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# **RESEARCH PAPER**

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# Building a Sustainable Future: Innovative Application of China Clay from Kerala in LC3

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# Abstract

Cement manufacturing has significant adverse environmental impacts. Clinker production by heating a mixture of limestone, clay, and other minerals at a high temperature in a kiln releases a large amount of carbon dioxide ( $CO_2$ ) into the atmosphere, contributing significantly to global greenhouse gas emissions and climate change. There is a need for more sustainable and environmentally friendly alternatives to traditional cement. In the scenario where even geopolymer failed to be a sustainable successor of conventional cement, LC<sub>3</sub> is a potential solution. LC3 stands for Limestone Calcined Clay Cement, a blend of limestone, calcined clay, and a small amount of gypsum. The selection of clay is of utmost importance in creating the best outcome for LC3. The properties and performance of the resulting cement can vary depending on the specific type of clay used. Metakaolin is the supplementary cementitious material that contributes to the properties of LC3. Metakaolin is known for its high pozzolanic activity to form additional cementitious compounds. Two clays, namely Hindustan special China clay(C1) and Indian Clay(C2) collected from different parts of Kerala, were subjected to the EDAX test for chemical comparison with pure kaolin. By analysing the weight change over time, TGA provided information about the composition of the kaolinite present in the clay samples. C1 and C2 were activated by heating them in a muffle furnace at varying temperatures of 650–750 °C and holding durations of 30-90 minutes. The changes in the crystalline structure of these materials were evaluated using X-Ray Diffraction (XRD). This study aims to investigate the feasibility of utilising locally soured clay in Kerala for the production of LC<sub>3</sub> (Limestone Calcined Clay Cement).

**Keywords:** Metakaolin; EDAX test; X-Ray Diffraction (XRD); Kaolinite; Limestone Calcined Clay Cement; TGA (Thermogravimetric analysis)

# Introduction

Ordinary Portland Cement (OPC) is one of the most widely used types of cement in the construction industry. It is known for its versatility and durability, making it suitable for a wide range of applications in building and infrastructure projects. OPC is manufactured by grinding clinker, gypsum, and other additives, providing excellent compressive strength and stability in concrete structures. The production of Portland cement requires a great deal of energy and emits a significant amount of CO<sub>2</sub>, the process of chemical decomposition of components used in cement manufacturing also contributes to CO<sub>2</sub> emission (Sharma et al., 2021) and other greenhouse gases that contribute to global warming. During these years, cement manufacturing underwent several changes. While the initial set of innovations aimed to lower the energy needed to produce clinker, the majority of contemporary clinker kilns today are close to achieving the highest thermodynamically feasible efficiency. Different clinker compositions were introduced to address this problem. Geopolymer is thought to be a promising alternative to cement. However, geopolymer was first discovered 45 years ago. Geopolymer is not widely used as a cement alternative. In this case, replacing all the cement is not the best course of action; only a portion of the cement needs to be changed. By reducing the quantity of clinker used, this approach can effectively lower CO<sub>2</sub> emissions during the manufacturing process of cement. The significance of Imestone-calcined clay cement is now brought up.



Limestone-calcined clay cement (LC<sub>3</sub>) is one such newly developed ternary blended cement in which the alumina derived from the calcined clay and carbonate derived from the limestone react with one another. Combined with the traditional pozzolanic reaction of calcined clay and the filler effect of limestone, the three primary components (clinker, calcined clay, and limestone) produce a synergistic result (Antoni et al., 2012). Metakaolin is derived from kaolin through the process of calcination, which involves heating the kaolin clay ( $Al_2Si_2O_5(OH)_4$ ) at high temperatures to transform it into a reactive amorphous material. Clay-derived metakaolin is the most crucial element in this situation. The main challenge in producing LC<sub>3</sub> is locating the best clay. Here, determine whether Kerala clays are suitable to create LC<sub>3</sub>.

