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A comparative study on the effect of fineness of low-grade calcined clays on engineering properties of binary and ternary blended concretes

#	Name	Email Address	Degree	Affiliation	
1	MOODI, FARAMARZ	fmoodi@aut.ac.ir	Ph.D.	Department of civil and environmental engineering, Amirkabir University of Technology	
2	Fazelhashemi, Ali	alifh7777@gmail.com	MSc	Department of civil and environmental engineering, Amirkabir University of Technology, Tehran, Iran.	
3	GivKashi, Mohammad Rasul	mrgivkashi@aut.ac.ir	MSc Student	Department of civil and environmental engineering, Amirkabir University of Technology, Tehran, Iran.	
4	Banar, Rasoul	rbanar1995@gmail.com	Ph.D. Candidate	Department of civil and environmental engineering, Amirkabir University of Technology, Tehran, Iran	
5	Ramezanianpour, Amirmohammad	ramezanian@ut.ac.ir	Ph.D.	School of Civil Engineering, College of Engineering, University of Tehran, Tehran, Iran	
6	Ramezanianpour, Aliakbar	aaramce@aut.ac.ir	Ph.D.	Department of civil and environmental engineering, Amirkabir University of Technology, Tehran, Iran	

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Abstract

The fineness of low-grade calcined clay (CC) particles in limestone calcined clay cement (LC3) and calcined clay cement (C3) plays an important role in terms of grinding time and energy consumption. In this regard, two low-grade kaolinitic clays (kaolinite content of less than 40%) from domestic sources were firstly calcined and then ground using a laboratory ball mill to achieve three fineness values of ~ 8 ± 2, 20 ± 2, and 32 ± 2 wt% retaining on 45 μm sieve. According to experimental results, the substitution of finer low-grade CCs in binary C3 and ternary LC3 concretes had a marginal improvement in the compressive strength, bulk water absorption, and electrical resistivity by 4.7, 5.2, and 14.5% compared to their counterpart coarser low-grade CCs mixtures, respectively. However, the chloride ions migration coefficients of mixtures containing low-grade CCs with the lowest fineness were 81-107% and 100-134% of their counterpart mixtures with the highest fineness of CC particles at 28 and 91 days, respectively. Overall, the low-grade CCs with a fineness of ~32 ± 2 wt% retaining on 45 μm sieve by using less grinding time and energy consumption delivered a satisfactory mechanical and durability performance against chloride attack.

Keywords: Limestone calcined clay cement (LC3), Fineness, Low-grade calcined clay, Kaolinite content, Chloride ions attack

1. Introduction

The production and availability constraints of common supplementary cementitious materials (SCMs), such as fly ash, silica fume, blast furnace slag, and natural pozzolans prevent them from satisfying the increasing demands of the construction industry (Scrivener et al., 2018a; Riahi Dehkordi and GivKashi, 2024). On the other hand, kaolinitic clays and calcium carbonate minerals like aragonite and calcite are plentiful resources that have the potential to broaden the utilization of SCMs in blended cements (Scrivener et al., 2018a; Zolfagharnasab et al., 2021; Du and Dai Pang, 2020; Díaz et al., 2017). In fact, calcined clays (CCs) are clays that have been thermally activated through dehydroxylation in the temperature range of 650-850 ℃ (Richardson, 2002). By comparing different types of clay, kaolinite-based CCs have the highest pozzolanic reactivity and have the potential to improve the mixtures properties (Fernandez et al., 2011). A novel approach has been introduced, combining kaolinitic CCs with limestone powder (LP) as SCMs. This method has resulted in the development of ternary blends known as limestone calcined clay cement (LC3) (Scrivener et al., 2018a). Additionally, this new ternary blend also has the potential for use in advanced mixes like self-compacting concrete or self-compacting lightweight concrete (Mazloom et al., 2020; Mazloom and Salehi, 2018; Afzali-Naniz et al., 2021). Although other SCMs such as slag and fly ash are generally regarded as having no additional emissions and energy consumption, CC due to the need for calcination, does have an impact. However, the calcination process for clay involves significantly lower temperatures compared to the clinker production, and there is no CO_2 emitted during the decomposition of clay (Sharma et al., 2021). Studies have indicated that the energy needed for the clays calcination is approximately 60% of that needed for clinker production, and the CO2 emissions are around 30% of those generated by clinker production (Gettu et al., 2019). In terms of environmental impact, a study by Pillai et al. (2019) found that LC3 concretes, which exhibit comparable compressive strengths to Portland cement concretes, could potentially have a CO_2 footprint that is 16% to 30% lower.

Due to the use of metakaolin (MK) by other industries (mainly painting, paper, and ceramic industries), pure kaolinite clay has become more expensive (Barata and Angélica, 2011). Therefore, using CCs with low purity is proposed as an alternative option. Studies on LC3 production show that clay with at least 40% kaolinite can be used as a 50% substitution of Portland cement. So, the utilization of low-grade clays with reduced kaolinite content could potentially address these constraints and also facilitate the utilization of waste materials in cement manufacturing. Initiatives have been undertaken in this direction, such as incorporating 30% rejected clay brick (FRCB) in the formulation of LC3-50 (with a 50% clinker ratio) (Krishnan et al., 2020). Du and Dai Pang (2020) investigated the concrete durability with various replacement levels of cement by CC and LP. Permeability, electrical conductivity, RCMT, and water sorptivity of LC3 concretes were decreased compared to the control mixture. Maraghechi et al. (2018) concluded that the purity of CC has a remarkable effect on the concrete durability against chloride ion transport.

Investigating the influence of fineness of particles and kaolinite content of low-grade CCs on the technical properties of binary and ternary blends containing CC, respectively named calcined clay cement (C3) and limestone calcined clay cement (LC3), is one of the necessary studies in this field. For instance, Bishnoi et al. (2018) produced LC3 blends by mixing 50% clinker, 31% CC, 15% LP, and 4% gypsum on a laboratory scale. The test results showed that while Blaine's fineness test could be a useful initial indicator for controlling the grinding process, laser diffractometry can offer a more comprehensive evaluation of the grinding of all cementing constituents. Krishnan et al. (2018) discussed that the industrial production of LC3 concretes comprising 50% clinker, 30% CC, 15% LP, and 5% gypsum. The findings indicate that grinding-based improvement in fine particles of MK does not always lead to enhanced early-stage hydration of C_3S. In a research by Lapeyre et al. (2019), the role of the particle size distribution (PSD) of MK in the hydration kinetics of tricalcium silicate paste (C_3S–T1) was investigated. Also, Dumani and Mapiravana (2020) investigated the influence of different fineness of MK on the compressive strength of cementing pastes containing MK. The findings showed that the compressive strengths of the cementing pastes containing MK were not remarkably affected by the MK fineness, within the particle size range of 45 to 75 µm. This means that even the coarser MK can be used as a substitute for OPC, thus helping to decrease the expenses associated with grinding. Malacarne et al. (2021) investigated the influence of utilizing low-grade clay as a clinker substitute on the fresh properties of ternary cements. Their results demonstrated that the influence of the fineness of calcined limestone and clay on thixotropic and water demand of pastes is more considerable relative to their mineralogical compositions. Ayati et al. (2022) used eight types of low-grade CC as a replacement of cement. The index of pozzolanic activity and strength activity index of clays were determined. They also showed that mortars containing CC have higher compressive strengths than mortars containing pulverized fuel ash.

Li et al. (2022) studied the influence of calcined MK content on water demand, pozzolanic activity, and mixture efficiency. The results showed higher pozzolanic reactivity and improved compressive strength with increasing MK content. In addition, higher MK content and fineness increased the water demand significantly. Kluge and Assmann (2018) investigated the effect of different mills of CCs on the short-term strength (7 days) of mixtures. The variables used in this study were cement fineness, CC fineness, and the amount of CC by considering three variables for each factor. The results indicated that the effect of CC fineness was less significant compared to that of cement and the amount of cement. Also, with increasing fineness, the strength increased, but with increasing the amount of CC, the strength decreased. Andrés et al. (2015) studied the impact of fineness of clinker, LP, and CC on compressive strength and mercury penetration porosity. They showed that CC fineness is almost as important as clinker fineness, but LP's fineness has a significant effect only at early ages. Ferreiro et al. (2019) studied the influence of fineness and calcination temperature of raw clays on the LC3 binders performance. They showed that the 2-day strength of blended cement was affected by clinker substitution. In contrast, the long-term efficiency and strength were influenced by LP content, fineness, raw clay composition, and temperature. Pérez and Martirena-Hernandez (2020) studied the concurrent grinding of LC3 binders and assessed how grinding duration affects the PSD of CC, LP, and clinker in the performance of these blends. The findings suggested that as grinding time increased, fineness improved, leading to enhanced compressive strength, albeit with an increase in water demand.

CC fineness is the main influencing factor on rheology and water demand, while compressive strength is controlled by clinker fineness. Zunino and Scrivener (2020) compared two fragmentation methods to increase kaolinite content in low-grade CC. The findings indicated that kaolinite tends to be concentrated in finer particles after the grinding process, and the separation of particles can elevate the kaolinite content of CCs and enhance their pozzolanic reaction. Zolfagharnasab et al. (2021) investigated the durability of LC3 binders containing low-grade CCs against chloride attack. They showed that these mixtures could be recognized as a promising option with sufficient durability. Yadak Yaraghi et al. (2022) investigated the suitability of conventional tests for assessment of the durability and permeability of low-grade CC mixtures. Based on the findings, it was determined that the water absorption test based on BS 1881-122, could not effectively determine the penetration characteristics of binary and ternary binders containing CC. Siline and Mehsas (2022) researched to explore the impact of enhancing the MK fineness on its chemical reactivity. Their findings indicated that increasing the MK fineness does not appear to enhance its chemical reactivity. This conclusion was drawn based on the observation that the strength activity index remained unchanged. Luzu et al. (2022) investigated the optimal ratio of LC3 binders to obtain the highest possible packing density. The results showed that grain size distribution is not a critical parameter, especially in mortars, and adding LP fillers improves packing density. Also, Ferreiro et al. (2017) studied the impact of fineness and type of raw clay on the efficiency and strength of binders. The early strength of blended cements appears to be mainly independent of particle fineness. On the other hand, the long-term performance can be remarkably enhanced by increasing the fineness of CC.

Although research has been conducted in this area, studies are still limited and there are discrepancies between opinions among researchers. Most existing studies have only investigated these concrete's pozzolanic reactivity, hydration kinetics, and efficiency. Among the hardened properties, compressive strength has often been the only investigated parameter. Nevertheless, more research is still needed to attain a better finding of the effect of CC's fineness and kaolinite content on the mechanical, permeability, and chloride-induced durability of ternary LC3 and binary C3 concretes.

