.0	0.20	2.6	1.5	13.9	17.9
S.6	47.8	12.7	7.1	9.0	4.5	0.24	2.4	1.3	11.8	18.5
S.7	45.8	11.2	6.1	11.2	4.1	0.20	2.9	1.6	12.8	17.4
S.8	48.7	14.4	2.8	2.5	1.5	0.22	2.5	1.3	8.3	7.2

**Table 2.** XRF analysis results for CEM I 42.5R (S.0, control sample).

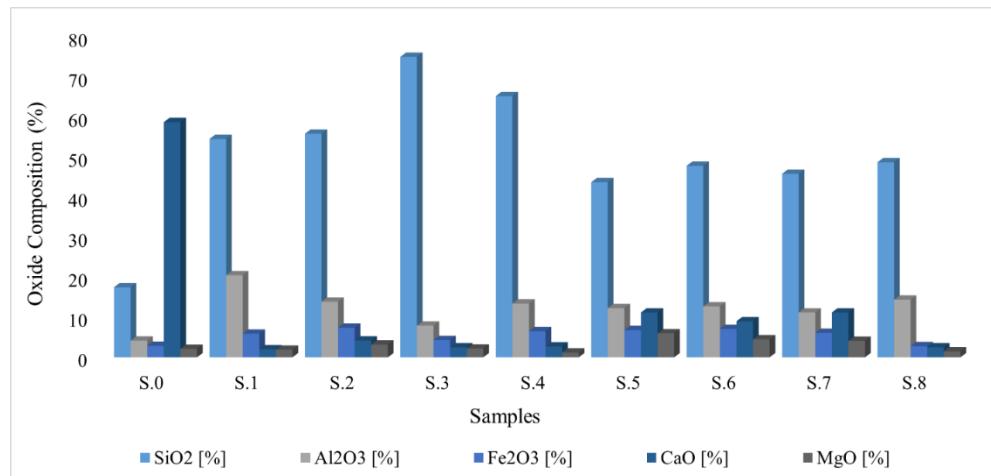
Oxide	Al <sub>2</sub> O <sub>3</sub>	CaO	SiO <sub>2</sub>	MgO	Fe <sub>2</sub> O <sub>3</sub>	SO <sub>3</sub>	K <sub>2</sub> O	Na <sub>2</sub> O	TiO <sub>2</sub>	L.O.I
%	4.17	58.72	17.49	2.14	2.89	2.52	0.68	0.27	0.23	9.88

The XRF results show that samples S.1, S.2, and S.4 exhibit pozzolanic potential due to their high kaolinite content and well-balanced alumina levels, making them highly suitable for metakaolin production. These clays have moderate L.O.I. values, supporting their effectiveness in calcination and subsequent use in cementitious applications.

On the other hand, S.5, S.6, and S.7 samples have high L.O.I. values and significant carbonate content, indicating that their reactivity could be negatively impacted during calcination. Their high CaO and MgO levels further limit their potential for pozzolanic activity. Additionally, S.3 and S.8 samples, despite their high silica content, have low kaolinite and alumina levels, making them less suitable for pozzolanic applications.

A more comprehensive analysis and discussion of these findings will follow in the discussion section.