## Study Area

Kaolin, known as China clay, is found in southern Kerala between Thiruvananthapuram and Kundara (Thiruvananthapuram and Kollam districts) and the Northern Kerala between Kannapuram Madayi - Cheruthazham in Kannur district to Nileswarm - Manjeshwaram in Kasaragod district. Kerala China clay is one of the finest quality clays and is world-class. The kaolin content in clay varies depending on the location and geological conditions, but it is generally available in sufficient quantities to support the local pottery and ceramics industry. These deposits are typically extracted through surface mining, and the resulting clay is then processed and refined for commercial use. Overall, the availability of kaolin-rich clay in Kerala has played an essential role in supporting the region's traditional arts and crafts and its modern industrial sector (Mineral Resources – Department of Mining and Geology).

This study mainly focuses on the China clay present in north Kerala. The Kerala Clays and Ceramic Products Limited (KCCP Limited) in Kannapuram provided the China clay, referred to as Hindustan special clay (C1). KCCP Limited used Hindustan special clay for Rubber & Fertilizer Industries. The second sample, Indian clay (C2), was sourced from the KCCP Limited mining plant in Nileswaram, in the Kasaragod district.

# Materials and Methodology

# Collection of raw materials

Department of Mining and Geology, Kerala, provided the geological databases that list the clay resources in Kerala. Samples were received from the sites where kaolin was found, i.e., Hindustan special clay (C1) and Indian clay (C2). The samples are then put through a pulveriser to ground them. Ordinary Portland 53 grade cement and limestone powder are collected from the local store. The sand used in this study was obtained from the concrete laboratory and underwent several tests to determine its properties in accordance with IS 2386 (Part 3): 1963 standards.

S. No.	Property	Value	Remarks
1	Loss of ignition from 400 to 800 °C	>7%	It is directly proportional to the kaolinite content in clay.
2	SiO <sub>2</sub>	<65%	To avoid too high Quartz content.
3	Al <sub>2</sub> O <sub>3</sub>	>24%	Required for approximately 50% kaolinite content.
4	$AI_2O_3$ / SiO <sub>2</sub> ratio	>0.38	Required for the main clay phase being Kaolinite.
5	Colour	Not important	As long as the above conditions are met

Table 1. Stand	ard properties	for selection of	kaolin clay
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# Analysis of the chemical composition of sample clays

Energy-Dispersive X-ray spectroscopy, also known as EDAX, is an effective analytical method for determining the chemical composition of materials. Awareness of the composition of silica and alumina present is vital in selecting clay. Clays with a high concentration of alumina and silica were ideal for use in LC<sub>3</sub> because they promote the formation of calcium silicate hydrates, the primary

binding agent in cement (Scrivener et al., 2018). The compatibility of sample clays is best understood when their chemical composition is compared against the pure kaolin.

The properties of clay samples are critical to the production of sustainable and environmentally friendly cement. Apart from analysing the clay samples, we will also compare them with the standard properties used for selecting kaolin clay. This comparison will assist in determining the suitability of locally sourced clay for producing sustainable cement.

#### XRD analysis and calcination of clays

Experimentation is necessary to determine the optimal temperature and holding duration for the perfect calcination. Dehydration, dehydroxylation, and recrystallisation are the three main aluminosilicate material-related events that may occur when clays samples are heated, as shown in the figure below. In the range of 100 to 150 °C, dehydration occurs. At temperatures ranging from 450 to 850 °C, the dehydroxylation process converts kaolin clay into metakaolin. When kaolin clay undergoes recrystallisation, the amorphous structure, i.e., metakaolin, that was created as a result of the water and hydroxyl groups being lost during the preceding stages is changed back into a crystalline structure. Recrystallisation takes place at temperatures exceeding 900°C (Hanein et al., 2021) which is not tolerable.

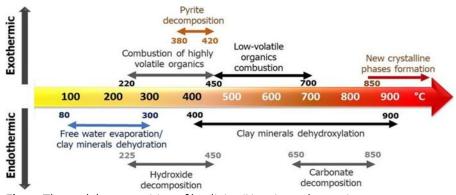


Fig. 1. Thermal decomposition of kaolinite (Hanein et al., 2021)

The crystal structure of minerals and other materials may be examined using the X-ray diffraction (XRD) method. As a result of calcination, Kaolin clay becomes metakaolin, a highly amorphous substance that lacks the characteristic crystal structure of kaolin clay.

