



Sustainable Render Mortars Using Limestone Calcined Clay Cement (LC3): Mechanical, Microstructural, and Environmental Evaluation

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Abstract

The cement industry significantly contributes to global greenhouse gas emissions. Since their limestone calcination process releases substantial amounts of carbon dioxide, Portland cement (OPC) production is a major environmental concern. This study evaluated the potential of limestone calcined clay cement (LC3) as a sustainable alternative to ordinary Portland cement (OPC), specifically in its application for render mortars. Three LC3 formulations were developed, designated as A, B, and C, featuring different proportions of Egyptian raw materials, including clinker (CK), calcined clay (CCL), limestone (LS), and 5% gypsum. These formulations were tested for their physical, mechanical, and microstructural properties. Results show that the most efficient low-energy binder that reduces carbon emissions while maintaining good strength is LC3(C) (with 50% clinker replacement). At 28 days, its compressive and flexural strengths reached 48.7 MPa and 6.9 MPa, respectively. In render applications, the R25B mix (25% LC3 with black pigment) offered the best combination of low cost, performance, and environmental benefits. It met EN998 standards, achieved strong compressive and flexural strengths, and exhibited better adherence and thermal performance compared to traditional mortars. Additionally, black pigment showed better compatibility with LC3 than red, enhancing workability and aesthetics. The research highlights the potential of LC3, especially LC3(C), as an eco-friendly solution for reducing the cement industry's environmental footprint and offers a practical approach for adopting sustainable practices in construction and addressing climate change.

Keywords: calcined clay, LC3, renders mortar, Pigments.

1. Introduction:

Ordinary Portland cement is one of the biggest products with a negative environmental impact and carbon footprint worldwide, as one ton of cement manufacturing produces about 860 kg of CO₂. In addition to using more energy, the calcination of limestone contributes to carbon emissions, which account for almost 7% of global human activity [1]. Nowadays, every study searches for methods to employ more cementitious materials, including furnace slag, silica fume, metakaolin, calcined clay, and others. Utilizing supplemental cementitious materials (SCMs) has tremendous potential to reduce carbon emissions and the consumption of virgin resources in the cement production process, especially for developing countries. Wider deployment is hampered by the restricted availability of SCMs in many nations or areas. Currently, over 80% of the SCMs used to reduce the clinker factor in cement are composed of fly ash, slag, or limestone [2]. However, the majority of these by-products are also undergoing an environmentally friendly change, and their availability does not align with the large-scale cement plant. In this case, limestone and kaolinite clays became significant sources of SCMs because of their extensive availability [3].

Limestone-Calcined Clay Cement (LC3), an innovative, affordable, and environmentally friendly binder made from calcined clay and limestone powder, was recently created. Calcined clays offer enormous potential to expand the use of supplemental cementitious materials as a partial replacement of clinker in cement and concrete, especially when combined with limestone (LC3 technology) [4]. Calcined clay, which acquires its characteristics when heated to 700–800 °C, and the plentiful global limestone supplies are the two main components of LC3 [5,6]. Adding limestone and calcined clay to the cement matrix can create a material with mechanical properties similar to those of pure Portland cements. Moreover, LC3 can reduce the amount of clinker and CO₂ emissions [7,8]. Due to the massive amounts of calcined clay found in emerging nations, the amount of limestone is widely distributed and always available. Many countries that rely on their own supply of raw materials prefer LC3 due to their inability to get substantial quantities of conventional supplemental cementitious materials, such as fly ash or slag [9–11]. When limestone carbonate and alumina from clay calcination are combined, carbo-aluminate phases are created, strengthening the material's characteristics and increasing its durability. [12]. Although the flexure and compressive strengths of mortars made of calcined clay and limestone, at early ages, decline as the amount of calcined clay and limestone increases, it won't have any detrimental effects on their long-term strength [13].

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Using calcined clay improves the pozzolanic, leading to improved long-term strength growth and superior resistance against aggressive environmental factors, including chloride penetration and sulfate attack [14]. Being permeable, limestone enhances workability. However, compared to OPC, its usage in calcined clay typically results in a higher water requirement. The reason is that it is much finer and has water-retaining qualities. In this case, it is crucial to use superplasticizers to produce the necessary new qualities. [15,16]

According to the European standard EN 197-1, calcined clay and limestone combinations are currently permitted up to a 65% clinker concentration (LC3-65): CEM IIB M(Q-LL). The proposed extension of this European standard with the CEM IIC class will allow down to 50% clinker, but does not include calcined clays in the list of substitute materials. However, it should be possible to include calcined clays (Q) in the future [17]. LC3s are already covered in many standards, but the optimal level of clinker content for cost and CO₂ savings may be somewhat less than current standards allow. LC3 blends are permitted by ASTM C595 to have a minimum clinker content of 45%, a pozzolan content of $\leq 40\%$, and a limestone content of $\leq 15\%$, though slag can be used to substitute these percentages. Many other countries have more flexible standards and Cuba has recently adopted a new standard covering LC3 [3]. Industrial trials conducted in Cuba and India have demonstrated the success of producing cement with only 50% clinker, mixed with a mixture of calcined clay and limestone. The cements produced had mechanical performance similar to a CEM I Portland cement, with clinker content above 90% [18]. The external wall coating, or the skin of the building envelope, plays a key role in offering the first or main layer of protection against climate-related problems. Traditional wall covering rendering mortars consist of a blend of aggregate, water, and one or more binders [19].

There are several factors for growing interest in LC3: (1) abundant reserves of limestone; (2) suitable clays are abundant; (3) it is very compatible with existing cement production processes that will require little modification; (4) it will operationally feasible in an existing plant; (5) similar or superior mechanical properties when compared to OPC; (6) better durability to chemical attack (e.g., sulfate, chloride); (7) better workability from the fine limestone and calcined clay particles; and (8) substantial environmental benefits, including reduction of CO₂ emissions and energy usage [20]. Depending on the amount of iron in the raw clay, the first color creation of calcined clay showed a variety of tints from grey to reddish, giving the cement system reddish tones. Initially, the unique coloring was seen as a barrier to LC3's widespread market acceptance. When the reaction was conducted under reducing conditions, researchers found that clays containing iron impurities produced magnetite through the calcination process rather than hematite. When calcined, the clay and the resulting cement both turn grey, like the common look of regular cement [21]. Although adding inorganic pigments to mortar has visual advantages, many intricate interactions affect the material's workability and hardened qualities [22-28]. The high pigment surface area (roughly ten times finer than cement) that adsorbs some of the mixing water and reduces fluidity is responsible for the observed 39% decrease in flow rate of red-colored mortars when the pigment concentration was raised from 3% to 12% of cement mass. Therefore, the allowable mixing ratio for red pigment must be set at less than 9% to achieve the right fluidity. It was discovered that when black pigment B was combined with cement mortar, the flow decreased; nevertheless, this should not be taken into account, because there was almost no change in fluidity. However, in this regard, the mixing ratio should be set to less than 6% [22].