**Figure 6.** Main oxides comparison for all samples (S.0-S.8).

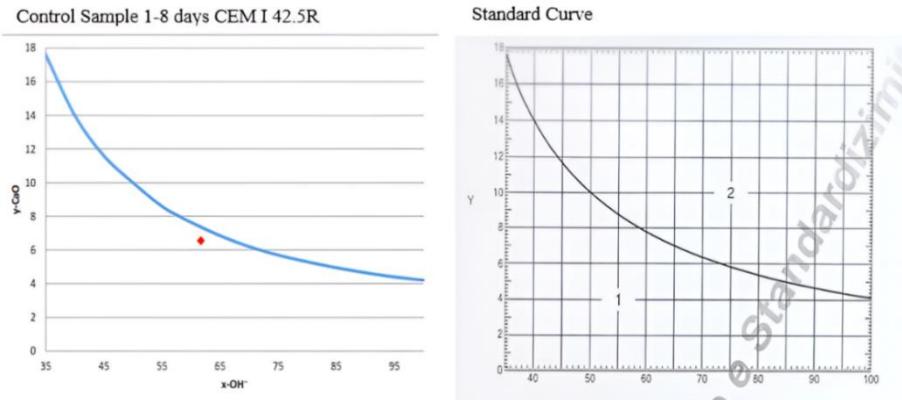


## 2.2 Pozzolanic Reactivity Results for the Two Best-Performing Samples

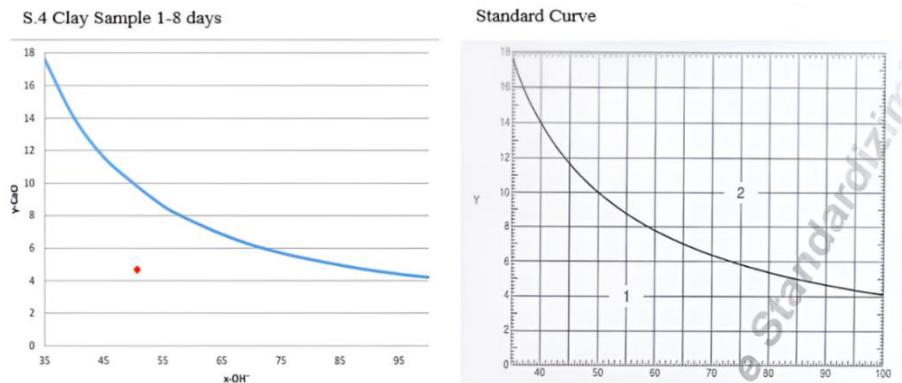
The pozzolanicity test was conducted to confirm the reactivity of the clay samples S.1 and S.4 and verify their potential as effective supplementary cementitious materials. While the XRF analysis provided insights into their oxide composition, where it was determined that those two samples show high kaolinite content, which is critical for pozzolanic reactivity, along with a favorable silica-to-alumina ratio, essential for the formation of metakaolin. Also, their moderate loss on ignition (L.O.I.) values suggest that they will maintain stability during calcination, the pozzolanicity test serves as a practical assessment to ensure that these clays exhibit the desired pozzolanic behavior under real-world conditions.

The shift of the red point indicates that the calcined clay demonstrates pozzolanic activity, as supported by the compressive strength results for the LC<sup>3</sup>-50 formulation. In the case of S.1, the point shifts even further, signifying a higher level of pozzolanic activity compared to the other samples. For pozzolanicity to be confirmed, the red dot must fall below the standard curve, which serves as a reference. These results are compared to the control sample and further validated by the standard curve. The compressive strength data consistently show improved performance for the S.1 clay sample, highlighting

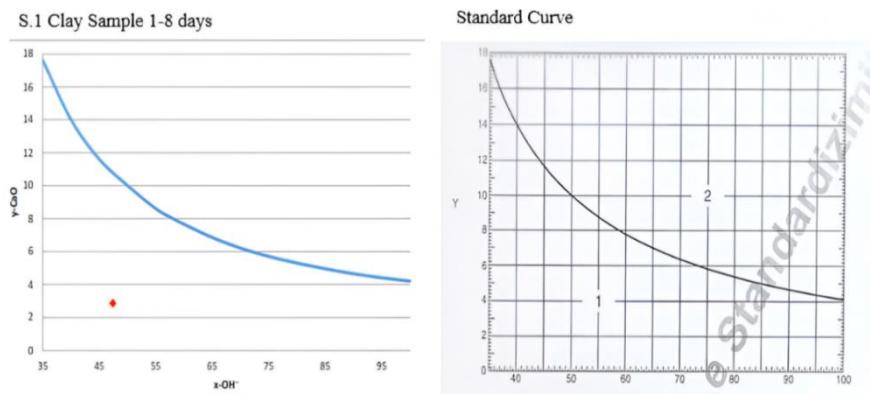
its superior pozzolanic potential and its suitability for cementitious applications.