This study aimed to fill the gap in existing studies regarding the effect of the fineness of low-grade CC on the technical properties of C3 and LC3 and compare the results with Portland cement (PC) concrete. In this regard, two types of low-grade CCs with kaolinite content of 19.4% and 28.7% from local resources were used. Low-grade CCs were ground by laboratory ball mill to achieve three fineness values of ~ 8 ± 2, 20 ± 2, and 32± 2 wt% retaining on 45 µm sieve. The results of ternary blends (containing 20% CC and 10% LP) were compared with corresponding binary blends (containing 30% CC). In other words, the potential of using lower fineness of low-grade CCs to produce LC3 concretes while maintaining desirable engineering properties has been studied. Also, a Distance-Based Approach (DBA) has been performed to compare the performance of different mix designs.

2. Experimental program

2.1. Material

In this research, a Type II Portland cement (PC) was used. The C_2S, C_3S, C_3A and C_4AF phases contents of PC were 17.75%, 58.64%, 5.10% and 11.81%, respectively. It should be noted that the used PC meets the requirements of ASTM C150 (2020). In addition, two types of low-grade CCs with different kaolinite contents were gathered from local resources located in north-western parts of Iran. The preparation of powdered samples of these raw clays for calcination are shown in Fig 1. Both raw clays were calcined in a fixed-bed electrical furnace at 800 ◦C for 1 hr. The calcination process of CCs is shown in Fig 2.

Clay sample 1
Kaolinite content: 19.4%

Clay sample 2
Kaolinite content: 28.7%

Fig 1. Samples of raw clays before calcination process (Bahman-Zadeh et al., 2022).

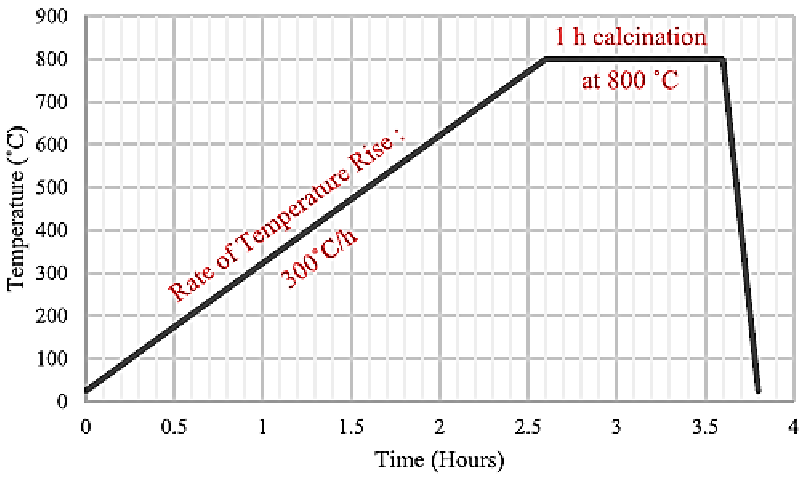


Fig 2. The process of calcination of CCs (Yaraghi et al., 2022).

The kaolinite content of the CCs was measured by determining the weight loss between approximately 400–600 degrees Celsius using thermogravimetric analysis (TGA) with the tangential method (Scrivener et al., 2018b). TGA was performed using the STA 504 instrument in an atmosphere of Ar gas, with a heating rate of 10 ℃ per minute. The IR spectrum of raw clay samples and TGA results are presented in Fig 3 and Table 1, respectively.

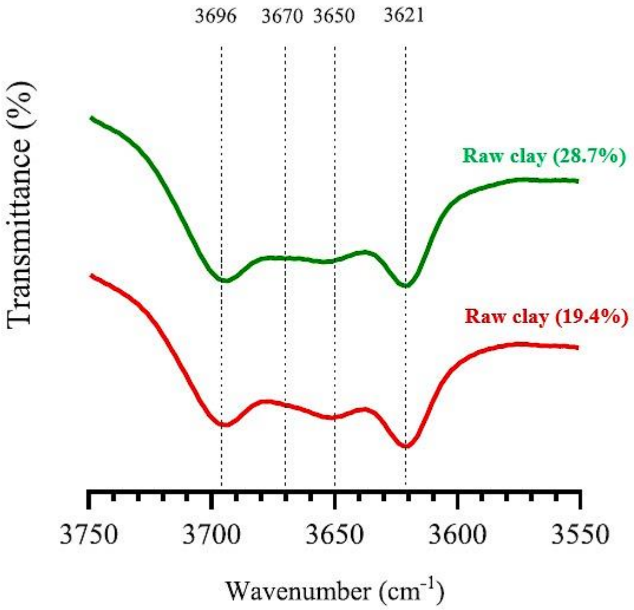


Fig 3. IR test spectrums of studied raw clays.

Table 1. TGA test results for CCs

Material	Approximate temperature range for dehydroxylation (℃)	Mass loss (%)	Amount of kaolinite from TGA test (%)
CC (19.4%)	540-640	2.71	19.4
CC (28.7%)	420-600	4.00	28.7

Binders including ASTM C150 Type II PC, two various CCs from different domestic deposits and LP. The chemical composition as achieved by X-ray fluorescence (XRF) and the physical properties of PC, CCs and LP are given in Table 2.

Table 2. Chemical Characteristics of cementitious materials by XRF analysis.

Chemical compounds	SiO_2	Al_2O_3	Fe_2O_3	CaO	Na_2O	K_2O	MgO	SO_3	L.O.I.	Density (gr/cm^3)
PC	21.62	4.29	3.88	64.89	0.41	0.78	2.29	2.41	1.52	3.10
CC (19.4%)	78.71	19.4	0.23	0.04	0.04	0.74	-	0.06	0.60	2.58
CC (28.7%)	71.30	22.6	0.09	2.70	0.05	0.33	0.09	0.01	2.57	2.55
LP	1.78	0.47	0.26	49.9	-	0.32	2.26	0.97	43.83	2.62

The composition of aggregates consisted of 55% sand, 20% fine gravel, and 25% coarse gravel, to be within the range recommended by the National Concrete Regulations. Table 3 shows the maximum size, density, and moisture percentage of aggregate components. Fig 4 shows the aggregate grading curve.

Table 3. Specifications of aggregates used include; Density, water absorption and moisture content

Aggregate	Maximum size (mm)	Density (gr/cm^3)	SSD (%)	Moisture available (%)
Coarse gravel	19	2.57	1.97	0.27
Fine gravel	12.5	2.55	2	0.5
Sand	9.5	2.54	2.2	0.65

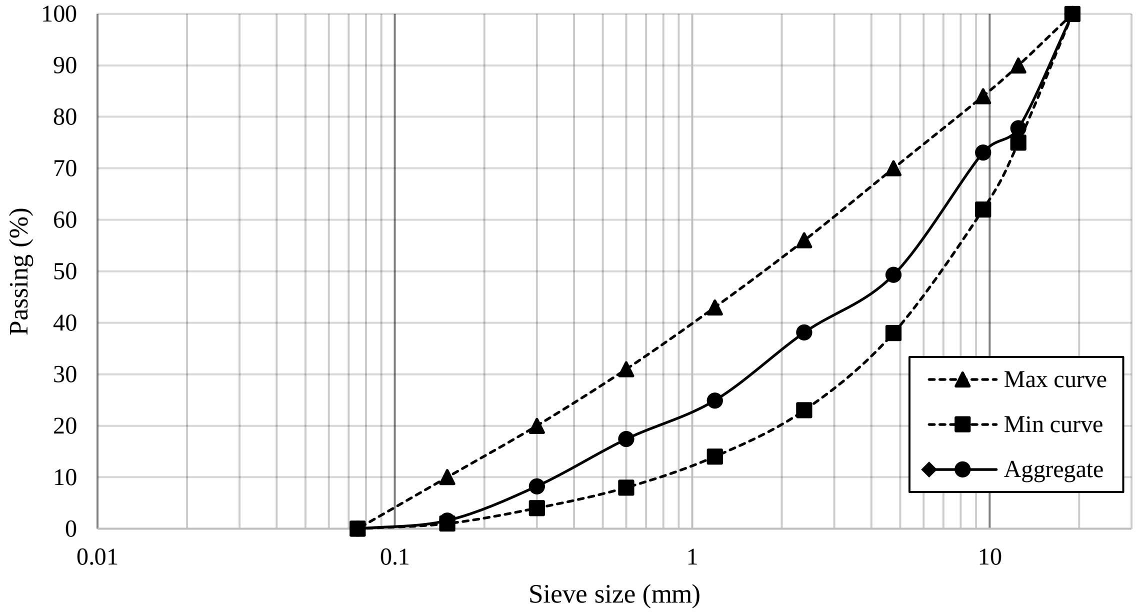


Fig 4. Aggregate grading curve with a maximum size of 19 mm

Considering that CCs and LP absorb water and reduce the workability of the mixture, and due to the need to keep the ratio of water to binder constant for making samples, using superplasticizer (SP) as a chemical admixture was inevitable in this research. The SP admixture used in this research was based on polycarboxylate ether according to the specification of G class of ASTM C494 standard (2020).

2.2. The process of preparing blended cements

In this research, in addition to ternary blends (including PC, CC, and LP), binary blends (including PC and CC) and a plain mixture containing PC have also been used to compare the results. In total, 13 types of binders were prepared in this research, and 13 mix designs were obtained by keeping other variables constant. Using low-grade CC with two different purity percentages and three different fineness values has been investigated.

The clays were calcined in a furnace at 800°C, and then the CCs were ground using a laboratory ball mill to make their particle diameter smaller than 4 mm. Low-grade CCs with 19.4% and 28.7% purity were used. CCs were crushed into three levels of fineness using a ball mill. All the CCs were passed through a 100-grade sieve during grinding. The percentage of retaining on the 45-micron sieve was used to measure fineness. The highest fineness, close to the fineness of PC, i.e., the remaining percentage of 8±2 on the 45-micron sieve, was selected. The lowest fineness, the lowest allowed limit according to ASTM regulations, i.e., 32±2 percentage of residue on a 45-micron sieve, and average fineness, 20±2% residue on a 45-micron sieve was selected. According to the ASTM standard, the maximum percentage of residue on the 45-micron sieve can be 34%. LP was also ground using a ball mill close to the fineness of PC, i.e., the remaining percentage was 8±2 on a 45-micron sieve. Fig 5 shows the process of calcining, crushing, and grinding of CCs.

a b c d

Fig 5. Preparation of blended cements; a) calcination of clay in the furnace, b) crushing machine, c) grinding machine, d) ball mill grinding

Binary blends were prepared with a ratio of 70% PC and 30% CC, and ternary blends with a ratio of 70% PC, 20% CC, and 10% LP. The particle size distribution curve of CCs obtained after production using a laser beam diffraction device is shown in Figures 6 and 7. Fig. 6 shows the cumulative particle size distribution and Fig. 7 shows their density distribution. By comparing the distribution of particles, it is clear that even though the percentage of residue on the 45-micron sieve was considered to be almost similar in the two types of CC, the CC (28.7%) particles were softer than the CC (19.4%) particles.