This study introduces a novel, cost-effective, and low-energy approach to producing a cementitious binder by substituting ordinary Portland cement (OPC) clinker with a blend of Egyptian gray clay, limestone, and gypsum. To create the limestone calcined clay cement (LC3), this research explored various ratios of calcined clay (25%, 30%), limestone (10%, 15%), and a constant 5% of gypsum within the clinker. Furthermore, the investigation extended to evaluate the performance of this newly developed LC3 binder as a colored mortar for decorative applications, besides being a low-energy binder. In accordance with EN 998-1/2010 standards, the mechanical and microstructural properties of these mortars were assessed after incorporating 2% inorganic pigments in black and red. The results confirm that this render mortar meets the mechanical performance requirements of EN 998-1/2010, suggesting its potential for widespread use. So, the research underscores the viability of LC3 as a colored render mortar, offering a sustainable alternative that reduces the carbon footprint of traditional cement mortars while enhancing building aesthetics and aiding in the preservation of older structures.

2. Materials and methods investigation.

2.1 Materials

The materials utilized in this study were limestone supplied by El-Arish Cement Company, located in the center of Sinai, Egypt, and ordinary Portland cement (OPC), which is composed of 95% clinker and 5% gypsum. The gray clay came from Egypt's Ras Abu Zneima Zone, which is located south of Sinai. Calcined clay (CCL) was made in a laboratory as illustrated in Figure 1. Dry Mix Company, El 10th of Ramadan, Sharqia, supplied all the materials needed to prepare the dry renders, including blank, hydroxypropyl methylcellulose (HPMC), limestone (LS), sand, and inorganic pigments (red and black as ferric oxide) that serve as fillers. The initial raw materials' particle size distribution is shown in Figure 2. It is observed that the particles in all materials are almost identical (Figure 2-a), with the majority of particles falling between 0.02 and 0.1 mm. While Sand's particle size (Figure 2-b) shows the majority of particles falling between 0.2 and 1 mm. XRF was utilized to ascertain the chemical composition of the raw materials employed in this project, as indicated in Table 1.

2.1.A Preparation of Calcined Clay:

The preparation of calcined clay, a basic ingredient in the preparation of LC3 binder, is shown in Figure 1. After being crushed to a size of 1 mm, gray clay was heated in a muffle furnace, which raised the temperature gradually over the course of two hours at a rate of 60°C/min from room temperature to 750°C. Due to the presence of iron oxide in the clay, this heating produced a discernible color shift from gray to light red. The faint red coloring was caused by the magnetite phase changing

into hematite after thermal treatment and without oxygen. Magnetite may change back to hematite during cooling if oxygen is present, giving the calcined material a reddish tint. On the other hand, magnetite will continue to be the stable phase in the absence of oxygen [29]. A small laboratory ball mill that could grind five kilograms of calcined clay was then used to finely grind the material. The ground calcined clay's particle size distribution is shown in Figure 2-a; roughly 10% of the material was left as residue after sifting through a 45-micron mesh.

Table 1 : XRF of raw materials used in this work

Oxides	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	SO ₃	Na ₂ O	K ₂ O	TiO ₂	P ₂ O ₅	CL	LOI	Sum
Clinker	21.03	5.83	4.57	65.9	1.17	0.72	0.22	0.19	0.09	0.10	0.02	0.08	99.92
Gypsum	0.12	0.15	0.04	31.20	2.05	43.70	0.12	0.10	0.00	0.00	0.06	22.00	99.54
gray Clay	53.38	30.80	3.74	0.02	0.15	0.12	0.13	0.11	1.84	0.04	0.03	9.58	99.94
Calcined clay	55.65	34.20	5.24	0.23	0.01	0.15	0.15	0.13	2.45	0.11	0.07	1.50	99.92
Limestone	0.38	0.08	0.02	54.40	0.96	0.07	0.21	0.25	0.00	0.00	0.02	43.60	99.99
Sand	96.33	1.20	0.53	0.09	0.10	0.05	0.12	0.51	0.00	0.00	0.01	0.95	99.89
Whit cement	20.65	4.95	0.43	64.88	1.08	2.45	0.10	0.56	0.42	0.02	0.04	4.20	99.75



Figure 1: Calcination processes of gray clay.

2.1.B Mineralogical and microstructure characterization.

The mineralogical composition of calcined clay and gray clay was examined using Fourier-transform infrared spectroscopy (FTIR) and X-ray diffraction (XRD). Gray clay displays sharp peaks corresponding to two crystalline phases: quartz and kaolinite. The disappearance of Kaolinite Peaks from the calcined clay pattern confirms the completion of the thermal treatment of kaolin. This indicates that the kaolinite has undergone a thermal transformation when heated by losing its structural water. The corresponding peaks of quartz are still visible and sharp in the calcined clay pattern [30, 31]. This indicates that quartz is a thermally stable crystalline phase and remains unchanged after the calcination process. Additionally, a broad, low-intensity hump observed in the calcined clay pattern confirms the presence of the amorphous metakaolin phase identified as unhydrated kaolinite.

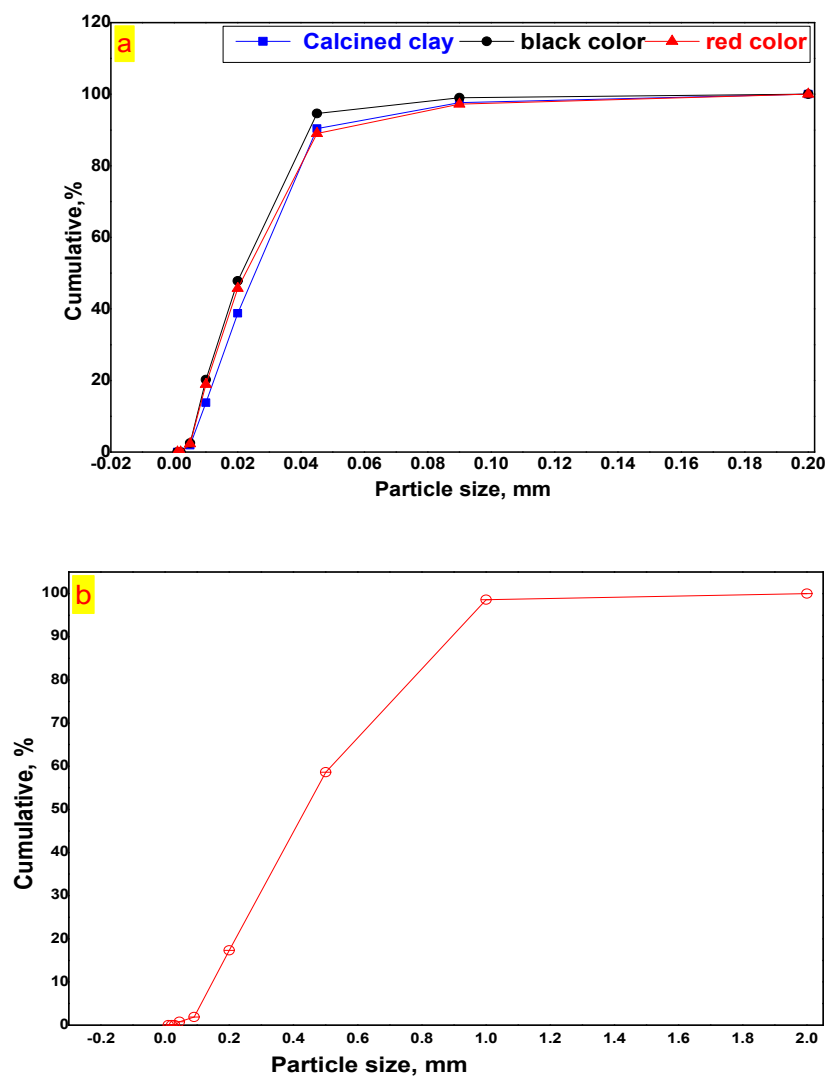


Figure 2: Particle size distribution of: a) initial raw materials, and b) sand.