**Figure 7.** Pozzolanicity graph for the control sample CEM I 42.5R (S.0).



**Figure 8.** Pozzolanicity graph for the S.4 clay sample.

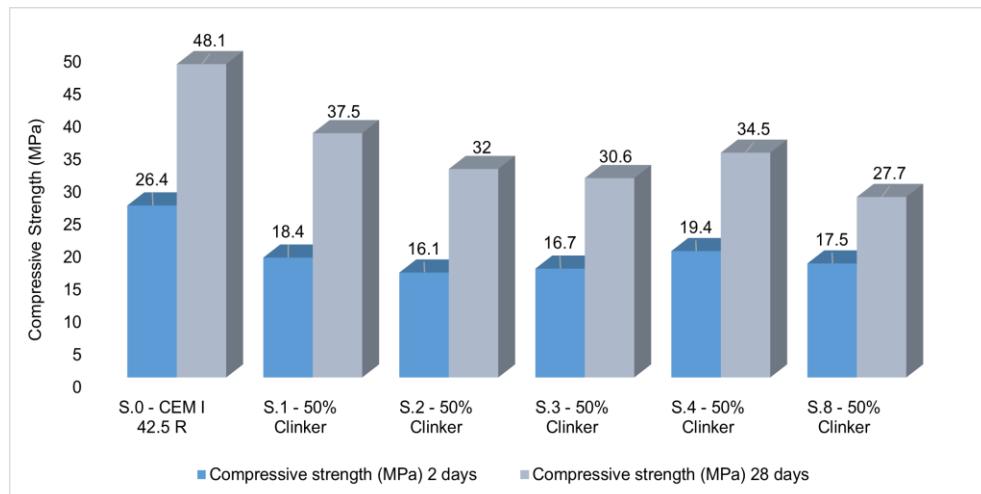


**Figure 9.** Pozzolanicity graph for the S.1 clay sample.

## 2.3 Mechanical Strength Data

### 2.3.1 Compressive strength results

The results of the compressive strength for the different clay samples after 2 and 28 days are presented in the following graph. Prior to the formation of the samples, all clays were calcined at 800°C for 1 hour to activate their pozzolanic properties. This calcination process enhances the reactivity of the clays, ensuring that the subsequent compressive strength analysis reflects their true performance in cementitious applications.



**Figure 10.** Compressive strength analysis results for all samples after 2 and 28 days of curing.

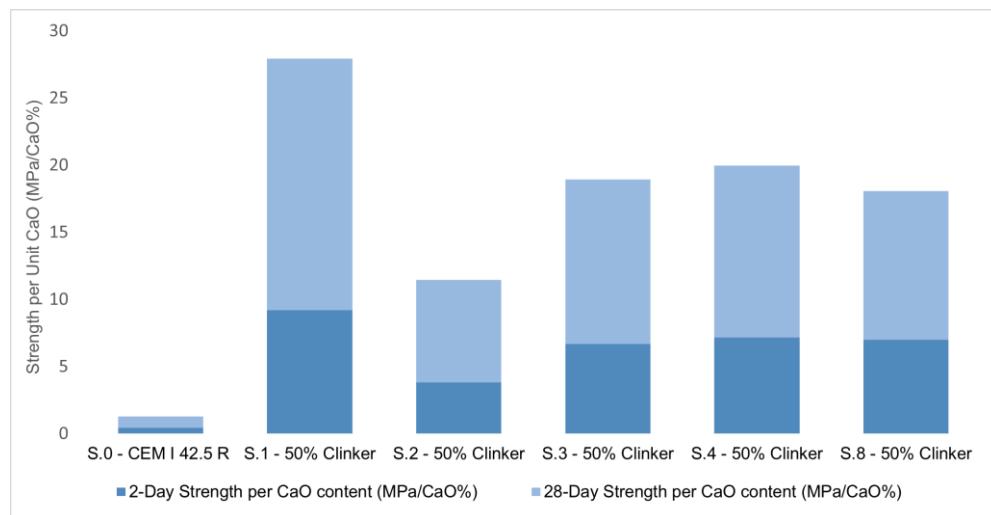
Among the clay samples, S.1 clay type demonstrated the highest compressive strength at both 2 days (18.4 MPa) and 28 days (37.5 MPa), followed closely by S.4. The other clay samples, including S.8 and S.2, showed lower strengths, indicating less pozzolanic activity or lower reactivity in the cement mixture.