The crystal's diffraction pattern, which reflects the arrangement of atoms in the crystal structure, is represented by the peaks in X-ray diffraction (XRD). When X-rays are focused on a crystalline substance, the atoms in the crystal lattice diffract the light, producing a distinctive diffraction pattern that may be examined to ascertain the crystal structure and composition of the substance. Atomic planes that are perpendicular to the Z axis, such as oo1 and oo2, are referred to as basal planes. Atomic planes that are not perpendicular to the Z axis, such as o20 and 110, are referred to as prism planes. The table below provides the reference peaks for kaolinite (Sachan and Penumadu, 2007).

Type of peaks	Numbering	Peaks	2θ(degrees)
Basal peaks	Kı	001	12.3
	K4	002	24.8
	K6	003	37.6
	K8	004	51.1
Prism peaks	К2	020	19.8
	K3	110	20.3
	К5	130	35.1
	K7	202	38.3

Table 2. Position of basal and prism peaks in the XRD pattern of kaolinite

In the case of kaolin, a decrease in peak intensity can indicate the transformation of the kaolin into metakaolin, as this transformation typically involves the loss of crystallinity and the conversion of crystalline phases into the amorphous phase of metakaolin (Khaled et al., 2023). The disappearance of the above-shown peaks in X-ray diffraction (XRD) analysis of kaolin can be an indicator of metakaolin formation (Elimbi et al., 2011). By examining the kaolinite peaks in the XRD test findings, the optimal dehydroxylation holding time and temperature range are identified.

The process of large-scale calcination by using a muffle furnace is carried out on locally sourced clay to compensate for the necessity of calcined clay for the production of LC<sub>3</sub> mixes. The temperature and duration of the calcination process are determined by XRD analysis. The resulting calcined clay will then be used in the production of LC<sub>3</sub> cement.

#### Determination of kaolinite content using thermogravimetric analysis (TGA)

The percentage of kaolinite content in clay can be calculated by Thermogravimetric Analysis (TGA) formula given below (Avet and Scrivener, 2018). The basic principle of TGA is to measure the change in mass of a sample as it is heated or cooled under controlled conditions. The kaolinite content wt%kaolinite is obtained from the mass loss during kaolinite dehydroxylation wt%kaol-OH by a formula (Avet and Scrivener, 2020) shown below. The analysis of the TGA graph provides information on the mass loss during kaolinite dihydroxylation.

wt%kaolinite = wt%kaol – OH × 
$$\left(\frac{Mkaolinite}{2Mwater}\right)$$

**Equation 1:** Formula to find kaolin content from mass loss during dihydroxylation Where Mkaolinite (258.16 g mol-1) and Mwater (18.02 g mol-1) refer to kaolinite and molecular weights of water, respectively.

On TGA thermograms, the mass losses during kaolinite dehydroxylation are within the range of the values (between 10 and 13.9%) that are typically reported for kaolinite (Gasparini et al., 2013). The degree of irregularity in the kaolinite structure and the quantity and kind of impurities have an impact on the dehydroxylation temperature (Cheng et al., 2010).

## Fineness of calcined clays

The fineness of the calcined clay is an important factor in the production of LC<sub>3</sub> as it affects the reactivity and strength development of the resulting concrete. Finer particles have a larger surface area, which promotes better pozzolanic reaction with the cement, leading to improved strength and durability of the concrete. Moreover, a fine particle size distribution can help to reduce the porosity of the concrete, resulting in higher density and lower permeability. The high fineness of SCM enhances clinker hydration and compensates for dilution effects, possibly due to a nucleation effect observed in calorimetry (Andrés et al., 2015). Therefore, controlling the fineness of the calcined clay is crucial for the production of high-quality LC<sub>3</sub> concrete.

The determination of the fineness of the calcined clay involved using a  $45\mu$  sieve for wet sieving as per IS 1727:1967. The percentage of the calcined clay that was retained on the sieve was then calculated, and it was ensured that this percentage was less than 34% in accordance with the guidelines set out in IS 3812 (Part 1): 2013.