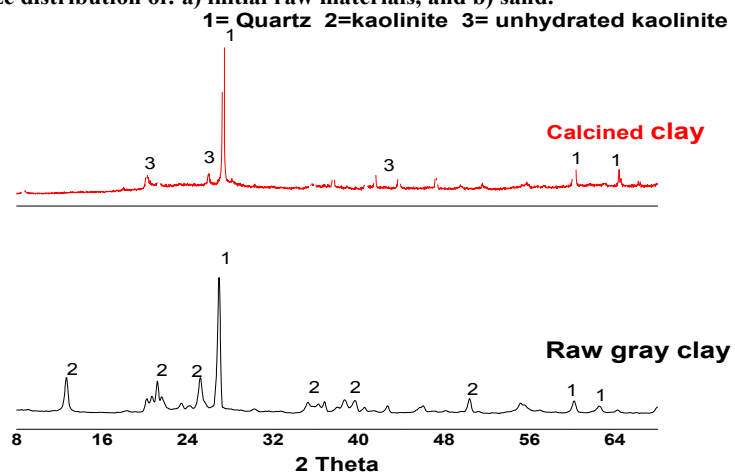


Figure 3: XRD patterns of raw gray clay and calcined clay.

Figure 4 shows gray clay spectra both before and after calcination. In gray clay, stretching band (at 3683 cm^{-1}) corresponding to inner hydroxyl groups and OH bending band (at 915 cm^{-1}) corresponding to the Al-OH, are displayed [30, 32]. The disappearance of these peaks in calcined clay indicates effective completion of the dehydroxylation process. There are also noticeable strong peaks at 1003 cm^{-1} and 1063 cm^{-1} , which represent the Si-O bond stretching vibrations. In calcined clay, the Si-O stretching vibrations become less defined and may merge into a broad band, indicating an amorphous structure formation. Furthermore, the absorption peaks in calcined clay and kaolinite clay at 2354 cm^{-1} and 2162 cm^{-1} are caused by CO_2 vibrations adsorbed on the clay particles' surface [33]. Lastly, Si-O Al VI bending vibrations and Si-O bending vibrations are linked to the bands at 532 cm^{-1} and 430 cm^{-1} , respectively.

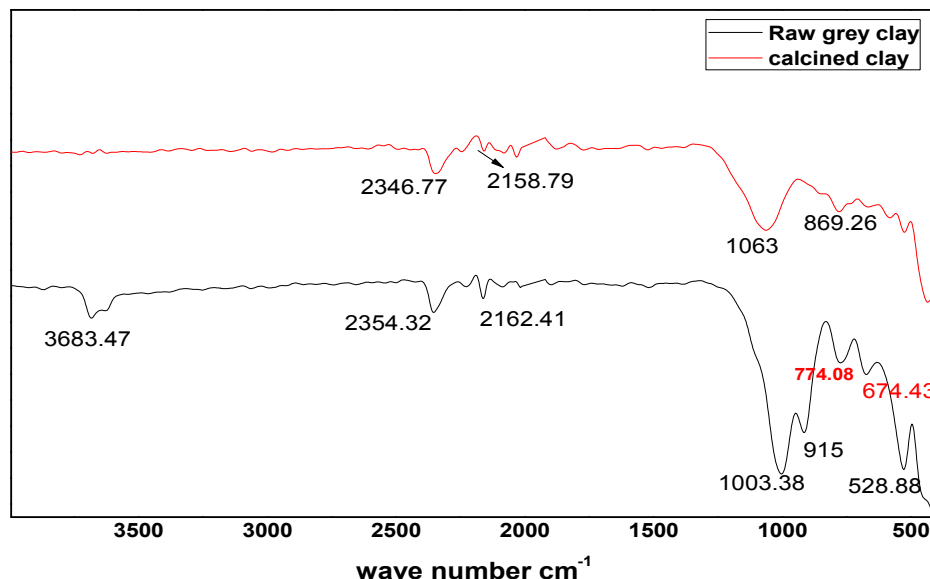


Figure 4: FTIR spectra of gray clay and calcined clay.

2.1.2 Preparation of limestone calcined clay cements (LC3) and render mortars:

Three distinct systems of limestone calcined cement mortars, LC3(A), LC3(B), and LC3(C) with varying amounts of clinker (CK), calcined clay (CCL), limestone (LS), and 5 % gypsum, are prepared as shown in Table 2. Additionally, a reference sample of ordinary Portland cement was made using a constant cement/sand ratio of 1:3 in accordance with EN196-1. Table 3 displayed the specific surface area or fineness for several cement systems. Moreover, Table 4 displays various render mortar formulations, in addition to the blank sample (white cement), with a surface area of $5200\text{ cm}^2/\text{g}$, compressive strength of 48.6 MPa at 28 days, and a whiteness degree of about 93%. The prepared render mortar utilized two types of inorganic pigments: black (B) and red (R). The widespread use of white cement for decorative applications is well-documented [34]. Yet, its production is associated with considerable expense and a high environmental impact, largely due to CO_2 emissions. Consequently, this study explores a replacement material that offers a lower-cost limestone calcined clay cement (LC3), which has a good environmental impact and reduces carbon footprint.

Table 2: Mix design of preparation of limestone calcined cement mortars.

preparation of LC3 binder, %					preparation of LC3 mortar, gm						
Sample No	CK	GY	CCL	LS	CK	GY	CCL	LS	Sand	water	C/S
OPC	95.00	5.00	0.00	0.00	427.50	22.50	0.00	0.00	1350.00	225.00	0.30
LC3(A)	60.00	5.00	25.00	10.00	270.00	22.50	112.50	45.00	1350.00	237.00	0.30
LC3(B)	55.00	5.00	25.00	15.00	247.50	22.50	112.50	67.50	1350.00	240.00	0.30
LC3(C)	50.00	5.00	30.00	15.00	225.00	22.50	135.00	67.50	1350.00	242.00	0.30

Table 3: Fineness of LC3 and OPC by EN 196-6

Samples. NO	Blaine, cm ² /g	Sieve Residue	
		90 μ m	45 μ m
Blank OPC	3750	0.00	8.00
LC3A	5850	1.50	14.50
LC3B	6100	1.00	13.60
LC3C	6250	1.00	12.00

Table 4: Mix design of preparation of render mortars.

Sample No	white OPC%	LC3(C)%	Sand%	LS%	In. pig %	HPMC%
Blank(B)	30.00	--	52.90	15.00	2.00	0.10
Blank(R)	30.00	--	52.90	15.00	2.00	0.10
R25B	---	25.00	58.90	15.00	1.00	0.10
R30B	---	30.00	53.40	15.00	1.50	0.10
R35B	---	35.00	47.90	15.00	2.00	0.10
R25R	---	25.00	58.90	15.00	1.00	0.10
R30R	---	30.00	53.40	15.00	1.50	0.10
R35R	---	35.00	47.90	15.00	2.00	0.10

2.2 Test methods

2.2.1 Mixing and curing process

The mortars were made using the ratios mentioned above in tables (2) and (3). The amount of water used was assessed to provide sufficient consistency and workability; after 15 jolts, the flow value on the flow table was roughly 150–185 mm. For LC3 mortars, the corresponding EN 459-2:2010 water quantity was 240 ± 2 . The mortar was made using a constant ratio of 450:1350 in accordance with the process described in EN 196-1. Water was added during the first few seconds of mixing, followed by 150 seconds of mechanical mixing, 30 seconds of scraping the boundaries, and more mixing. Mortars were then produced in 40 x 40 x 160 mm metallic prismatic molds. The samples of mortar were removed from the molds and cured for 28 and 90 days at a temperature of 20 ± 3 °C and a relative humidity of $90 \pm 5\%$. Finally, samples were kept in a climatic room for 24 hours at 20 ± 3 °C and $65 \pm 5\%$ RH the day before testing began.