### 2.3.2 Compressive strength performance per unit of calcium oxide

Illustrating strength efficiency per unit CaO content is important, as it provides a normalized measure of how effectively the calcium oxide in each binder contributes to mechanical performance. This allows for a direct comparison

between different binder systems, independent of variations in overall CaO content, and helps identify formulations that maximize strength development while minimizing clinker usage. The graph below presents the cumulative compressive strength per unit CaO content at 2 and 28 days for all binder systems.

The results show that the blended systems incorporating S.1 and S.4 clay types exhibit the highest strength per unit CaO at both 2 and 28 days, outperforming the control CEM I 42.5R. This indicates that despite their low CaO content, these materials are highly reactive and efficient in forming strength-contributing hydrates. The superior performance can be attributed not only to their high kaolinite content (e.g., 30.5% for S.1 and 22.7% for S.4), but also to the favorable combination of amorphous silica and alumina that enhances pozzolanic reactivity.



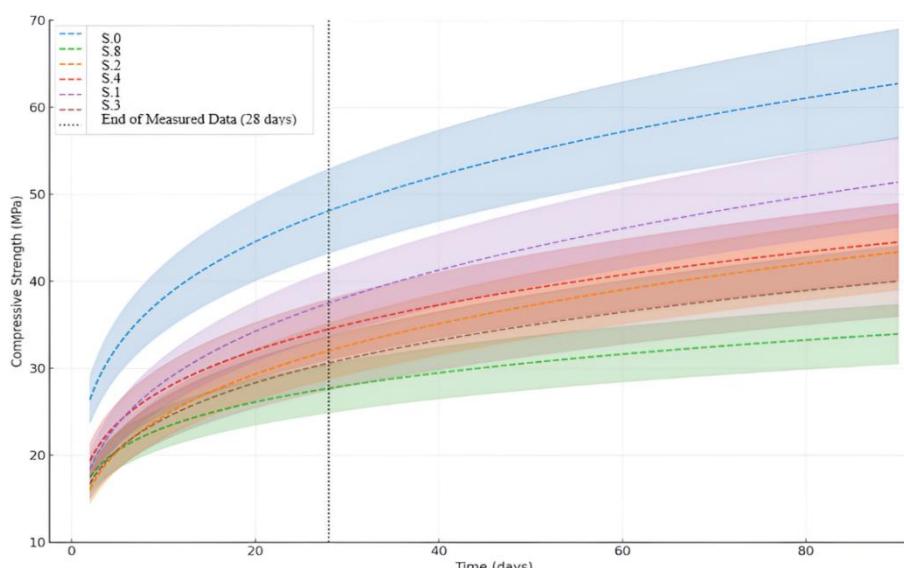
**Figure 11.** Strength contribution of CaO in binder systems at 2 and 28 days.

In contrast, S.2 and S.3 samples show lower strength/CaO ratios, likely due to lower kaolinite content, reduced calcined reactivity, or a higher fraction of inert or crystalline phases. The control sample (CEM I 42.5R), while high in absolute strength, shows very low CaO efficiency, as a large portion of its strength derives from high clinker content rather than optimized CaO use. These results suggest that clay-based systems can significantly improve the ecological footprint of cement by reducing clinker factor without

compromising mechanical performance, provided that their reactivity and mineralogical profile are favorable.

### 2.3.3 Long-term compressive strength prediction

In the above sections, the compressive strength was measured for all samples up to 28 days of curing. To assess the long-term strength development of the binders, the evolution of compressive strength was predicted up to 90 days using a power-law model ( $y = a \cdot x^b$ ), with parameters  $a$  and  $b$  derived from the experimental data up to 28 days. The graph below shows the predicted evolution up to 90 days for CEM I and the five clay binder systems, with shaded areas representing  $\pm 10\%$  uncertainty bounds for each prediction.