#### Mix design and workability

The mix design in this study is aimed at producing an LC<sub>3</sub> with 50% and 25% replacement of clinker from OPC 53 grade cement. The ratio of calcined clay and limestone in the replacement portion of OPC was kept at 2:1 as per standard LC<sub>3</sub> composition (The Material – LC<sub>3</sub>, n.d.).

Five different trial mixes are created consisting of LC<sub>3</sub> with 25% and 50% of each clay sample, along with a control mix. The procedures for making LC<sub>3</sub> involve mixing OPC, calcined clay, and limestone in specific ratios as mentioned in the table. First, the OPC and calcined clay are dry-

mixed thoroughly. Next, the limestone is added to the dry mix. LC<sub>3</sub> trial mixes are prepared just before use.

A mini flow table test is carried out on all trial mixes to determine the water-cement ratio that yields optimal workability for the fresh mortar, according to IS: 5512:1983.

$$Flow(\%) = \frac{(Spread \ diameter - Base \ diameter \ of \ mould)}{Base \ diameter \ of \ mould} \times 100$$

Equation 2: Equation for workability

The minimum flow diameter of  $110 \pm 5\%$  after 25 drops were fixed as per IS: 4031 (Part 7):1988

Table 3. '	Various trial	l mixes used	in tł	ne stud	y
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	OPC	Calcined clay	Limestone
LC3 0%	100	0	0
LC3 25%	75	16.7	8.3
LC3 50%	50	33.3	16.7

#### Casting and testing

The process involves casting 50mm x 50mm x 50mm cubes of LC3 using different mix proportions of OPC 53 grade, calcined clay, limestone, and sand in accordance with the specifications mentioned in the table. A cement-sand ratio of 1:2 and the water-cement ratio determined from the workability test were used for casting. First, all the dry materials were weighed and mixed manually in a dry condition until the mixture became homogeneous. Then, water was added to the mix to achieve the desired water-to-binder ratio, and the mixing was continued for approximately 3-4 minutes until a uniform consistency was obtained. The mortar was then poured into 50 mm cube moulds, and proper compaction was ensured using a tamping rod. After compaction, the moulds were left undisturbed for 24 hours to allow the mortar to set. The cubes were then de-moulded and cured in water at room temperature for 3,7, and 28 days before conducting any tests. The same procedure was followed for the control mix, except that only OPC 53 grade and sand were used without any additions of calcined clay or limestone. IS: 4031 (Part 7): 1988 specifies the procedures to be followed during the casting, filling, and finishing of the specimens, as well as the curing process to be used to achieve the desired strength of the concrete. Six cubes are cast for each trial mix to test the compressive strength, this methodology is followed to ensure the accuracy and reliability of the test results.

A material or structure's ability to endure axially directed pushing forces is known as its compressive strength. 50mmx50mmx50mm cast specimens were tested for compressive strength after various curing times of 3, 7, 14, and 28 days. The specimens are set up in a universal testing machine, the cube is loaded, and the load at failure is recorded as the failure load. The compressive strength is calculated using the formula P/A, where P is the maximum load applied to the cube, and A is the cross-sectional area of the cube. The compressive strength is expressed in N/mm<sup>2</sup> or MPa (Mega pascal). The average of six cube test results is taken as the compressive strength value for a particular mix. The compressive strength was obtained from the different trial mixes of OPC 53 grade, calcined clay, and limestone are compared with the values of the control mix, which is prepared using only OPC 53 grade.

#### **Results and Discussion**

#### Analysis of chemical composition of sample clays

Clay samples from Indian Clay and Hindustan Special China Clay were obtained from two different centres of KCCPL. The Hindustan Special China Clay was in the form of a fine powder, whereas the Indian clay was hardened. The Indian clay was ground into a fine powder using a pulveriser, and both clay samples were then sieved using a 90-micron sieve. The chemical composition and physical properties of these clays were compared to those of pure kaolin clay.