2.2.2 Evaluation of the consistency of fresh mortar (by flow table) EN 1015-3

The average diameter of a test sample is measured to calculate the flow value. To perform the test, first prepare a mold (60 mm tall, 100 mm base, 70 mm top) and place it in the middle of a flow table. The mold is filled in two layers, each tamped 10 times. After leveling the top and cleaning the base, the mold is removed 15 seconds later. The table is then jolted 15 times in 15 seconds. Finally, the spread diameter of the mortar is measured with calipers in two perpendicular directions and recorded. The mean of these two measurements is the flow value.

2.2.3 Determination of bulk density

A minimum of three identical cubes with the same age and mix composition were used for each measurement before assessing the compressive strength. Both water and air (saturated surface dry) were used to weigh the suspended samples. The Archimedes method was used to compute the density using the following formulas:

$$\text{Bulk density} = \frac{\text{Saturated weight}}{\text{Volume of sample}} \quad (\text{g/cm}^3)$$

$$\text{Volume sample} = \frac{\text{Saturated weight-suspended weight}}{\text{Density of water}} \quad (\text{cm}^3)$$

$$\text{Bulk density} = \frac{\text{Saturated weight}}{\text{Saturated weight-suspended weight}} \quad (\text{g/cm}^3)$$

2.2.4 Determination of compressive strength and flexural strength

Compressive and flexural strengths were measured using prismatic test specimens measuring 40 x 40 x 160 mm in accordance with EN196-1. A compressive strength test was performed on each half after the specimen was split in two for the flexural test. The Automatic Compression and Bend test plant, Toni PRAX testing machines, were used to perform the compressive and flexural strength tests. Data was collected using a computer that was linked to a Zwick test Xpert system.

2.2.5 Thermal conductivity test

The thermal conductivity, or k-value, determines how much energy is needed to heat and cool structures. In solids, conduction heat transfer is accomplished by free electron energy transport and molecular vibrations. A laser comp heat flow meter for 314 instruments with S/N: 11061327 was used in the thermal lab at the Housing and Building National Research Center (HBRC) to measure the thermal conductivity of the mortars used in this study. ASTM C518 is a standard test procedure that uses a heat flow meter instrument to determine the steady-state thermal transmission qualities of materials. This technique evaluates the thermal resistance and conductivity of materials such as building supplies and insulation. The test involves placing a specimen of a specific size (30 x 30 cm x 1 cm) between two plates—one of which is heated and the other cooled—and measuring the heat flow through it. To provide a controlled temperature differential (temperature gradient) throughout the specimen, the plates are kept at varying temperatures. The mean temperature difference across the specimen was 32.5 to 45 °C. For five hours, under test conditions of 25°C and 55% relative humidity, the heat flux direction through the sample was upward from the hot plate to the cold plate.

2.2.6 Adherence to the substrate (pull-off test)

A render's ability to tolerate normal and tangential stresses at the interface with the substrate, even after the hardening process, is known as adherence. A digital or manual pull-off adhesion testing machine with a dolly (type Proceq DY-216 Elcometer 506 (UK)) is usually used for the test. It is crucial to clean the coated surface and the dolly before starting the test. Then, after 28 days, the dolly is affixed to the covered surface using the prepared adhesive. The adhesive should be left to cure at room temperature. After the curing process is finished, the actuator of the machine is placed over the dolly, and pressure is exerted until the bond breaks. A hydraulic pump can be used to create the pressure.

3. Result and discussion

3.1 Limestone Calcined Clay Cement (LC3)

Limestone Calcined Clay Cement (LC3) is a newly proposed low-carbon cement, which can effectively reduce energy consumption and carbon emissions of the traditional cement industry without changing the basic mechanical properties of cement-based materials [1]. In this study, the mechanical and physical properties are determined.

3.1.1 Setting time and water requirement

The initial and final setting times of the prepared cement pastes, as well as the water of standard consistency, are listed in Table 5. The findings demonstrated that the LC3 standard consistency (%) is higher than that of the OPC. This is because calcined clay is mainly composed of amorphous metakaolin, which permits water to penetrate and dissolve. Moreover, amorphous phases offer a high surface area for water absorption [12]. Furthermore, mixes with a higher clay percentage also require more water. Although the limestone addition might help with some water demand and improve flowability, superplasticizers are nearly always needed in practice to maintain the appropriate workability [35]. The presence of limestone in the binder causes a significant reactivity with the calcium aluminates present in both the calcined clays and clinker, as well as with portlandite to form aluminate hydrate phases during the early stage. The results demonstrate that the presence of both limestone and calcined clay reduces the initial and final setting times [36]. The results also indicate that calcined clays and limestone reduced setting time. A high rate of hydration in the presence of reactive metakaolin in calcined clay may have contributed to the shorter setting time by accelerating the nucleation of calcium silicate hydrates (C-S-H) that resulted in a build-up of bridges between cement particles [37]. Moreover, the addition of Gypsum to Clinker enables calcium aluminate phases to react and produce calcium sulfoaluminate hydrates, which postpone setting [38].

Table 5: Water consistency and setting time of different mixes of the LC3 system

Mix.NO	CK	CCL	LS	GY	W/C	Setting time, (min)	
						I.T	F.T
OPC	95.00	0.00	0.00	5.00	26.00	168.00	250.00
LC3A	60.00	25.00	10.00	5.00	29.80	150.00	220.00
LC3B	55.00	25.00	15.00	5.00	31.00	140.00	205.00
LC3C	50.00	30.00	15.00	5.00	31.50	135.00	195.00

3.1.2 Mechanical properties of LC3 mortars

The compressive and flexural strengths, shown graphically in Figs. 5a–b, indicate that increasing curing time up to 90 days boosts both strengths for all LC3 mortar mixes (Blank, LC3A, LC3B, and LC3C) because of the ongoing hydration process. Additionally, alumina and limestone may react to form carbo-aluminate hydrates. These products occupy space and contribute to improving durability and strength [39]. Since the pozzolanic reaction is just beginning and clinker content is significantly lower, LC3's compressive and flexural strengths are also less than those of the blank OPC. The lowest strength of LC3 is linked to the low workability of the cement mixture. Poor compaction of the fresh mortar increases pore volume in the hardened mortar, leading to lower strength [40, 41]. The high pozzolanic reactivity of the calcined clay content results in reduced dissolved portlandite in the liquid phase, which inhibits full hydration of metakaolin phases and thus limits ultimate strength [42]. Segregation of aggregate in the cement mortar, caused by excess water beyond that needed for hydration, is another possible reason for the low development of both compressive and flexural strength [43, 44]. All these findings suggest that although adding limestone and calcined clay may weaken the mortar's early-age strength, they do not negatively impact its long-term compressive and flexural strengths [45].