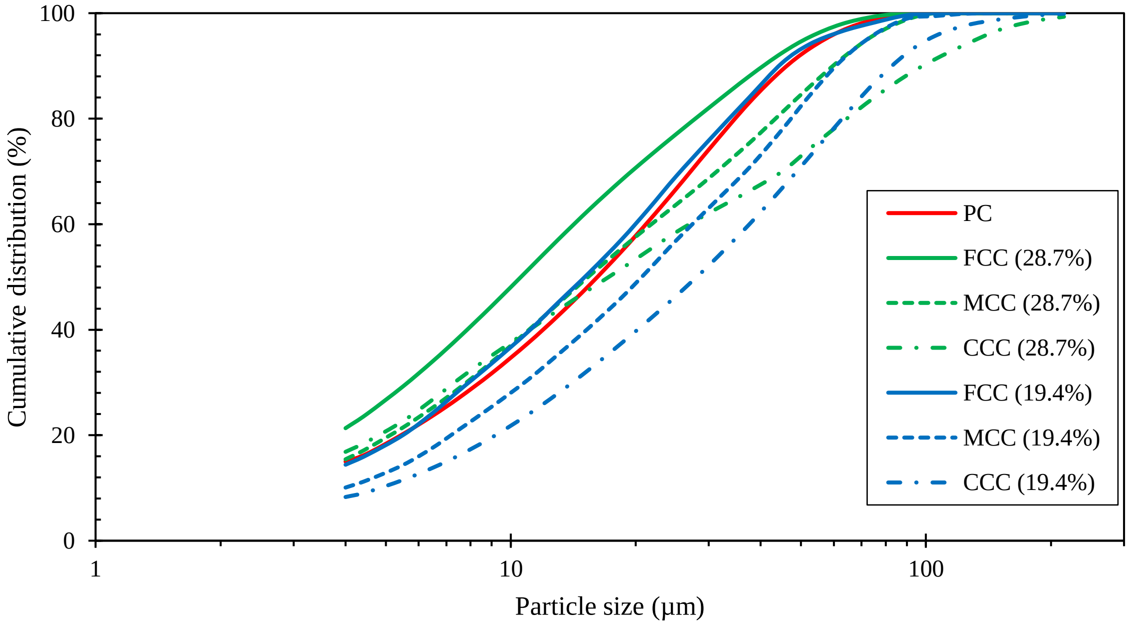


Fig 6. Comparison of cumulative particle size distribution

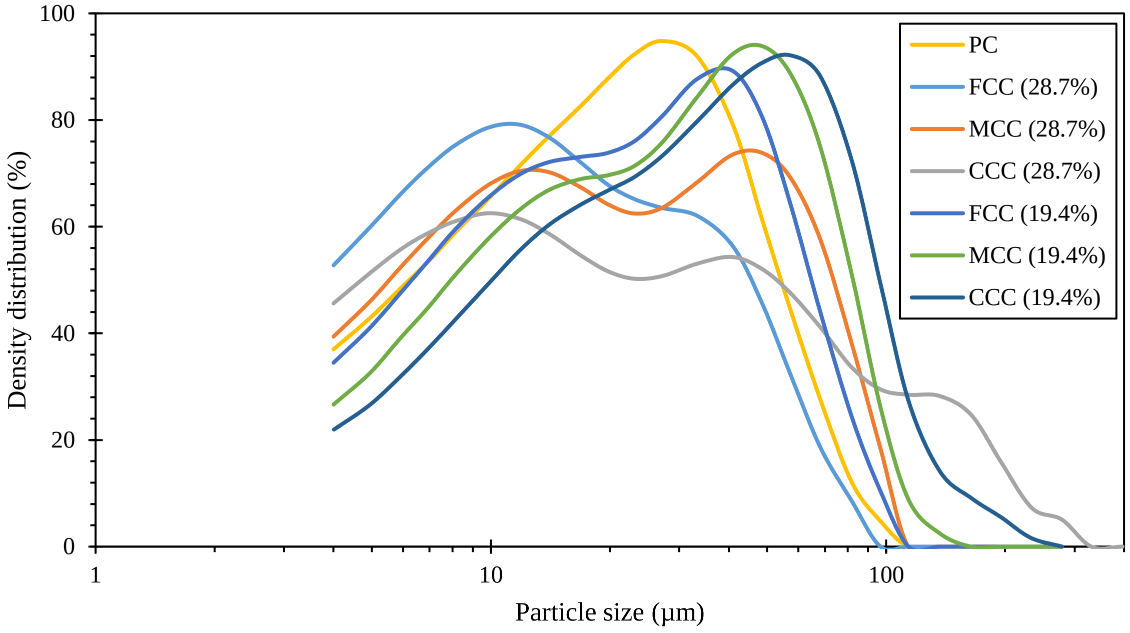


Fig 7. Comparison of particle size density distribution

2.3. Mixing procedure

Table 4 shows the design of the mixtures used in this study according to the components. In this study, thirteen concrete mixtures with total binder contents of 400 kg/m^3 and constant water to binder ratio of 0.4 were considered. In binary blends, the amount of PC replaced by CCs was 30%. In ternary blends, the substitution levels of PC were 20% for CCs and 10% for LP. The slumps of all mixtures were adjusted between 30 and 60 mm (S1-S2) using SP. The dosages of SP have been expressed as weight percentages of binders.

The concrete mixing process was done using a 60-liter pan mixer. The following steps outline the sequence for mixing materials and preparing concrete samples:

- Mixing gravel and sand with 1/3 of water for 1 min;
- Adding dry SCMs to mix and mixing for 0.5 min;
- Adding PC, the remaining water, and mixing for 1.5 min;
- Modifying the slump by incorporating SP and thoroughly mixing for 1.5 min;
- Molding concrete samples and 24 hour-curing under a plastic sheet at room temperature (~25 ˚C);
- Demolding samples and curing until testing days in a saturated limewater (RH~100%, ~21 ˚C).

Table 4. Specifications of concrete mix designs (Kg/m^3)

Mix. ID	Binders			Gravel	Sand	Water	SP (gr)
	PC	CC	LP				
OPC	400	797.3	-	797.3	967.0	160	60
FC3 (19.4%)	280	787.7	-	787.7	955.3	160	74
MC3 (19.4%)	280	787.7	-	787.7	955.3	160	67
CC3 (19.4%)	280	787.7	-	787.7	955.3	160	67
FC3 (28.7%)	280	788.2	-	788.2	955.9	160	63.9
MC3 (28.7%)	280	788.2	-	788.2	955.9	160	58.1
CC3 (28.7%)	280	788.2	-	788.2	955.9	160	54.2
FLC3 (19.4%)	280	788.2	40	788.2	955.9	160	59.7
MLC3 (19.4%)	280	788.2	40	788.2	955.9	160	55.1
CLC3 (19.4%)	280	788.2	40	788.2	955.9	160	55.4
FLC3 (28.7%)	280	788.6	40	788.6	956.3	160	57.2
MLC3 (28.7%)	280	788.6	40	788.6	956.3	160	54.1
CLC3 (28.7%)	280	788.6	40	788.6	956.3	160	51.3

(F: Fine, M: Medium, C: Coarse), (C3: Calcined Clay Cement), (LC3: Limestone Calcined Clay Cement), (CC: Calcined Clay), (LP: Limestone Powder), (SP: Superplastisizer)

2.4. Test methods

As previously stated, the aim of this research was to assess the durability of concretes with varying levels of fineness of low-grade CCs. Therefore, the conducted experiments were divided into three main categories: mechanical properties, permeability, and durability (Fig 8).

2.4.1. Compressive strength

The compressive strength test (Fig 8-c) was performed at 3, 7, 28, 90, and 180 days of curing according to the ASTM C39 standard (2021). Cubic samples with dimensions of 10 cm were stored to test the compressive strength until the test age in saturated lime water. For each test age, the average results of three samples were reported.

2.4.2. Bulk water absorption

Bulk water absorption of the samples was conducted based on the BS 1881-122 standard (2020). As mentioned in previous studies (Yaraghi et al., 2022; Antoni, M., 2012), ettringite exhibits strong stability in LC3 blends attributed to the incorporation of LP and the creation of CO3-AFm compounds, with the presence of this phase noted in matured samples. However, ettringite becomes unstable at temperatures exceeding 50°C; thus, preparation techniques involving high temperatures are not advised (Scrivener et al., 2018b). Therefore, following the sample preparation procedure outlined in BS 1881-122 for water absorption measurement (drying concrete samples for 72 hours at 105°C) could lead to notable modifications in the microstructure of LC3 binders and affect the mixtures. Consequently, utilizing BS 1881-122 for evaluating the permeability of LC3 concrete presents significant limitations, and adjustments to the sample preparation method are necessary for this type of mixture. Alternative techniques such as extended drying at 50°C and vacuum drying of specimens could be considered for the modified water absorption test preparation (Yaraghi et al., 2022). For this reason, all studied samples were kept at 50˚C for 14 days and then placed in a sealed room for 24 hours to overcome this drawback.

2.4.3. Water sorptivity

To measure the amount of water sorptivity, disk-shaped samples with a diameter of 10 and a height of 5 cm, which were obtained by cutting cylindrical samples of 20 x 10 cm, were used, and the test was performed according to the ASTM C1585 standard (2020). First, the samples were kept in an oven at a temperature of 50°C for 14 days. Then after weighing, the samples were placed in a container of water, so that the water level was 2±1 mm above the bottom of the samples, while the water with the bottom of the sample should be in complete contact (Fig 8-d). It should be noted that after removal from the oven, the samples were completely covered with insulating tape to prevent the absorption of moisture from the environment. For each sample, three specimens were prepared at the ages of 28 and 90 days.

2.4.4. Electrical resistivity

For this test, cylindrical concrete samples with dimensions of 10x20 cm were made and stored in saturated lime solution until the test age. For each mixture design, three samples were tested and the average results were reported. The electrical resistance results were read in all four directions in each sample. The test was performed according to ASTM C1760 (2021) at 7, 28, 90, and 180 days. Fig 8-b shows a picture of this experiment.

2.4.5. Rapid chloride migration test (RCMT)

This test was performed according to NT BUILD 492 (1999) at the ages of 28 and 90 days. One sample was tested for each age. Samples with a diameter of 10 cm and a height of 5 cm obtained by cutting cylindrical samples were tested. Fig 8-a shows a view of the RCMT test.

2.4.6. Mercury intrusion porosimetry (MIP)

Mercury intrusion porosimetry (MIP) was performed on paste samples that had been cured in limewater for 28 days. To stop the hydration process, the samples with dimensions of around 10 mm × 10 mm × 3 mm, underwent treatment using the solvent exchange technique outlined in Reference (Snellings et al., 2018). Thermo Finnigan Pascal 440 and 140 porosimeters, featuring a maximum mercury injection pressure of 182 MPa, were employed in this investigation. The contact angle between the solid and mercury was adjusted to 140°, and the surface tension of mercury was determined as 0.480 N/m. These parameters were used to compute the pore radius.

a b c d

Fig 8. Pictures of different experiments; a) Chloride ion migration, b) Surface electrical resistance, c) Compressive strength test, d) Capillary water absorption.