Materials	Pure Kaolin	Hindustan Special China Clay	Indian Clay
SiO₂	49.28	44.97	47.94
Al <sub>2</sub> O <sub>3</sub>	35.69	37.77	36.43
Fe₂O <sub>3</sub>	0.97	1.57	0.03
TiO₂	0.06	0.50	0.93
MgO	0.07	Traces	0.12
LOI	12.55	11.1	11.6

 Table 4. Chemical composition of kaolin clay used for the present study



Fig. 2. Pulveriser

A comparison of the chemical composition of both clays obtained by the EDAX test showed many resemblances to that of pure kaolin (Mohsen and El-Maghraby, 2010). Determining the SiO<sub>2</sub> and  $Al_2O_3$  content is crucial for evaluating the suitability of the clay sample for LC<sub>3</sub> preparation.

# Comparison of standard properties of clays

According to the given table, Hindustan special China clay and Indian Clay exhibit comparable properties. The loss on ignition of both the clays from 400°C to 800°C is greater than 7%, indicating their thermal stability.

S. No.	Property	Hindustan special China clay	Indian Clay	Conditions
1	Loss on ignition from 400°C to 800°C	11.1	11.6	>7%
2	SiO2	44.97%	47.94%	<65%
3	$AI_2O_3$	37.77%	36.43%	>24%
4	$AI_2O_3$ to $SiO_2$ ratio	0.839	0.759	>0.38

Table 5. Suggested properties of China clay for use in LC3 production

The SiO<sub>2</sub> content of both clays is less than 65%, which is within the acceptable range for clay used in LC<sub>3</sub>. The Al<sub>2</sub>O<sub>3</sub> content of both clays is greater than 24%, which is desirable for their application in LC<sub>3</sub>. The Al<sub>2</sub>O<sub>3</sub> to SiO<sub>2</sub> ratio of both clays is greater than the minimum acceptable value of 0.38. Overall, both clays exhibit satisfactory physical properties for their use in LC<sub>3</sub> manufacturing.

# Calcination of clays and XRD analysis

Metakaolin is the anhydrous calcined form of the clay mineral kaolinite. Metakaolin (MK) is a pozzolanic material. It is obtained by the calcination of kaolinitic clay at a temperature ranging between 500 °C and 800 °C. The clay samples are exposed to varying temperatures and holding duration conditions, as shown in the table. The raw material input in the manufacture of metakaolin (Al<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>) is kaolin.

# Selection of optimum temperature and holding duration by XRD analysis

X-ray diffraction (XRD) analysis was carried out to determine the optimum temperature and holding duration for the calcination of kaolin clay samples C1 and C2. The XRD patterns were obtained for both samples at various calcination temperatures and durations. The results showed that for C1, the optimum calcination temperature and holding duration were 750°C and 30 minutes, respectively. For C<sub>2</sub>, the optimum conditions were found to be 650°C and 30 minutes. The XRD analysis provides important information about the phase transformation and structural changes occurring during the calcination process. The diffraction patterns obtained for the kaolin clay samples at different calcination temperatures and durations show distinct changes in the intensity and position of the diffraction peaks, indicating the transformation of the clay mineral structure.

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S. No.	1	2	3	4	5	6
Temperature(°C)	650	650	650	750	750	750
Duration(min)	30	60	90	30	60	90

Table 6: Optimum temperature and holding duration



Fig. 3. Small scale calcination

For C1, the XRD pattern obtained at 750°C for 30 minutes showed the disappearance of kaolin peaks, indicating the completion of the dehydroxylation process. The absence of reference peaks indicated that the desired product was obtained. On the other hand, for C2, the XRD pattern obtained at 650°C for 30 minutes showed a decrease in the intensity of peaks corresponding to the formation of metakaolin, indicating the dehydroxylation of the clay. The difference in calcination temperatures between C1 and C2 indicates that the energy consumption for the calcination process of C1 is lower compared to C2. This indicates that C2 is more suitable for LC3 production compared to C1. By selecting these optimum conditions, we were able to ensure the maximum transformation of the clay minerals into metakaolin while minimising energy consumption and processing time.