**Figure 12.** Compressive strength prediction of CEM I and clay-based binder systems up to 90 days.

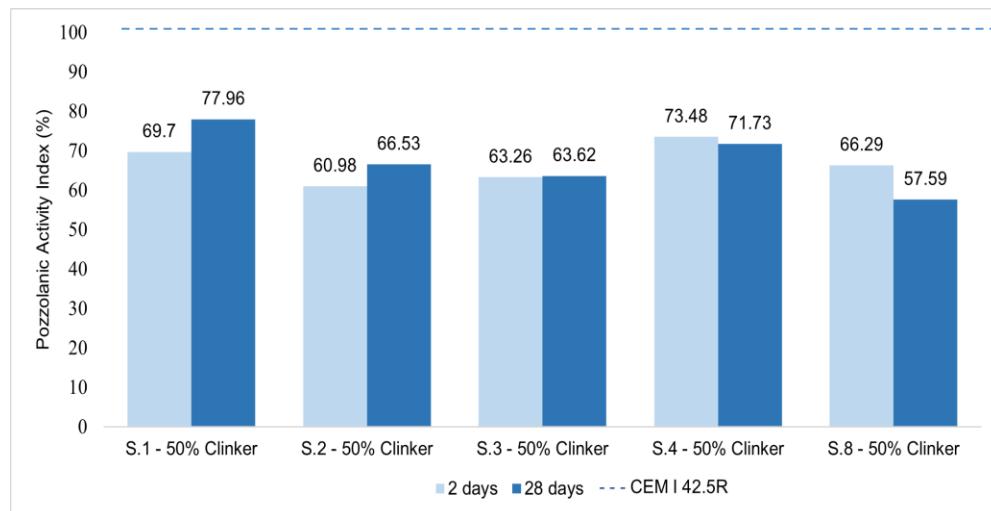
CEM I (S.0) demonstrates the highest rate of strength development over time, followed by S.1 and S.4 samples. The curves show that while some clay blends start with lower early strength, several, particularly S.2 and S.1, exhibit a steeper growth trajectory, narrowing the performance gap at later ages. This

highlights the long-term pozzolanic activity of these materials and their potential to serve as sustainable alternatives in cementitious systems.

### 2.3.4 Pozzolanic activity index (PAI)

The graph presented below provides a comparison of the percentage of resistance (compressive strength) exhibited by each binder system in relation to the control sample. This comparison highlights the variations in performance between the different clays, offering insight into their relative effectiveness as pozzolanic materials. By evaluating the resistance percentages or else pozzolanic activity index, it is possible to identify which samples exhibit the most promising results in terms of strength development over time.

The results show that S.1 type demonstrates the highest performance, with 69.7% of the control sample's resistance after 2 days and 77.96% after 28 days, indicating its strong pozzolanic potential. S.4 also performs well, achieving 73.48% resistance at 2 days and 71.73% at 28 days. Other formulations, such as S.2 and S.3, show moderate resistance, ranging from 60% to 66% at both time intervals. In contrast, S.8 exhibits the lowest resistance, particularly after 28 days, suggesting it has the least pozzolanic activity among the samples tested. These results highlight the varying performance of the clays in cementitious applications.



**Figure 13.** Pozzolanic activity index at 2 and 28 days for clay-based binders, compared with CEM I 42.5R (100%).

## Discussions

The XRF results provide a detailed view of the elemental composition of the clay samples, allowing us to assess their pozzolanic potential. S.1 and S.4 samples exhibit high kaolinite content, indicating strong potential for metakaolin formation. S.6 and S.2 also show promising kaolinite levels, with moderate alumina content suggesting good reactivity upon calcination.

However, based on the L.O.I. rubble (>11%), S.5, S.6, and S.7 samples were not calcined, as their high L.O.I. values indicate significant water loss and possible carbonate decomposition, which can lead to undesirable changes in their oxide composition. This aligns with their higher CaO and MgO content, suggesting the presence of carbonate phases that might limit their pozzolanic efficiency. In contrast, S.1, S.2, and S.4 samples show moderate L.O.I. values, supporting their suitability for calcination and pozzolanic applications. MgO content is generally low across the samples, with the highest values found in S.5, S.6, and S.7 samples.