3. Results and discussion

3.1. Compressive strength

Fig. 9 indicates the compressive strength development of C3 and LC3 concretes between 3 and 91 days. At all ages, the compressive strengths of all concretes containing CCs were lower than OPC concrete. It can be due to the dilution effect of using 30% SCMs (20% CC + 10% LP) instead of clinker (Yaraghi et al., 2022). As it is illustrated, C3 mixtures containing CC (28.7%) showed higher compressive strength in comparison with CC (19.4%) binary mixtures. Elevating the kaolinite content in CCs typically leads to enhanced compressive strengths of C3 concretes. This improvement can be primarily attributed to the increased reactivity of CCs, particularly when they possess higher purity levels of MK (Zolfagharnasab et al., 2021). However, in LC3 concretes, there was no remarkable difference between compressive strength results of CC (19.4%) and CC (28.7%) ternary mixtures.

Comparing the results of binary and ternary samples containing CCs, ternary LC3 (19.4%) concretes had a compressive strength comparable to that of binary C3 (19.4%) concretes. In comparison, the compressive strength of LC3 (28.7%) concretes was lower than corresponding C3 concretes. The comparable compressive strength of C3 and LC3 concretes containing low-grade CC (19.4%) attributed to the filling effect of LP and the reactions of alumina-containing phases in CCs with $CaCO_3$ from LP (synergic reactions) leads to the formation of additional CO_3-AFm phases. Therefore, it appears that in LC3 (19.4%) mixtures, the filler effect of LP and synergistic reactions may compensate for the lower pozzolanic reactivity of CC at 19.4%. However, when CC content is increased to 28.7%, the impact of the pozzolanic reaction on compressive strength may outweigh the positive effects of using LP and CC together. Specifically, replacing 20% of OPC with CC in LC3 (28.7%) mixtures could result in the formation of fewer pozzolanic hydrates compared to binary mixtures containing 30% CC (Bahman-Zadeh et al., 2023).

In order to understand the effect of the fineness of CC particles on C3 and LC3 concretes, the relative compressive strength of each mixture compared to their corresponding mixtures with the highest fineness of CCs (i.e., FC3 or FLC3 mixtures) is demonstrated in Fig 10. Generally, the coarser CCs containing mixtures showed less compressive strength than finer CCs mixtures. The fineness of CCs had more influence at early ages, so the difference between the minimum and maximum relative compressive strengths of mixtures varied from 19% to 6% at

3 and 91 days, respectively. This better early age-compressive strength of finer CC mixtures can be attributed to the smaller PSD of CC can lead to improved particle packing, filling the gaps between clinker grains (Poon et al., 2001). Additionally, the finer CC particles can act as nucleation sites, promoting nucleation effects and potentially enhancing the overall performance of the cementitious system (Andres et al., 2015). However, the results confirmed that the coarser CCs obtained by less grinding time and energy consumption, had the great potential of being used in C3 and LC3 concretes in terms of compressive strength.

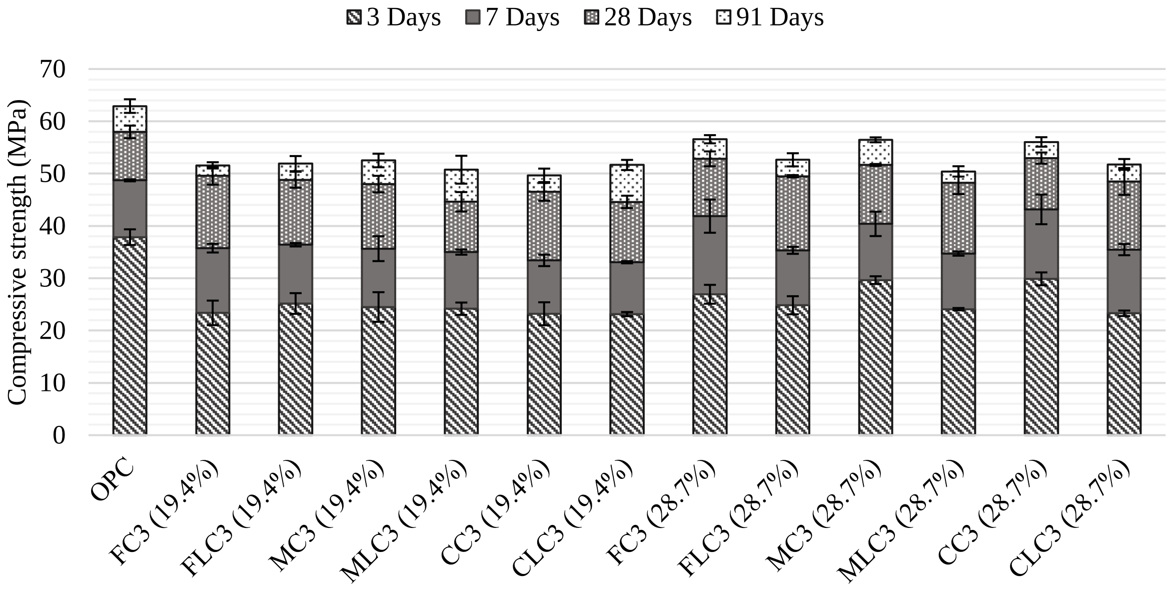


Fig 9. Compressive strength of studied mixtures.

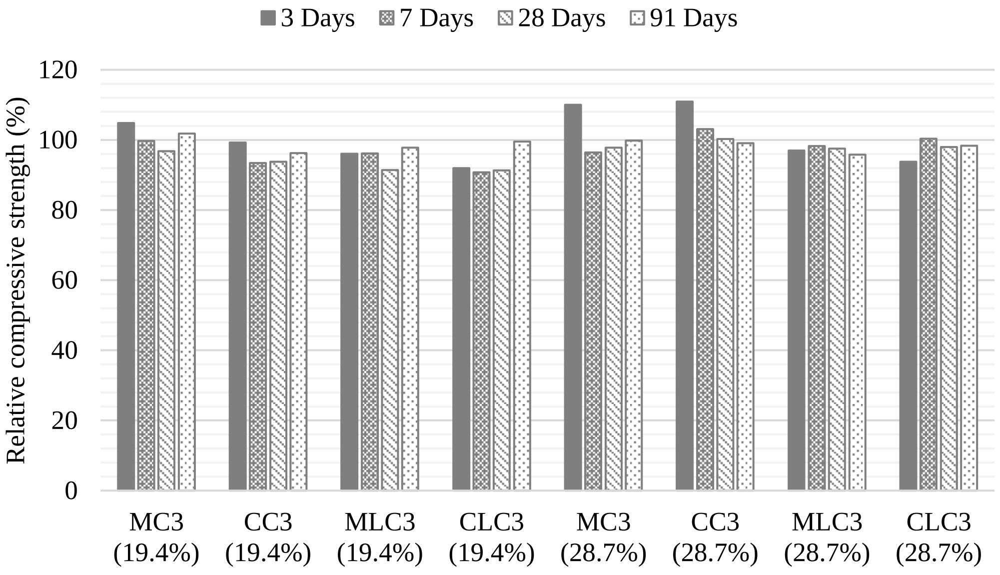


Fig 10. Relative compressive strength of mixtures compared to the corresponding mixture with the finest CCs.

3.2. Bulk water absorption

The results of water absorption test are shown in Fig 11. According to the results, incorporating CCs (either in binary or ternary binders) led to increased permeability by up to 34% compared to OPC mixture. These findings are in contradiction with the results of (Zolfagharnasab et al., 2021; Yaraghi et al., 2022), which reported lower water absorption for CCs mixtures. These contradictory results can be attributed to difference in test methods. Both of them (Zolfagharnasab et al., 2021; Yaraghi et al., 2022) have determined that using the BS 1881-122 method may change the hydrated cement system and result in inaccuracies when assessing permeability. The lower water absorption of binary blends containing CCs compared to their counterpart ternary mixtures is primarily due to the higher amounts of CCs in C3 concretes and the greater formation of pozzolanic products. This enhancement is more noticeable in the CC (28.7%) mixtures, which was up to 34% difference between CC3 (28.7%) and CLC3 (28.7%). Moreover, because of the higher pozzolanic reactivity of CC (28.7%) compared to CC (19.4%), the water absorption rates of CC (28.7%) mixtures were slightly lower than those of CC (19.4%) blends. It can be concluded that the blends made with the finer CCs exhibited a little less and almost identical water absorption values compared to the coarser CCs. The water absorption of mixtures with the highest fineness of CC, including FC3 (19.4%), FLC3 (19.4%), and FLC3 (28.7%) was 20%, 7%, and 3% lower than CC3 (19.4%) and CLC3 (19.4%), respectively. However, in FC3 (28.7%), the water absorption was 9% higher than in CC3 (28.7%). This exception can be due to the agglomeration of finer platy particles of CC around the clinker grains that resulted in higher permeability (Andres et al., 2015).

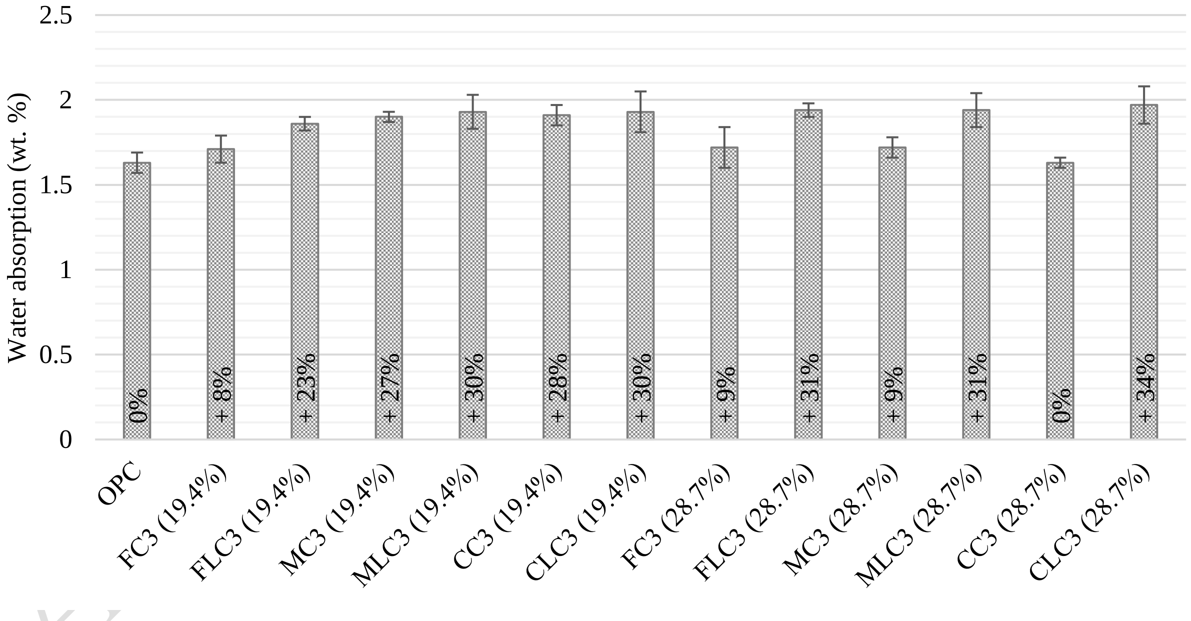


Fig 11. 28-day water absorption values and difference with OPC mixture.