3.1.3 XRD analysis

The XRD pattern for the LC3(C) mix at 28 and 90 days of curing is shown in Figure 6. Ettringite ($C_6AS_3H_{32}$, d values 4.25, 2.13 $\text{\AA}$), calcium silicate hydrate (CSH, d values 2.28, 1.97, 1.67 $\text{\AA}$), portlandite (CH, d values 2.46, 1.82 $\text{\AA}$), quartz Q, d values 3.34, 1.54 $\text{\AA}$, and calcite from carbonation, d values 3.03, 1.65 $\text{\AA}$, are found to be the primary phases of the hydrated mortars. Due to improved pozzolanic reactions, peak intensities rise with curing time up to 90 days [46]. On the other hand, the pozzolanic reaction also causes the CH peak's intensity to decrease over time. Additionally, reduced amounts of dissolved portlandite are caused by the presence of calcined clay (CCL) and the high pozzolanic reactivity that goes along with it [41]. Ettringite's growth peaks over time as a result of its gradual conversion to monosulphate. Ettringite is stabilized when monosulphate interacts with CO_3^{2-} ions to generate carboaluminate and ettringite [47, 48]

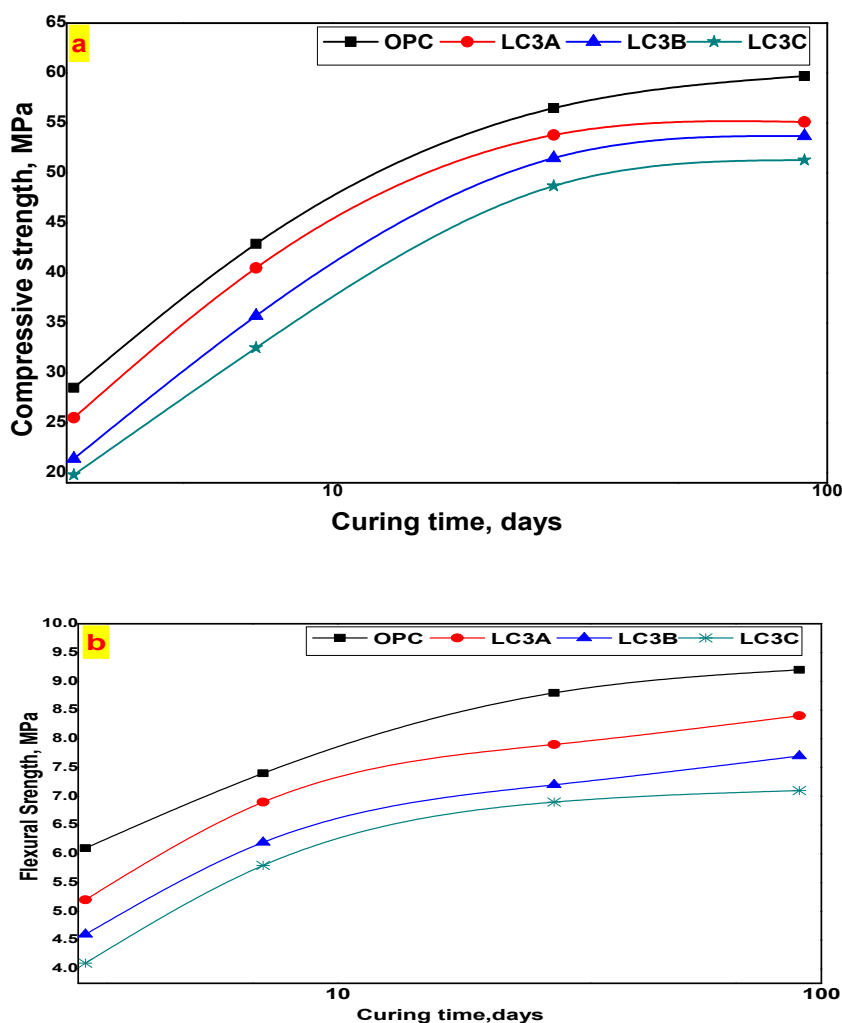


Figure 5: The mechanical characteristics of different mortar mixes (LC3 and blank OPC) after 90 days of curing, a) compressive strength, b) flexural strength.

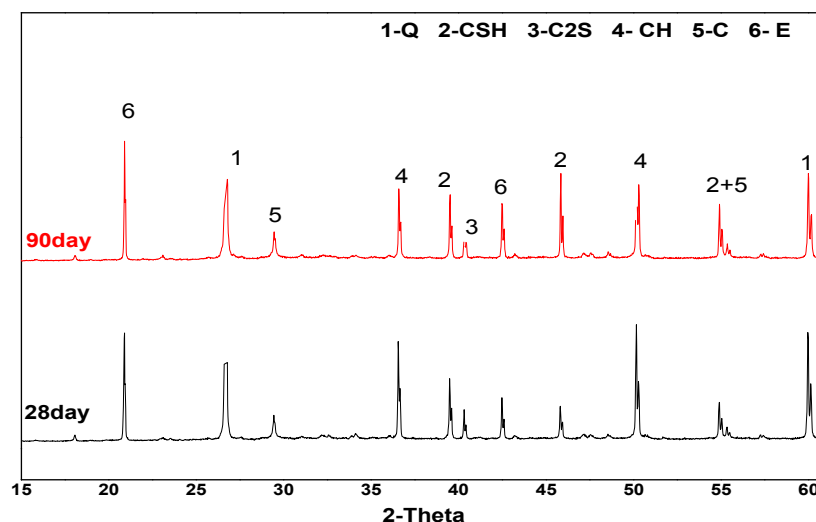


Figure 6: The XRD patterns of hydrate cement mortars for LC3C at 28 and 90 days.

3.2 Characterization of Render based on LC3 as binder:

LC3 cement is a promising alternative to traditional Portland cement and appears to be a more cost-effective option. Preliminary studies indicate that it is cheaper to manufacture than plain Portland cement and may also be less expensive than various blends of supplementary cementitious materials (SCMs), depending on local availability and pricing. The lower cost of LC3 is a significant advantage that could increase its adoption as a general-purpose cement. This study specifically evaluated the use of LC3 in render mortars, where its cost-effectiveness was a key benefit. The research assessed the physical and mechanical properties of these mortars, demonstrating the viability of LC3 as a binder.

3.2.1 Workability, Water demand, Setting time, and flow value:

The water needed, setting time, and flow rate of prepared render mortars were as indicated in Table 6. It has been noted that when the LC3 level increases, the setting time consequently decreases while the water binder ratio increases. Furthermore, according to EN1015-3, the workability decreases as LC3 content increases, resulting in low flow values that mix R35B and R35R show the lowest flow rate. Because of the decreased workability, more water is needed to keep the mortar's typical consistency. On the other hand, a higher water /cement ratio may lead to an excessively wet render and a deficient curing with implications on the cohesion of the mortar [49]. The findings also indicate that both setting time and flow value decrease as the pigment /cement (P/C) ratio increases. Red pigment, a byproduct of iron recycling, exhibits lower fluidity due to its tendency to absorb water, which is strongly influenced by the size and shape of its particles [22]. In contrast, when black pigment is used, the R25B mix demonstrates higher fluidity compared to R25R. This is because black pigments as ferric oxide, are generally spheroidal in shape and consist of a blend of fine powder and carbon black, which enhances overall black chromaticity. Moreover, larger particle sizes increase water demand, leading to improved fluidity. [22].