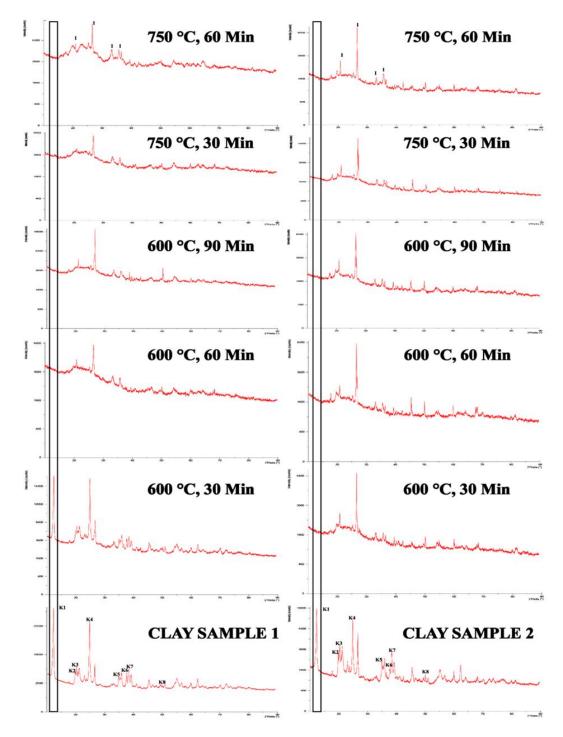


Fig. 4. XRD of calcined C1&C2 various temperature and holding duration
\*K1 – K8: Denotes the peak corresponding to kaolinite.
\*I: Denotes the impurity present in the clay
\*Vertical slender box shows the disappearance of a peak

# Large scale calcination for tests

Large-scale calcination is performed in a muffle furnace to obtain calcined clay. The Hindustan Special China Clay is then subjected to full calcination using a large muffle furnace, with the selected optimum temperature and holding duration of 750°C for 30 minutes. This condition has been chosen to reduce the duration of calcination. At the same time, the ideal condition for Indian clay is chosen as 650°C for 30 minutes.



Fig. 5. Calcined clay and calcination of clays using muffle furnace

# Calculation of kaolinite content in clays Thermogravimetric analysis (TGA)

The TGA analysis of kaolin clay samples C1 and C2 shows a mass loss of 11.1% and 11.6%, respectively. The kaolinite content of C1 and C2 was found to be 79.587% and 83.172%, respectively. These values suggest that both samples are relatively stable up to the temperature at which the mass loss occurs. The mass loss in TGA analysis suggests that both kaolin clay samples C1 and C2 have an almost similar level of thermal stability (Cheng et al., 2010). The higher kaolinite content in C2 may be due to differences in the depositional environment or geological history. Research conducted at EPFL (Alujas et al., 2015; Avet et al., 2016) has demonstrated that incorporating only 40% kaolinite content in a blend of LC3-50% is enough to produce mechanical properties similar to those of plain Portland cement after approximately 7 days. So, both the clays are compatible with LC3 production as per the obtained kaolin content.

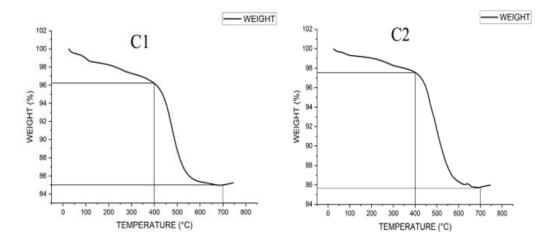


Fig. 6. TGA thermograms of Hindustan special China clay and Indian clay

Checking the properties of constitue	nts
Specific gravity of sand	2.6
Fineness of sand	3.6
Fineness of sand	1.746 g/cc
Maximum bulking of sand	29.4%
Loose density of sand	1.592 g/cc
Percentage voids	32.84%

# Preliminary tests Checking the properties of constituents

# Fineness of calcined clays

The fineness of the calcined clays was determined using a 45µ sieve, ensuring that the percentage retained on the sieve was less than 34% as per IS 3812 (Part 1): 2013.