3.3. Water sorptivity

Table 5 illustrates the initial and secondary rates of water absorption after 28 and 91 days of curing. According to the findings, the water sorptivity of the mixtures is reduced as the curing time advances, as a result of the completion of hydration reactions of cementitious materials that alter the micro-pore structure and diminish the water transport rate of capillary suction. From 28 to 91 days of curing, the initial rate of water absorption for OPC, C3 binary, and LC3 ternary concretes has been reduced by up to 33%, 53%, and 45%, and the secondary rate of water absorption reduced by up to 36%, 72%, and 54%, respectively. As a general deduction, all C3 binary mixtures except MC3 (19.4%) and CC3 (19.4%) showed lower initial and secondary rates of water absorption after 91 days. Nevertheless, until 28 days, only binary mixtures including FC3 (19.4%), FC3 (28.7%), and CC3 (28.7%) had similar or lower initial and secondary rates of water absorption. These can be due to the filling effect of the finer particles in the cementitious matrix of binary mixtures containing the finest CCs in FC3 (19.4%) and FC3 (28.7%) and the highest content of metakaolin in CC3 (28.7%) and FC3 (28.7%) mixtures. Generally, LC3 ternary mixtures had more S_i and S_s relative to their corresponding C3 binary mixtures which can be attributed to the lower content of CCs in LC3 mixtures. In binary blends containing CC (19.4%) and ternary blends containing CC (28.7%), by increasing the fineness of CCs the initial rate of water absorption was reduced; however, in other cases, it remained almost constant.

Table 5. Initial and secondary rates of water absorption

Mixture ID	28 Days		91 Days	
	$S_i \times 10^{-4}$ (mm/√s)	$S_s \times 10^{-4}$ (mm/√s)	$S_i \times 10^{-4}$ (mm/√s)	$S_s \times 10^{-4}$ (mm/√s)
OPC	128.7	33.9	85.7	21.9
FC3 (19.4%)	134.0	21.4	63.1	11.7
FLC3 (19.4%)	166.7	37.7	101.6	21.8
MC3 (19.4%)	142.9	41.2	96.6	11.4
MLC3 (19.4%)	157.6	34.7	99.6	24.9
CC3 (19.4%)	136.6	36.1	100.7	15.2
CLC3 (19.4%)	159.0	34.4	98.3	25.8
FC3 (28.7%)	133.3	17.9	78.6	10.6
FLC3 (28.7%)	137.1	34.1	84.7	29.9
MC3 (28.7%)	140.3	36.4	84.4	12.9
MLC3 (28.7%)	175.2	22.8	96.6	27.4
CC3 (28.7%)	112.8	36.3	76.9	10.5
CLC3 (28.7%)	150.4	33.1	136.5	15.2

S_i : Initial rates of water absorption, S_s : Secondary rates of water absorption.

3.4. Electrical resistivity

Fig. 12 shows the surface electrical resistivity of concretes. The electrical resistivity of concrete is directly influenced by the permeability, the ionic composition of the pore solution, and the connectivity of pores in the concrete microstructure (Ramezanianpour et al., 2011). At 7 days, the electrical resistivity of C3 binary blends incorporating CC (19.4%) was up to 10.3% lower than that of the OPC mixture. It can be due to the dilution effect, pozzolanic reactivity, and lower kaolinite content of CC (19.4%) at early ages. Besides, C3 binary concretes containing CC (28.7%) showed higher electrical resistivity compared to binary CC (19.4%) mixtures by about 1.5 times after 91 days. After 28 and 91 days of curing, all CC containing concretes displayed higher electrical resistivity. This enhancement is primarily associated with the refinement of the concrete's pore structure, as well as the increased tortuosity of the paths for ion transport within the concrete pore solution (Du and Dai Pang, 2020). All ternary LC3 concretes demonstrated lower electrical resistivity in comparison with their corresponding binary mixtures. This difference was more remarkable between binary and ternary concretes containing CC (28.7%). It seems that the replacement of LP in LC3 concretes resulted in lower electrical resistivity due to the lower kaolinite content of ternary binders as opposed to binary mixtures, as also reported by Yadak Yaraghi et al. (2022). In comparison with three different fineness of CCs, Fig 13 showed that the binary and ternary binders demonstrated slightly better electrical resistance relative to coarser CC particles mixtures. In other words, at 91 days, the electrical resistivity of FC3 (19.4%), FLC3 (19.4%), FC3 (28.7%), and FLC3 (28.7%) was 6%, 7%, 1%, and 4% than those of CC3 (19.4%), CLC3 (19.4%), CC3 (28.7%), and CLC3 (28.7%), respectively. Thus, the mixtures with lower fineness (coarser particles) of CCs had comparable electrical resistivity while using less energy consumption and grinding time compared to higher fineness (finer particles) mixtures containing CCs.

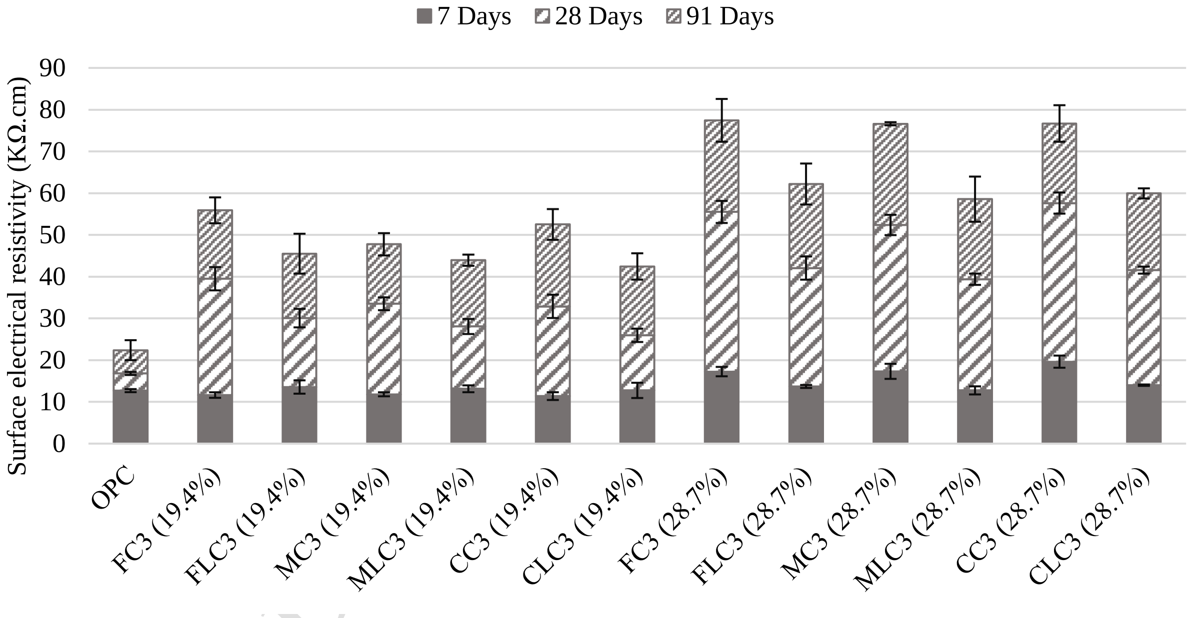


Fig 12. Surface electrical resistivity of concrete mixtures.

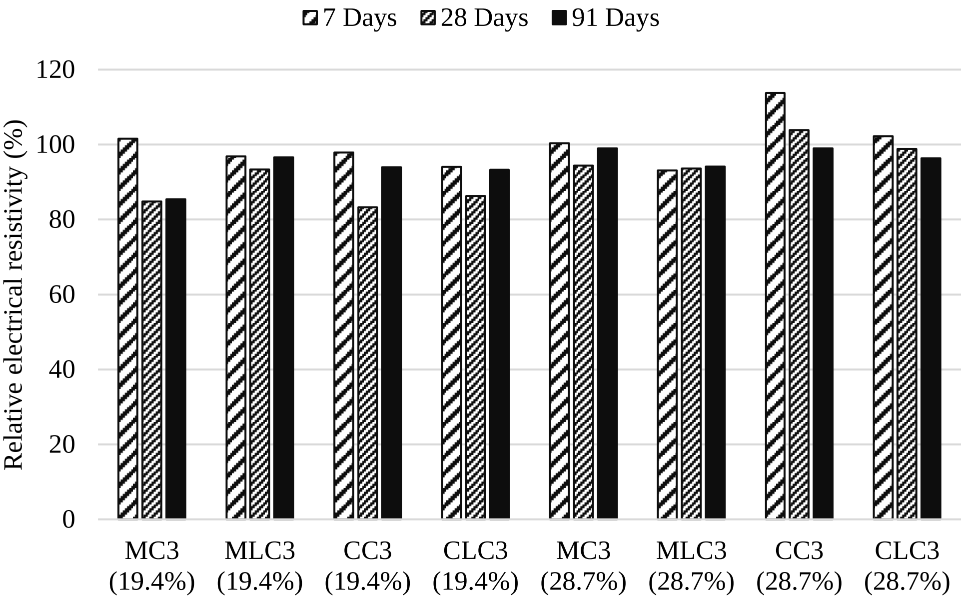


Fig 13. Relative electrical resistivity of mixtures compared to the corresponding mixture with the finest CCs.

3.5. Rapid chloride migration test (RCMT)

The RCMT results for concrete specimens after 28 and 91 days of curing are demonstrated in Fig 14. The results show that 30% replacement of PC drastically reduced the D_{nssm} values for concretes incorporating SCMs, especially at 90 days. The C3 binary mixtures containing CC (19.4%) outperformed the counterpart ternary LC3 mixtures at 28 and 91 days. In binary and ternary blends containing CC (28.7%), LC3 mixtures had comparable D_{nssm} values at 28 days, but after 91 days of curing, C3 mixtures showed lower D_{nssm} relative to LC3 concretes. This could suggest that the creation of carboaluminate compounds was not sufficient to offset the impact of using reduced amounts of low-grade CC in LC3 mixtures (Yaraghi et al., 2022). As expected, the C3 and LC3 concretes containing the higher kaolinite content of CC (28.7%) had remarkably reduced the chloride coefficient compared to CC (19.4%) containing mixtures. As for various fineness of CCs mixtures, results in Fig 15 revealed that the chloride ions migration coefficient of mixtures with the coarser particles were from 81-107% and 100-134% of their counterpart mixtures with the finest CC particles at 28 and 91 days, respectively. Therefore, by the progress of cementitious materials' hydration and pozzolanic reaction at later ages, the durability of mixtures containing the lower fineness of CC against chlorides enhanced significantly compared to their corresponding mixtures with higher fineness. In C3 and LC3 concretes, the fineness of low-grade CCs was more effective in the reduction of D_{nssm} values at early ages until 28 days.