Table 6: The water demand, setting time, and flow rate of render

Mix. NO	W/S %	Setting time(min)		Flow value (mm)
		I.T	F.T	
Blank (B)	26.30	350.00	540.00	167.00
Blank (R)	25.90	340.00	525.00	165.00
R25B	22.50	390.00	550.00	179.00
R30B	23.60	340.00	480.00	172.00
R35B	24.40	290.00	410.00	166.00
R25R	22.20	360.00	530.00	177.00
R30R	23.20	310.00	420.00	170.00
R35R	23.90	270.00	380.00	164.00

3.2.2 Mechanical properties of the produced render:

The prepared render's flexural and compressive strengths were graphically represented in Figures 7- a and –b. Due to the progressive formation of hydration products, such as calcium carboaluminate, CAH, and CSH, which densify in the pores, the results demonstrated that both compressive strength and flexural strength increased as the curing period increased up to 28 days. Theoretically, a mortar's strength increases as the amount of binder it contains increases. Alternatively, too much binder causes the mortar to shrink and break. The mortar must have a low modulus of elasticity and a high tensile strength to better withstand the stress placed on it and prevent cracking [50] and [51]. All the mixes are confirmed to be appropriate for render mortar and meet EN998-2 and EN 1015-11 criteria. Additionally, hydroxypropyl methyl cellulose prevents water from escaping; it has been confirmed to increase mortar cohesiveness [52].

According to the results, both compressive and flexural strength rise when inorganic pigment (IOP) increases. This is explained by the fact that inorganic pigment can adsorb a lot of available water, which lowers the mortar's actual water-to-cement ratio [53]. In addition, they have a micro-aggregate filling action that can increase the density of mortars by filling their internal pores [54]. Our findings indicate that black pigment has better mechanical properties than red pigment. This could be because black pigment has a large specific surface area, which provides nucleation sites that speed up cement hydration and improve overall hydration, thereby enhancing the overall hydration process and improving compressive strength [55]. These results agree with other research, which established that less than 6% of the weight of the cement to be used was the ideal pigment mix ratio for assuring the characteristics of colored mortar [53].

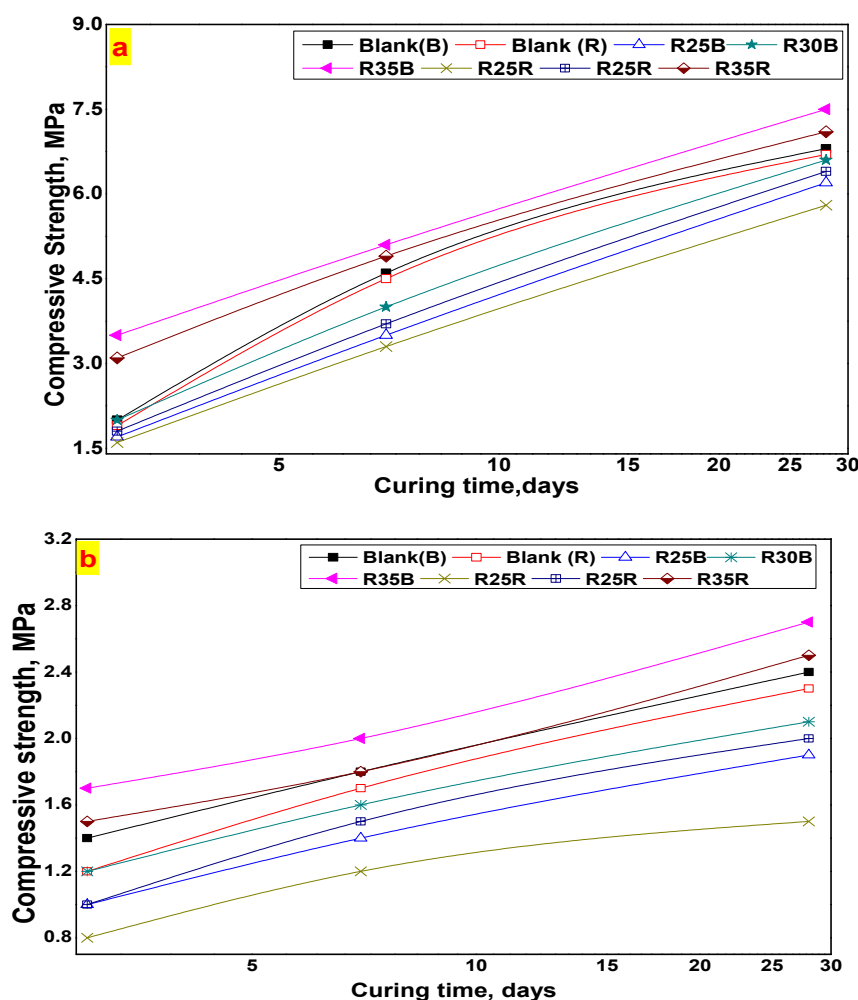


Figure 7: The mechanical characteristics of different colored render mortar mixes (LC3 and blank OPC) after 28 days, a) Compressive strength, b-) Flexural strength.

3.2.3 Comparison of several attributes between conventional cement render mortar (blank) and environmentally friendly LC3 colored render mortar (R25B)

Performance, cost, and environmental impact must all be carefully considered when choosing colored rendering mortars. In the second phase of this study, the proposed mixes were compared with a blank cement render mortar in terms of microstructural characteristics, porosity, bulk density, mechanical adhesion, and thermal conductivity. The results indicate

that, although all colored mixes exhibited mechanical performance comparable to the blank, they were generally more cost-effective and associated with lower CO₂ emissions. Among the investigated blends, R25B emerged as the most suitable option, combining adequate mechanical strength, a lower carbon footprint. A low binder content in R25B plays a crucial role in reducing carbon emissions and lowering costs compared to both the blank and other blends. Furthermore, it permits the incorporation of additional inorganic black pigment to achieve the desired decorative color, while maintaining the pigment-to-cement (P/C) ratio below the 10% limit specified in EN 13914-1 (2005). This allows up to ~2.5% pigment addition without compromising performance [56]. Conversely, the use of red pigment with limestone-calcined clay cement proved less efficient. Due to the natural gray shade of the binder, red pigmentation increased the P/C ratio above 10%, offering no economic advantage. For instance, achieving a dense red tone in R25 required ~5% red pigment, which increased the P/C ratio by approximately 20%. Even at 2% dosage, the resulting mortar showed only a light-red coloration with negligible aesthetic benefit. Such outcomes negatively affect both cost and mechanical performance. Overall, darker-toned colors, especially near-gray shades, are best suited for limestone-calcined clay cement, balancing aesthetics, cost, and sustainability. The R25B mix was identified as the optimal choice, combining low cost, reduced CO₂ emissions, and higher pigment capacity. Except for red, pigments generally improved mortar performance, with black pigment notably enhancing strength and durability [57-59].