Table 8.	Fineness	of clays
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	Sieved using 45µ sieve				
Clay sample	Percentage passed	Percentage retained			
C1	85	15			
C2	74	26			

To check the workability of trial mixes



Fig. 7: Flow table test

The minimum flow diameter of 110  $\pm$  5% after 25 drops are considered necessary for easy placement of mortar in moulds. The flow is determined by measuring the increase in the average base diameter of the mortar mass after at least four equispaced measurements, expressed as a percentage of the original base diameter. This flow measurement is a crucial factor in ensuring that the mortar is of the appropriate consistency to allow for easy and effective placement in the desired location. Equation (3) represents the equation to find the flow percentage, where D<sub>avg</sub> is the average base diameter and D<sub>o</sub> is the original base diameter.

Flow(%) = 
$$\left(\frac{\text{D avg} - \text{D}_0}{\text{D}_0}\right) \times 100$$

Equation 3. Formula for workability

400g of sand and 200g of LC3 cement are used for mixes, with a ratio of calcined clay to the limestone of 2:1. This ratio is in accordance with the standards of LC3 composition.

When comparing the water-cement ratio between C1 and C2, it is evident that there is a significant increase in the ratio as the content of the calcined clay and limestone mix increases. However, C2 exhibits better workability with a lower water-cement ratio, indicating that it is more suitable for replacing conventional cement. There are problems associated with an increased water-cement ratio in a mix. Firstly, it can lead to a decrease in the compressive strength of the final product. Additionally, it can cause an increase in porosity and permeability, leading to decreased durability and an increased risk of moisture ingress. Furthermore, an increased water-cement ratio can lead to shrinkage and cracking of the material as it dries and ages (Chan et al., 2018). Therefore, it is important to maintain a suitable water-cement ratio in any mix to ensure the desired properties and performance of the final product. The flow table test was conducted to determine the water-cement ratio of each mix design, and these values were utilised for casting the cubes.



Fig. 8. Weighing and mix preparation for flow table test



LC3-50%



OPC

9	5	 -	-	 -

Table 9. Flow table test results of various mixes					
Trial mix name	Trial No.	Water added(grams)	Water cement ratio	Flow %	Remarks
LC3 0%	1	90	0.45	98.75	
	2	95	0.475	106.75	Appropriate
LC3 50% C1	1	140	0.7	106.25	Appropriate
LC3 25% C1	1	120	0.6	95	
	2	125	0.625	107.25	Appropriate
LC3 50% C2	1	120	0.6	83.75	
	2	130	0.65	109.25	Appropriate
LC3 25% C2	1	115	0.575	106	Appropriate

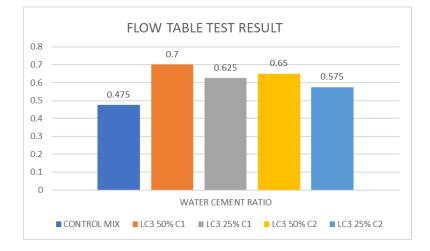


Fig. 10. Graphical representation of water cement ratio of various mixes

# Mortar Compressive strength



Fig. 11. Mortar cubes of Cement, LC3 50% C1, LC3 25% C1, LC3 50% C2, LC3 25% C2

The compressive strength of  $5 \text{ cm} \times 5 \text{ cm} \times 5 \text{ cm}$  cubes was determined, and the compressive strength of LC3 cubes with different mix designs was compared with that of OPC cubes. The diagram shows the compressive strength (in MPa) of various mix designs of cement mortars cured for different time periods. The control mix (without any clay) shows the highest compressive strength values at all curing periods.

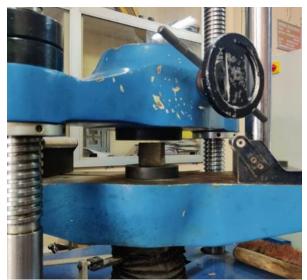


Fig. 12. Determination of Compressive strength using UTM

However, the LC3 mix designs with 25% substitution of C1 and C2 clays show some improvement in compressive strength as the curing period increases but still remain lower than the control mix. But C2 has the potential to achieve comparable compressive strength results to OPC 53 grade, whereas C1 falls short in this aspect. The graphical representation shows that as the amount of clay content increases, there is a decrease in the rate of strength gain. The slope of strength gain over time is lower for both clays with 50% substitution compared to other mix designs. The decrease in the availability of portlandite due to the use of calcined clay with high metakaolin content in a low clinker system can result in a lower degree of clinker hydration (Cardinaud et al., 2021). This can ultimately reason lead to a decrease in the compressive strength of the LC3 50%.