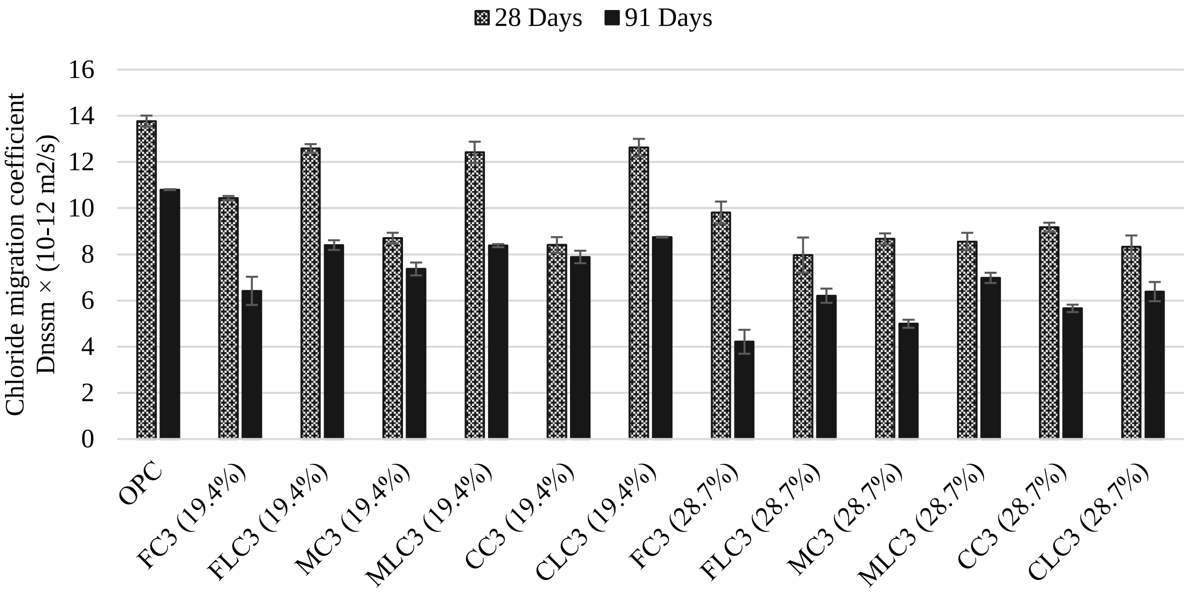


Fig 14. Results of the rapid chloride migration test.

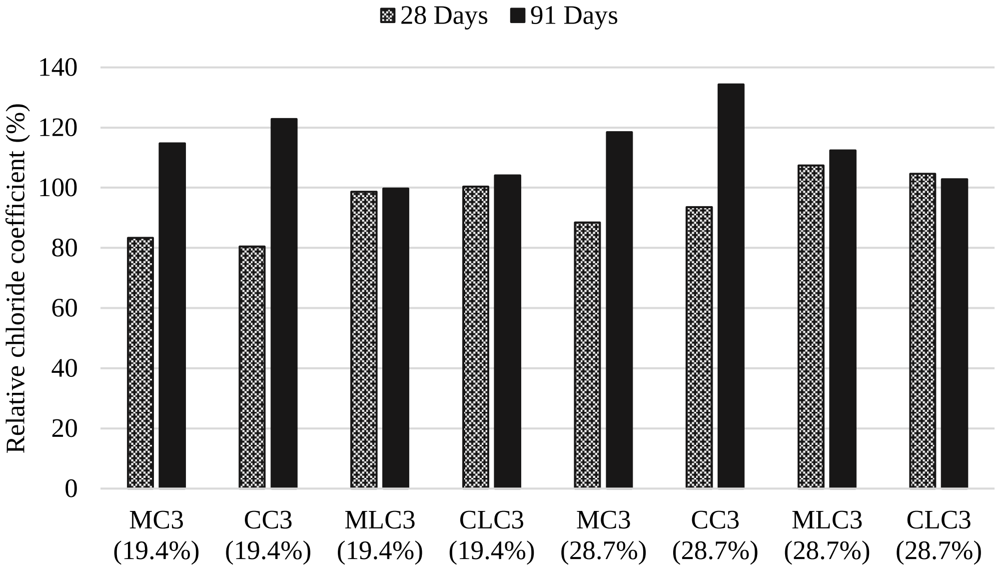
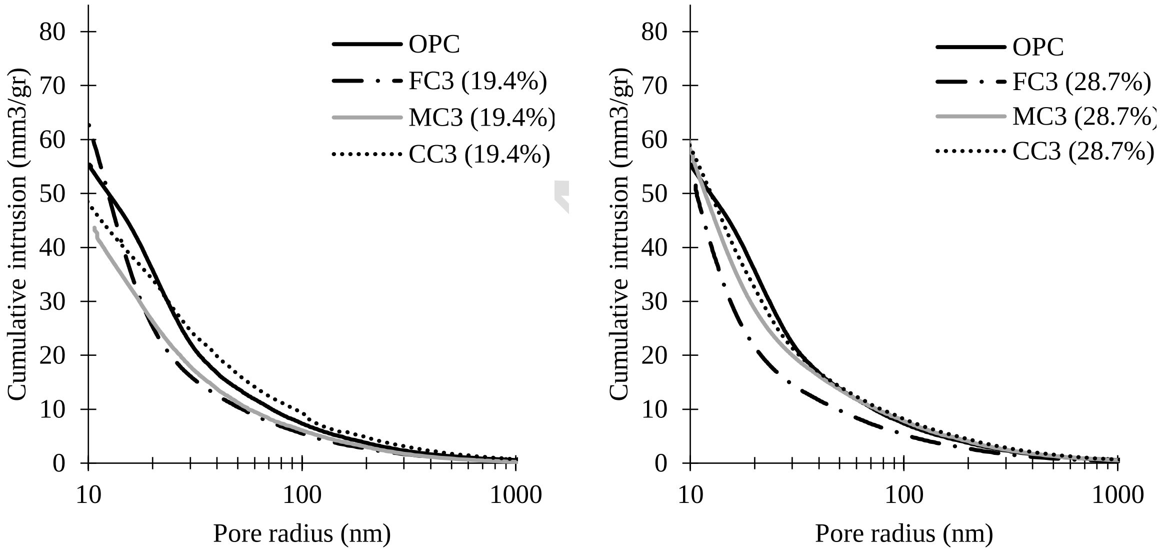


Fig 15. Relative chloride ions migration coefficient of mixtures compared to the corresponding mixture with the finest CCs.

3.6. Mercury intrusion porosimetry (MIP)

The cumulative intrusion of mercury in cementitious pastes is shown in Fig 16. Generally, the C3 binary pastes exhibited a more refined pore structure and finer pore connectivity compared to OPC paste. In addition, in LC3 ternary pastes (except for CLC3 (19.4%) and CLC3 (28.7%) due to the coarser particles of CC), the impermeability of pastes was enhanced in comparison with the control paste. According to the results, the blends made with finer CCs (i.e., FC3 and FLC3) demonstrated a lower total porosity and significantly greater pore refinement compared to the blend containing coarser CCs. This pore refinement may be attributed to the combined effect of enhanced particle packing resulting from a much smaller PSD, which could fill the gaps between cement grains (Poon et al., 2001) and the higher pozzolanic reaction of the CCs as similarly stated by other researchers (Andres et al., 2015; Bahman-Zadeh et al., 2022). The pozzolanic hydrates formation in the pastes containing CC may contribute to the densification of the microstructure and pore refinement (Bahman-Zadeh et al., 2022). In addition, the threshold pore diameter (as defined in Ref. (Scrivener et al., 2018b)) and average pore diameter of pastes are given in Table 6. It can be seen that by increasing the fineness of CCs in binary and ternary blends, the average diameter of pores had changed towards smaller values and a reduced threshold pore diameter was achieved. In addition, the C3 binary pastes had smaller average pore diameters than their corresponding LC3 ternary blends. A similar trend for threshold pore diameter has been observed for binary and ternary binders containing CCs except for MC3 (19.4%) and CC3 (19.4%) relative to their corresponding LC3 pastes. Also, the average pore diameter of all CC containing pastes was smaller than OPC paste (19.4 nm) except for MLC3 (19.4%) and CLC3 (19.4%) which were 25.2 and 26.1 nm, respectively. In addition, based on the results in Table 6, the threshold and average pore diameters of mixtures containing CC (28.7%) were smaller than CC (19.4%) mixtures. This further refinement of the porosity in higher kaolinite content CC pastes was observed in previous studies(Avet and Scrivener, 2018; Bahman-Zadeh et al., 2022).



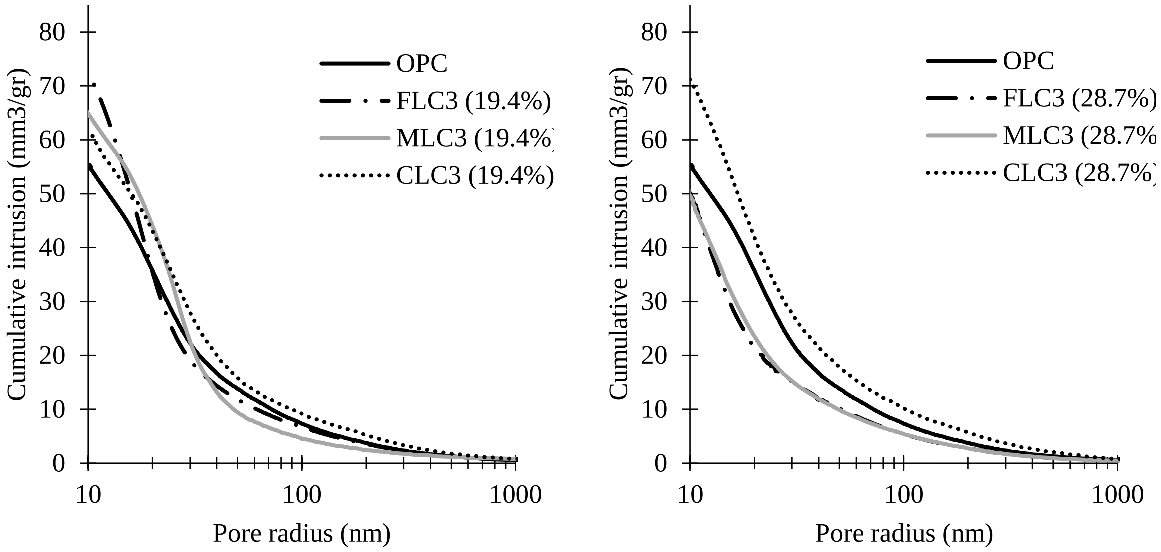


Fig 16. MIP results of CC pastes in comparison with PC paste.

Table 6. Threshold and average pore diameters of studied mixtures.

Mixture ID	Threshold pore diameter (nm)	Average pore diameter (nm)
OPC	50.6	19.4
FC3 (19.4%)	26.6	11.6
MC3 (19.4%)	54.7	11.5
CC3 (19.4%)	110.2	10.0
FLC3 (19.4%)	32.9	17.0
MLC3 (19.4%)	43.3	25.2
CLC3 (19.4%)	69.1	26.1
FC3 (28.7%)	27.1	11.3
MC3 (28.7%)	37.0	10.5
CC3 (28.7%)	42.5	12.6
FLC3 (28.7%)	26.7	11.7
MLC3 (28.7%)	39.5	10.2
CLC3 (28.7%)	59.2	16.4

3.7. Discussion

In this study, a Distance-Based Approach (DBA) (Kashi et al., 2017) is utilized for comparing different mix designs and achieving an optimal design. In this technique, the first step is to determine decision criteria. Two criteria of strength and durability have been selected as decision criteria. The durability criterion includes various experimental results such as bulk water absorption, water sorptivity, electrical resistivity, RCMT, and MIP. Table 7 illustrates the normalized values of decision criteria.