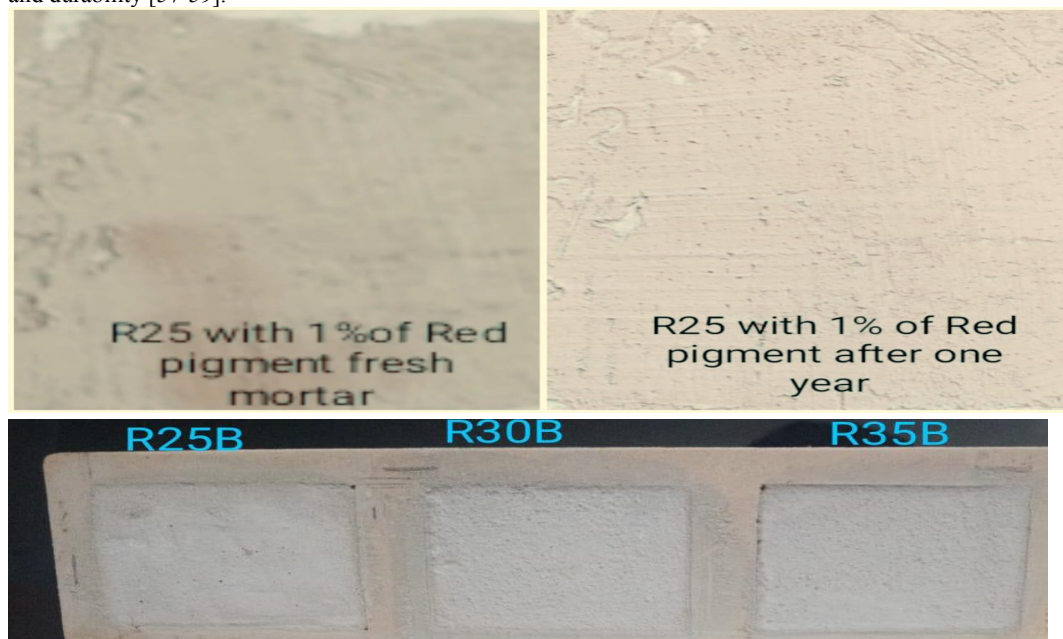


Figure 8: The visual appearance of colored render mortars with black and red pigments on the substrate.

3.2.4 XRD of hardened render mortars:

The XRD of the chosen render mortar mix based on LC3 binder (R25B) is shown in Figure 9. The primary phases that have been recognized are Ettringite (E, C₂ASH₃2, d values 3.39, 2.75 Å), calcium silicate hydrate (CSH, d values 3.05, 2.56, 2.09 Å⁰), portlandite (CH, d values 4.09, 2.19 Å⁰), quartz (Q, d values 3.21, 1.76 Å⁰), and calcite from carbonation with d values 4.33, 2.19 Å⁰. The hydration of cementitious systems involves the formation of ettringite, calcium silicate hydrate (CSH), portlandite, quartz, and calcite. As hydration progresses, the intensity of original cement phases (C₃S, C₂S) decreases, while portlandite reacts with calcined clay in a pozzolanic reaction to form more CSH. Calcium carbonate reacts with C₃A to form calcium carboaluminate, and calcined clay can further react with limestone to produce mono- and hemi-carboaluminate, refining the pore structure. Over time, ettringite decreases as monosulphate forms, which can then combine with carbonate ions to stabilize ettringite and carboaluminate [47, 48].

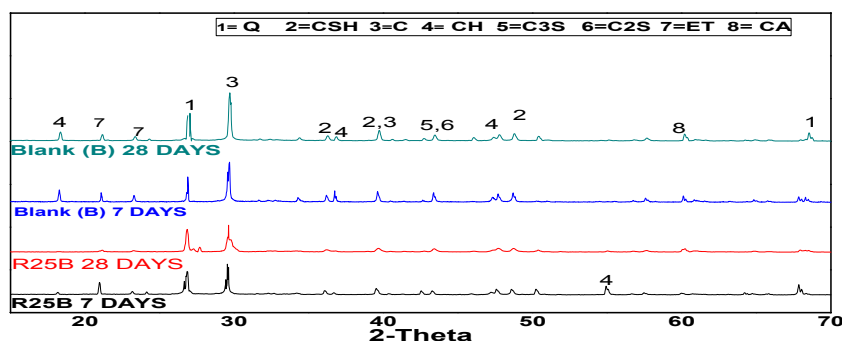


Figure 9: XRD patterns of render based on LC3 and blank sample at 7, 28 days.

3.2.5 Fourier transform infrared spectroscopy (FTIR):

The FTIR data, illustrated in Figure 10, were collected for R25B and the blank samples examined at 7 and 28 days of curing. The absorption bands have been annotated to correspond with the detected components. An asymmetric stretching vibration related to carbonates, denoted as $\nu(\text{C-O})$, was observed at approximately 1419 cm^{-1} . Additionally, the out-of-plane bending vibrations associated with carbonates were detected at 870 cm^{-1} and 712 cm^{-1} for all specimens. The intensity of these carbonate bands in R25B decreased with curing time due to the consumption of limestone in the pozzolanic reaction that produces carbo-aluminates. The peak at 1419 cm^{-1} may be attributed to calcium carboaluminate (CC) or other CO_3^{2-} species. The stretching vibration band for sulfates, $\nu(\text{S-O})$, appeared at 1062 cm^{-1} and diminished in R25B, likely due to sulfates reacting more strongly with the available aluminates from CCL. The S-O stretching band, typically found at 1062 cm^{-1} in ettringite or mono-sulfoaluminate, also decreased as the hydration process advanced. For the 7-day cured specimens, the $\nu(\text{Si-O})$ absorbance associated with calcium silicate hydrate (C-S-H) was observed at around 871 cm^{-1} . After 28 days of curing, this band shifted to 969 cm^{-1} . The movement of the $\nu(\text{Si-O})$ band to a higher wavenumber indicates the transformation of dimeric Si-O bonds into three-dimensional (3D) polymeric Si-O-Si units during polymerization in C-S-H [60], which correlates with improved strength in the reference samples.

In R25B, a doublet centered around 780 cm^{-1} (with peaks at 780 and 868 cm^{-1}) was noted at both 7 and 28 days. This was attributed to an increase in C-S-H resulting from the progression of the pozzolanic reaction, along with quartz impurities (Si-O vibrations) present in the CCL, which were also evident in the original materials. As the curing period lengthened, the $\nu(\text{Si-O})$ band shifted towards higher wavenumbers, ultimately reaching 868 cm^{-1} for specimens cured for 28 days. This transition indicates that the C-S-H had fully transitioned to a 3D polymerized Si-O structure [61].

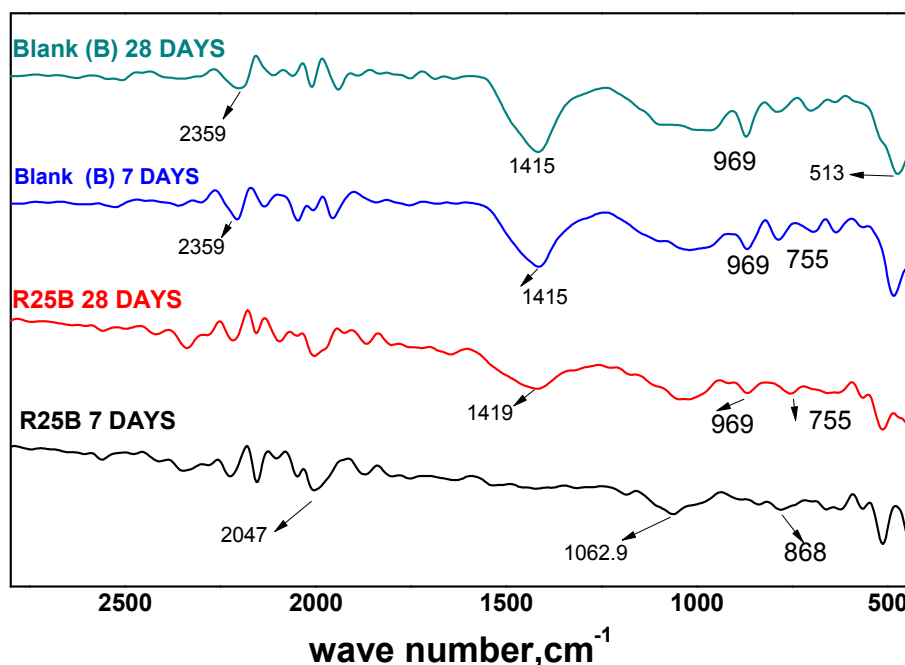


Figure 10: FTIR patterns of render mortars based on LC3 (R25B) and blank sample after 7 and 28 days of curing.