Interestingly, the LC<sub>3</sub> mix designs with 25% substitution of C<sub>1</sub> and C<sub>2</sub> clays show a similar trend to that of the control mix in terms of compressive strength gain with curing days. At 28 days, the compressive strength of the LC<sub>3</sub> mix designs with 25% substitution of C<sub>2</sub> clay is even parred with that of the control mix, which indicates the potential of this mix design in achieving comparable strength to OPC.

Overall, the results suggest that the use of calcined clays as partial replacements for clinker in cement production can lead to the reduced compressive strength of the resulting cement mortars. However, carefully selected mix designs with appropriate substitution levels and curing periods can improve the strength and potentially achieve comparable strength to OPC.

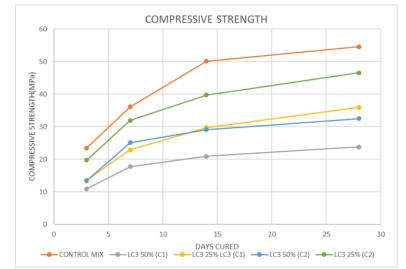


Fig. 13. Graphical representation of compressive strength during various curing age

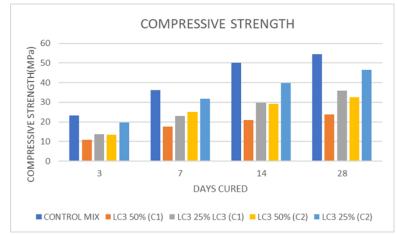


Fig. 14. Comparison of compressive strength of various mixes in different curing periods

# Conclusion

The chemical composition of both clays obtained by the EDAX test showed many resemblances to that of pure kaolin, and determining the SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> content is crucial for evaluating the suitability of the clay sample for LC3 preparation. The chemical composition of both clays obtained by the EDAX test showed many resemblances to that of pure kaolin, and determining the SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> content is crucial for evaluating the suitability of the clay sample for LC<sub>3</sub> preparation. The kaolinite content values suggest that both samples are relatively stable up to the temperature at which the mass loss occurs, and both the clays are compatible with LC3 production as per the obtained kaolin content. XRD analysis determined the optimum calcination conditions for kaolin clay samples C1 (750°C, 30 minutes) and C2 (650°C, 30 minutes), maximizing metakaolin transformation while minimizing energy consumption and processing time. Higher clay composition increases water-cement ratio, impacting strength and durability of cement mortar; C2 has lower ratio than C1 but slightly higher than control mix. Higher clay content leads to slower strength gain, particularly with 50% substitution, as indicated by reduced slope of strength gain over time compared to other mix designs. The decrease in the availability of portlandite due to the use of calcined clay with high metakaolin content in a low clinker system can result in a lower degree of clinker hydration, ultimately leading to a decrease in the compressive strength of the LC3 50% which is also substantiated by SEM analysis. Impure sources of kaolin clays can be utilized to achieve desirable properties in cementitious materials, allowing for the use of abundant lowergrade kaolinite clays, with a minimum 40% kaolinite content required for LC3 manufacturing. Partial replacement of clinker with calcined clays in cement production may initially reduce compressive strength; however, with carefully selected mix designs, appropriate substitution

levels, and proper curing, strength can be improved and potentially match that of OPC. Both clays are suitable for LC<sub>3</sub> production: C1 for low-strength cement and C2 as a 25% cement substitute matching OPC 43 grades, aligning with the increasing demand for cement alternatives.

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#### **Author Contributions**

NKAR, VS, AUP and RB conceived the concept, wrote and approved the manuscript.

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# Availability of data and materials

Not applicable.

# Competing interest

The authors declare no competing interests.

# Ethics approval

Not applicable.



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