Table 7. Normalized values of different decision parameters.

Criterion	1. Strength	2. Durability				
Mix. ID	Compressive strength	Bulk water absorption	Water sorptivity	Electrical resistivity	RCMT	MIP
OPC	2.70	1.63	0.50	-1.95	-2.18	-0.84
FC3 (19.4%)	-0.43	0.98	1.66	-0.02	0.11	0.61
FLC3 (19.4%)	-0.29	-0.25	-1.17	-0.65	-1.12	0.63
MC3 (19.4%)	-0.40	-0.57	-0.13	-0.51	0.34	0.91
MLC3 (19.4%)	-0.81	-0.82	-0.85	-0.79	-1.06	-0.39
CC3 (19.4%)	-0.92	-0.65	-0.03	-0.38	0.28	-1.92
CLC3 (19.4%)	-0.95	-0.82	-0.87	-0.93	-1.23	-2.09
FC3 (28.7%)	0.76	0.90	1.37	1.49	0.95	0.67
FLC3 (28.7%)	-0.29	-0.90	0.05	0.36	0.91	0.82
MC3 (28.7%)	0.75	0.90	0.39	1.35	1.06	0.43
MLC3 (28.7%)	-0.61	-0.90	-1.00	0.11	0.50	0.59
CC3 (28.7%)	1.01	1.63	1.48	1.62	0.71	0.87
CLC3 (28.7%)	-0.51	-1.14	-1.38	0.28	0.74	-0.28

After determining the decision criteria, they need to be standardized for calculating the distance index. Ultimately, the distance between each alternative for the optimal state is derived from Equation 1. Another point regarding calculating the distance index is that different criteria need to be weighted to determine their importance. Various weighting scenarios are considered. Equal importance between two criteria and considering only one criterion are among the investigated scenarios. Furthermore, Fig 17 depicts the changes in the distance index for different mix designs based on different weights.

$$D_k = \left\{ \sum_j \left[W_j * \left(z_{kj} - z_{*j} \right) \right]^2 \right\}^{0.5} \qquad \text{(Eq. 1.)}$$

Where: k= Number of alternatives, D_k= Distance index for the kth alternative, j= Number of criteria, z_{kj}= Standardized value of the kth alternative with respect to criterion j, z_{*j}= Benchmark value of criterion j, W_j= Weight of criterion j.

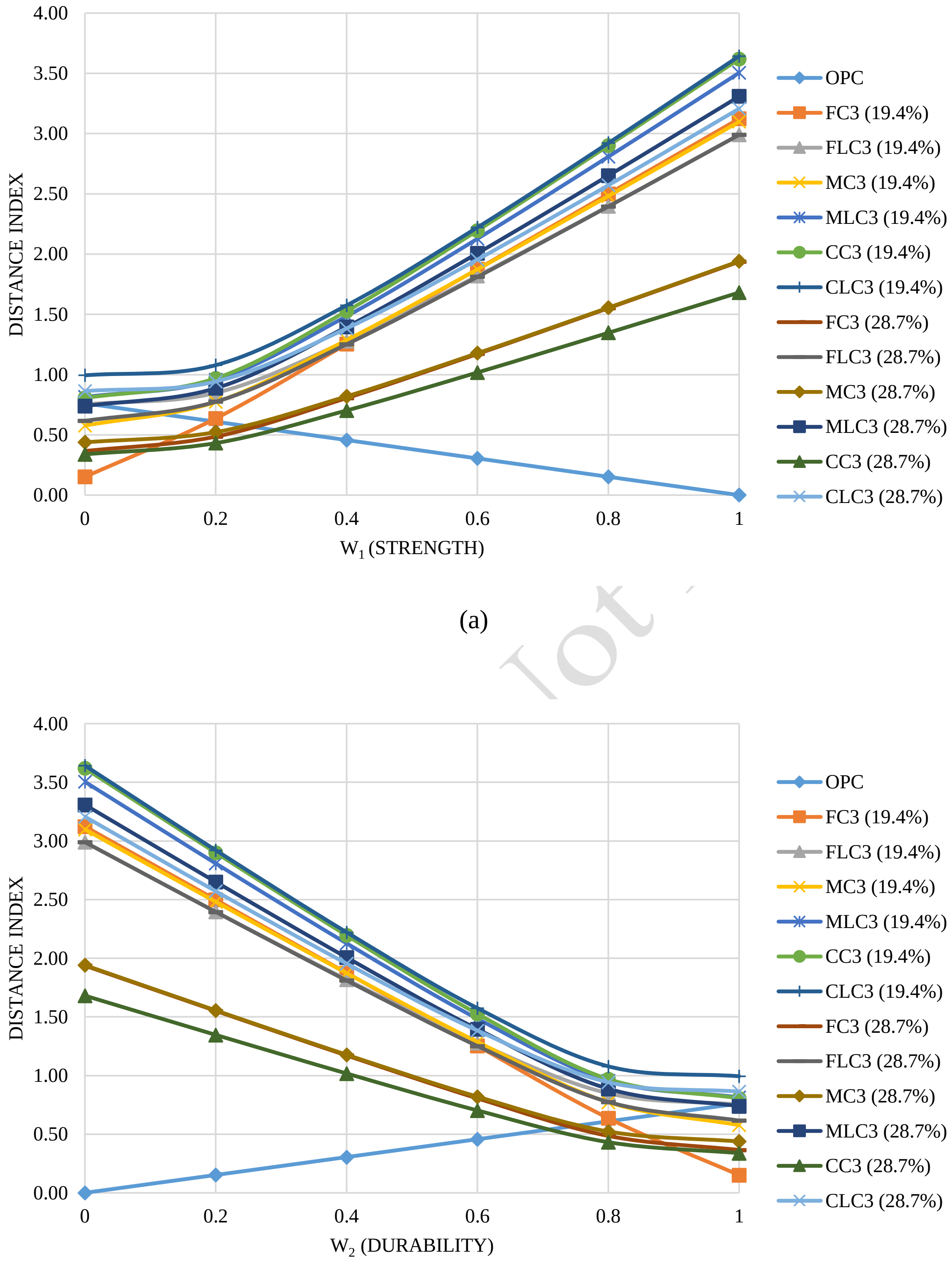
According to the DBA concept, the optimal alternative has the lowest distance index. According to the results in Table 8, the mixture of OPC will be the best design when the importance of both decision criteria is equal. Among clay-containing specimens, the binary combination CC3 (28.7%) shows the best performance considering both strength and durability criteria. Considering only the strength criterion, the OPC sample outperforms the binary and ternary mixtures containing CC considerably. This is due to the decrease in strength caused by using CCs. Also, as indicated by the durability criterion, FC3 (19.4%), CC3 (28.7%), and FC3 (28.7%) will be the best mixtures, respectively. The finer particle size and higher grade of CC-containing mixtures have improved the performance. However, among ternary mixtures containing CC and LP, FLC3 (28.7%) will be the best mixture.

In general, according to the results in Table 8 and Fig 17, increasing the fineness and grade of CCs in mixtures reduces their distance index, and they exhibit better performance in various tests. Only in CC3 (28.7%) sample, the effect of increasing particle size was not observed in the results. Additionally, the distance index for all ternary mixtures increased compared to their corresponding binary mixtures, indicating decreased strength and durability.

Table 8. Distance index for different W_j

Mix. ID / W_j	W_1=W_2=0.5	W_1=1	W_2=1
OPC	0.38	0.00	0.76
FC3 (19.4%)	1.56	3.12	0.15
FLC3 (19.4%)	1.54	2.99	0.75
MC3 (19.4%)	1.58	3.10	0.58
MLC3 (19.4%)	1.80	3.50	0.81
CC3 (19.4%)	1.85	3.62	0.81
CLC3 (19.4%)	1.89	3.64	0.99
FC3 (28.7%)	0.99	1.94	0.37
FLC3 (28.7%)	1.53	2.99	0.62
MC3 (28.7%)	0.99	1.94	0.44
MLC3 (28.7%)	1.69	3.31	0.74
CC3 (28.7%)	0.86	1.68	0.34
CLC3 (28.7%)	1.66	3.21	0.86

According to the results in Fig 17a, with an increase in the importance of strength, FC3 (19.4%) preferred relative to OPC mixture. When the weight of the strength criterion is less (more important to the durability criterion), binary mixtures containing CC with a grade of 28.7% will be preferable. FC3 (19.4%) will have the lowest distance index when the weight is less than 0.1. In the distance of 0.1-0.3 (weight of strength), CC3 with a 28.7% grade will be better. According to the results, a significant difference in the distance index between binary and ternary mixtures is observed. This is due to the effect of adding LP in ternary blends of LC3 concrete. Due to considering two decision criteria, the interpretation of the results in Fig 17b is very similar to Fig 17a. Only when the importance of the durability criterion exceeds 0.7, the results obtained for samples containing CC with the lower grade will be the preferred option. However, it is necessary to mention that the environmental impact of using CC compared to Portland cement is considerable, and further studies need to consider other criteria such as environmental and economic assessments, including carbon footprint, energy consumption, and cost.



(a)

(b)

Fig 17. Distance index values according to different weighting; a) Strength criterion, b) Durability criterion

4. Conclusion

This study mainly focused on the effect of the fineness of low-grade calcined clays (CCs) on their reactivity in binary and ternary concretes. For this purpose, two kaolinitic low-grade CCs were ground by laboratory ball mill to achieve three fineness values of ~ 8 ± 2, 20 ± 2, and 32± 2 wt% retaining on 45 μm sieve named fine, moderate and coarse CCs, respectively. Experimental investigations of binders with the 30% substitution of the PC by CCs (C3 binary blends) or combined 20% CCs and 10% LP (in LC3 ternary blends) were compared with the control mixture without CCs and LS. It is worth noting that the experiments were conducted in a controlled laboratory environment, so the results may not completely represent how the C3 and LC3 concretes would perform in actual construction situations. The following conclusions can be drawn:

- Utilizing low-grade CCs (kaolinite contents < 30 wt%) in C3 and LC3 concretes resulted in lower compressive strength, higher water absorption, higher electrical resistivity, and lower chloride ions migration coefficient than PC concrete.
- Binary blends incorporating higher kaolinite content CC (28.7%) showed superior performance in comparison with corresponding LC3 concretes in terms of compressive strength, bulk water absorption, and electrical resistivity. However, in binary and ternary blends containing CC (19.4%), the difference between these results was insignificant.
- Regarding RCMT results, the C3 binary mixtures containing CC (19.4%) outperformed the counterpart ternary LC3 mixtures at 28 and 91 days. In binary and ternary blends containing CC (28.7%), LC3 mixtures had comparable D_{nssm} values at 28 days, but after 91 days of curing, C3 mixtures showed lower D_{nssm} relative to LC3 concretes.
- Regarding the chloride ions migration coefficient of mixtures with various fineness of CCs, results revealed that the chloride ions migration coefficient of mixtures with the coarser particles were 81-107% and 100-134% of their counterpart mixtures with the finest CC particles at 28 and 91 days, respectively.
- Based on MIP results, the blends made with the highest fineness of CCs demonstrated a lower total porosity and a considerably greater pore refinement compared to the blend made with the coarser CCs. This can be attributed to the filling effect and higher pozzolanic reaction of the highest fineness-containing mixtures.
- Substitution of finer low-grade CCs in C3 and LC3 mixtures had a marginal improvement in the compressive strength, bulk water absorption, and electrical resistivity by up to 4.7, 5.2, and 14.5% relative to coarser low-grade CCs mixtures, respectively. In other words, the mixtures with the CCs' fineness of ~32 ± 2 wt% retaining on 45 μm sieve performed a comparable compressive strength, bulk water absorption, and electrical resistivity with the counterpart mixtures containing CCs with higher fineness of 8 ± 2 wt% retaining on 45 μm sieve.
- Overall, C3 and LC3 concretes containing low-grade CCs with the fineness of ~32 ± 2 wt% retaining on 45 μm sieve obtained using less grinding time and energy consumption delivered a satisfactory mechanical and durability performance compared

to their corresponding CC mixtures with the fineness of ~8 ± 2 wt% retaining on 45 μm sieve.