3.2.6 Bulk density of hardened render mortars

Table 7 shows the bulk density obtained from a selection of hardened render mortars based on LC3 binder (R25B) with blank dry mix. Render mortar's bulk density is determined by the degree of hydration. The hydration products fill a portion of the pore volume as the hydration process goes on because their volume is more than double that of the unhydrated cement; this raises the bulk density of hardened mortar. According to the data, bulk density increases up to 28 days after curing. This could be because the production of new hydration products fills some pores. The bulk density of R25B is lower than of blank, as it has less hydration products such as CSH, CAH, and more carboaluminate content with low density [62]. Moreover, the rate of hydration of R25B is lower than that of blank so the values of bulk density of R25B are lower than those of blank, at all curing times. Additionally, the density of the CSH produced by the pozzolana reaction is lower than that of the CSH produced by hydration of the blank. Consequently, the bulk density of render mortar R25B is reduced [62].

Table 7: The bulk density of hardened render mortars (R25B) after 3, 7, and 28 days of curing.

Sample No	Bulk density g/cm ³		
	3d	7d	28d
Blank	2.485	2.527	2.586
R25B	2.238	2.247	2.251

3.2.7 Porosity and water absorption of render mortars

Table 8 presents the porosity and water absorption of hardened render mortars made with LC3 binder (R25B) and a blank OPC. It was observed that the porosity of hardened mortars decreases with curing time for both mixes. As hydration progresses, the hydration products occupy a portion of the pore volume, leading to a reduction in the porosity and water absorption of render mortars over time. Furthermore, new C-S-H gel fills the capillary voids formed at early ages, depending on the degree of hydration and the quantity of new solid products that occupy the original pores [63]. The results also indicate that R25B exhibits greater porosity than the reference sample, likely due to the higher sand content, which can contribute to a more porous structure. The presence of pores in the samples may result from voids not filled by the solid paste compounds, with irregular or spherical shapes attributed to entrained air trapped during mixing [64], as confirmed by the results in section (3.2.2). Additionally, the porosity of R25B is higher than that of the blank sample, which may be due to the lower loose bulk density of expanded aggregates [65]. In contrast, the findings indicate that the reference sample has higher water absorption values than the R25B sample; this could be attributed to the production of more hydration products that facilitate the absorption process. Moreover, the increase in inorganic pigment in the blank sample may have led to greater water absorption. Higher concentrations of pigments can result in significant changes in the mortar's microstructure, often leading to increased water absorption. The pore ratio decreased, resulting in lower water absorption values when black pigment was used [57]. In render mortars, a higher cement-to-sand ratio typically results in lower porosity. This is because a higher cement content creates a denser matrix with fewer voids after hardening. Conversely, a higher sand content increases the overall volume of the mortar, potentially creating more space for pores, thereby increasing porosity [66].

Table 8: Porosity and water absorption of colored render mortar mixes R25 and blank sample.

Sample.NO	Porosity, %			Water absorption, %		
	3d	7d	28d	3d	7d	28d
Blank	4.446	2.631	2.325	19.689	18.984	17.887
R25B	6.869	4.070	3.510	14.781	14.179	13.652

3.2.8 Adherence to the Substrate (pull-off) and thermal conductivity.

The property known as adherence enables render to adapt to typical stresses at the substrate interface even after the hardening process is complete. Table 10 presents the mechanical adherence of the render based on the suction of the substrate. Since a higher binder concentration in mortars causes more cracking, which may result in a loss of adhesion, the results indicate that the pull off render mortars based on LC3 (R25B) are more than blank [56]. In contrast to blank, which has poor porosity and an excessive absorption capacity that tends to quickly desiccate coating, halting hydration of its contents and resulting in lower adherence, R25B has higher porosity, which results in superior absorbent substrates. Higher water-absorbing mortars lessen the adherence of renders [67]. The thermal conductivity values are shown in Table 9. Since heat conduction in materials is linked to the combination of vibrational vibrations and electron movement, it is directly tied to the microstructural and compositional properties of the composite. The findings indicate that the R25B sample has a lower thermal conductivity than the reference sample; this could be because of its larger porosity, which makes it lighter and has a higher pore volume fraction. Better compactness and reduced porosity of the specimen are also indicated by a higher conductivity coefficient, although the thermal insulation effect is diminished [23]. The low conductivity of porous materials makes them ideal for thermal insulation in terms of building thermal performance. The type of material used in a building can have a big impact on its thermal performance. The low conductivity of porous materials makes them more suitable for thermal insulation [68].

Table 9: The pull-off and thermal conductivity of the colored render mortar R25B and blank OPC.

Sample No	Pull off N/mm ²	Thermal conductivity W/m.k
Blank	0.26	0.64
R25B	0.60	0.49

4. Conclusions:

This study demonstrated that limestone calcined clay cement (LC3), particularly the LC3(C) system with a 50% clinker factor, offers a sustainable, low-energy binder with a significantly reduced carbon footprint compared to ordinary Portland cement (OPC). Although its compressive strength is slightly lower at early ages, the long-term performance remains acceptable due to the pozzolanic activity of calcined clay and the synergistic reactions with limestone.

The results reveal several important conclusions:

- While compressive strength is lower than that of OPC at early ages, long-term performance is acceptable due to pozzolanic activity.
- LC3 mortars exhibit higher water demand and shorter setting times than OPC.
- R25B render mortar is identified as the best mix, offering compliance with EN998 standards, lower costs compared to OPC mortars, and a reduced CO₂ footprint and environmental impact.
- Compared to red pigment, black pigment was found to be more compatible with LC3, providing higher fluidity, better mechanical properties, and a shade that is closer to the binder's natural gray.

The findings indicate that LC3 is a viable alternative to Ordinary Portland Cement (OPC) for both structural and decorative render mortars. Utilizing LC3 can help the construction industry shift towards more sustainable practices by decreasing energy consumption and reducing CO₂ emissions while still meeting performance standards. Its cost-effectiveness and suitability for areas with abundant clay and limestone, like Egypt, further support its large-scale use. Moreover, LC3-based mortars offer comparable or even superior durability against environmental stressors like chloride ingress and sulfate attack, making them ideal for coastal regions and aggressive soil conditions. This extends building lifespan, reduces frequent repairs, and promotes local construction materials development, reducing reliance on imported cement. Continued research is expected to lead to even more advancements in sustainable building materials, ultimately promoting an eco-friendly construction industry.

5. Conflicts of interest

There are no conflicts to declare.

6. Formatting of funding sources

Not applicable

7. References

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