A higher MgO content can indicate the presence of dolomitic impurities, which may reduce pozzolanic efficiency or affect cement hydration negatively. S.3 is silica-rich but has low alumina and kaolinite content, making it less suitable for metakaolin production. Likewise, S.8, with the lowest kaolinite content among all samples, shows limited reactivity and is less suitable for pozzolanic applications.

Overall, the XRF results indicate that samples S.1, S.2 and S.4 are the most promising for pozzolanic activity. S.6 samples also shows good potential; however, since its L.O.I. exceeds 11%, it was not calcined, limiting its usability in pozzolanic applications. S.5 and S.7 samples have high CaO and MgO levels, along with L.O.I. values above 11%, indicating the presence of carbonates, which negatively impact their pozzolanic reactivity and led to them not being calcined. S.3 and S.8 samples, despite their silica content, have low kaolinite and alumina levels, making them less suitable for pozzolanic applications. Overall, samples S.1, S.2, and S.4 stand out as the most favorable candidates for metakaolin production and cementitious applications.

The pozzolanic test, was crucial in confirming the ability of the selected clays to react with calcium hydroxide in the cement matrix. Among the samples, S.1 and S.4 performed the best due to their high silica and alumina content, which significantly enhanced their pozzolanic reactivity. The S.1 sample showed the most substantial shift in the red dot, indicating a higher pozzolanic activity compared to the others. The compressive strength results, both at 2 days and 28 days, further validated these findings, with S.1 and S.4 exhibiting the highest compressive strengths. These samples demonstrated a consistent and significant improvement in strength over time, reflecting their ability to contribute effectively to the cement matrix. The high pozzolanic activity of these clays, as shown by both the red point shift and compressive strength results, underscores their potential for enhancing the long-term strength and durability of concrete.

By identifying which clays show the most promise in terms of compressive strength and pozzolanic reactivity, this study lays the foundation for future research aimed at optimizing these materials for industrial-scale applications.

### **Conclusions**

The findings of this study highlight both the potential and the limitations of using locally available calcined clays as supplementary cementitious materials. The results indicate that only two of the tested clays were able to achieve a compressive strength classification of 32.5R when replacing 50% of clinker, highlighting the challenges in identifying suitable materials for large-scale clinker substitution.

The pozzolanic activity analysis confirmed that materials such as schist (S.8) and pumice (S.3) are highly stabilized and show little to no reactivity, making them unsuitable as supplementary cementitious materials. Despite some samples showing promising results, the tested clays remain significantly below the performance of highly reactive calcined clays, such as those used in LC<sup>3</sup> formulations, which can reach compressive strengths comparable to CEM I 42.5R (above 50 MPa). This suggests that the studied clays, while somewhat reactive, do not possess sufficient kaolinite content (>40%) to achieve optimal pozzolanic performance.

The comparison with the control sample (CEM I 42.5R) further validated that certain clays, retained a significant percentage of compressive strength,

supporting their potential role in blended cement formulations. However, the overall substitution potential of the local clays studied remains limited, with a practical replacement rate of approximately 10%, which is significantly lower than the 50% achieved in optimized LC<sup>3</sup> formulations. While the current reactivity of local clays may not match that of highly optimized alternatives, their use in blended cements could still offer significant environmental benefits, particularly in regions with limited access to other supplementary materials.

The economic viability of LC3 is limited by the high investment required and the relatively small local availability of reactive clay. The low potential for clinker replacement and long return on investment make industrial adoption unlikely without strong market demand or regulatory incentives.

Further research could improve the feasibility of incorporating calcined clay in cement production. Optimizing calcination conditions and mix design may enhance reactivity and performance. Investigating long-term durability, hydration kinetics, and mechanical properties will provide insights for practical applications. Expanding sampling and conducting pilot-scale trials with advanced characterization are needed to validate laboratory results, as the limited number of samples was a key study limitation. Calcined clays offer a pathway to reduce CO<sub>2</sub> emissions in cement, but successful implementation requires balancing performance, cost, and environmental benefits. Although full-scale LC3 use is challenging, these findings highlight the potential and limitations of calcined clays for sustainable cement production.

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