- According to the results of the distance index, mixtures containing low-grade CCs will be the more suitable option compared to the control mixture, when the importance of durability exceeds approximately 70%. This percentage will change with changes in decision criteria and alterations in the grade of CCs.

References

Afzali-Naniz, O., Mazloom, M., & Karamloo, M. (2021). Effect of nano and micro SiO2 on brittleness and fracture parameters of self-compacting lightweight concrete. *Construction and Building Materials*, *299*, 124354. https://doi.org/10.1016/j.conbuildmat.2021.124354

Andres, L. M., Antoni, M. G., Alujas Diaz, A., Martirena Hernandez, J. F., & Scrivener, K. L. (2015). Effect of fineness in clinker-calcined clays-limestone cements. *Advances in Cement Research*, *27*(9), 546–556. https://doi.org/10.1680/jadcr.14.00095

Antoni, M., et al. (2012). Cement substitution by a combination of metakaolin and limestone. *Cement and Concrete Research*, *42*(12), 1579–1589. https://doi.org/10.1016/j.cemconres.2012.09.006

ASTM C150/C150M-20. (2020). Standard Specification for Portland Cement. *ASTM International, West Conshohocken, PA*. https://www.astm.org/c0150_c0150m-20.html

ASTM C1585-20. (2020). Standard Test Method for Measurement of Rate of Absorption of Water by Hydraulic-Cement Concretes. *ASTM International, West Conshohocken, PA*. https://www.astm.org/c1585-20.html

ASTM C1760-12. (2021). *Standard Test Method for Bulk Electrical Conductivity of Hardened Concrete*. *ASTM International, West Conshohocken, PA*. https://www.astm.org/c1760-12.html

ASTM C39/C39M-21. (2021). Standard Test Method for Compressive Strength of Cylindrical Concrete Specimens. *ASTM International, West Conshohocken, PA*. https://www.astm.org/c0039_c0039m-20.html

ASTM C494/C494M-17. (2020). *Standard Specification for Chemical Admixtures for Concrete*. *ASTM International, West Conshohocken, PA*. https://www.astm.org/c0494_c0494m-17.html

Avet, F., & Scrivener, K. (2018). Investigation of the calcined kaolinite content on the hydration of Limestone Calcined Clay Cement (LC3). *Cement and Concrete Research*, *107*, 124–135. https://doi.org/10.1016/j.cemconres.2018.02.016

Ayati, B., Newport, D., Wong, H., & Cheeseman, C. (2022). Low-carbon cements: Potential for low-grade calcined clays to form supplementary cementitious materials. *Cleaner Materials*, *5*, 100099. https://doi.org/10.1016/j.clema.2022.100099

Bahman-Zadeh, F., Ramezanianpour, A. A., & Zolfagharnasab, A. (2022). Effect of carbonation on chloride binding capacity of limestone calcined clay cement (LC3) and binary pastes. *Journal of Building Engineering*, *52*, 104447. https://doi.org/10.1016/j.jobe.2022.104447

Bahman-Zadeh, F., Zolfagharnasab, A., Pourebrahimi, M., Mirabrishami, M., & Ramezanianpour, A. A. (2023). Thermodynamic and experimental study on chloride binding of limestone containing concrete in sulfate-chloride solution. *Journal of Building Engineering*, *66*, 105940. https://doi.org/10.1016/j.jobe.2023.105940

Barata, M. S., & Angélica, R. S. (2011). Pozzolanic activity of kaolin wastes from kaolin mining industry from the Amazon region. *Matéria (Rio de Janeiro)*, *16*, 797–810. https://doi.org/10.1590/S1517-70762011000300007

Bishnoi, S., Maity, S., Kumar, M., Saxena, S. K., & Wali, S. K. (2018). Pilot scale production of limestone calcined clay cement. *Calcined Clays for Sustainable Concrete: Proceedings of the 2nd International Conference on Calcined Clays for Sustainable Concrete*, 69–74. https://doi.org/10.1007/978-94-024-1207-9_12

BS 1881-122. (2020). *Testing concrete Method for determination of water absorption*. https://standardsdevelopment.bsigroup.com/projects/2020-01169

Díaz, Y. C., Berriel, S. S., Heierli, U., Favier, A. R., Machado, I. R. S., Scrivener, K. L., Hernández, J. F. M., & Habert, G. (2017). Limestone calcined clay cement as a low-carbon solution to meet expanding cement demand in emerging economies. *Development Engineering*, *2*, 82–91. https://doi.org/10.1016/j.deveng.2017.06.001

Du, H., & Dai Pang, S. (2020). High-performance concrete incorporating calcined kaolin clay and limestone as cement substitute. *Construction and Building Materials*, *264*, 120152. https://doi.org/10.1016/j.conbuildmat.2020.120152

Dumani, N., & Mapiravana, J. (2020). Evaluation of Age Strengths of Metakaolin Blend Pastes with Varying Fineness of Grind. *Calcined Clays for Sustainable Concrete: Proceedings of the 3rd International Conference on Calcined Clays for Sustainable Concrete*, 339–348. https://doi.org/10.1007/978-981-15-2806-4_40

Fernandez, R., Martirena, F., & Scrivener, K. L. (2011). The origin of the pozzolanic activity of calcined clay minerals: A comparison between kaolinite, illite and montmorillonite. *Cement and Concrete Research*, *41*(1), 113–122. https://doi.org/10.1016/j.cemconres.2010.09.013

Ferreiro, S., Canut, M. M. C., Lund, J., & Herfort, D. (2019). Influence of fineness of raw clay and calcination temperature on the performance of calcined clay-limestone blended cements. *Applied Clay Science*, *169*, 81–90. https://doi.org/10.1016/j.clay.2018.12.021

Ferreiro, S., Herfort, D., & Damtoft, J. S. (2017). Effect of raw clay type, fineness, water-to-cement ratio and fly ash addition on workability and strength performance of calcined clay – Limestone Portland cements. *Cement and Concrete Research*, *101*, 1–12. https://doi.org/https://doi.org/10.1016/j.cemconres.2017.08.003

Gettu, R., Patel, A., Rathi, V., Prakasan, S., Basavaraj, A. S., Palaniappan, S., & Maity, S. (2019). Influence of supplementary cementitious materials on the sustainability parameters of cements and concretes in the Indian context. *Materials and Structures*, *52*, 1–11. https://doi.org/10.1617/s11527-019-1321-5

Kashi, A., Ramezanianpour, A. A., & Moodi, F. (2017). Durability evaluation of retrofitted corroded reinforced concrete columns with FRP sheets in marine environmental conditions. *Construction and Building Materials*, *151*, 520–533. https://doi.org/10.1016/j.conbuildmat.2017.06.137

Kluge, W., & Assmann, B. O. (2018). Grinding of Calcined Clays and Its Effects on Cement Properties. *Calcined Clays for Sustainable Concrete: Proceedings of the 2nd International Conference on Calcined Clays for Sustainable Concrete*, 244–248. https://doi.org/10.1007/978-94-024-1207-9_39

Krishnan, S., Emmanuel, A. C., Shah, V., Parashar, A., Mishra, G., Maity, S., & Bishnoi, S. (2018). Industrial production of limestone calcined clay cement: experience and insights. *Green Materials*, *7*(1), 15–27. https://doi.org/10.1680/jgrma.18.00003

Krishnan, S., Gopala Rao, D., & Bishnoi, S. (2020). Why Low-Grade Calcined Clays Are the Ideal for the Production of Limestone Calcined Clay Cement (LC 3). *Calcined Clays for Sustainable Concrete: Proceedings of the 3rd International Conference on Calcined Clays for Sustainable Concrete*, 125–130. https://doi.org/10.1007/978-981-15-2806-4_14

Lapeyre, J., Ma, H., & Kumar, A. (2019). Effect of particle size distribution of metakaolin on hydration kinetics of tricalcium silicate. *Journal of the American Ceramic Society*, *102*(10), 5976–5988. https://doi.org/10.1111/jace.16467

Li, R., Lei, L., & Plank, J. (2022). Impact of metakaolin content and fineness on the behavior of calcined clay blended cements admixed with HPEG PCE superplasticizer. *Cement and Concrete Composites*, *133*, 104654. https://doi.org/10.1016/j.cemconcomp.2022.104654

Luzu, B., Trauchessec, R., & Lecomte, A. (2022). Packing density of limestone calcined clay binder. *Powder Technology*, *408*, 117702. https://doi.org/10.1016/j.powtec.2022.117702

Malacarne, C. S., Longhi, M. A., Silva, M. R. C., Gonçalves, J. P., Rodríguez, E. D., & Kirchheim, A. P. (2021). Influence of low-grade materials as clinker substitute on the rheological behavior, hydration and mechanical performance of ternary cements. *Case Studies in Construction Materials*, *15*, e00776. https://doi.org/10.1016/j.cscm.2021.e00776

Maraghechi, H., Avet, F., Wong, H., Kamyab, H., & Scrivener, K. (2018). Performance of Limestone Calcined Clay Cement (LC 3) with various kaolinite contents with respect to chloride transport. *Materials and Structures*, *51*, 1–17. https://doi.org/10.1617/s11527-018-1255-3

Mazloom, M., Karimpanah, H., & Karamloo, M. (2020). Fracture behavior of monotype and hybrid fiber reinforced self-compacting concrete at different temperatures. *Advances in Concrete Construction*, *9*(4), 375–386.

https://doi.org/10.12989/acc.2020.9.4.375

Mazloom, M., & Salehi, H. (2018). The relationship between fracture toughness and compressive strength of self-compacting lightweight concrete. *IOP Conference Series: Materials Science and Engineering*, *431*(6), 62007. https://doi.org/10.1088/1757-899X/431/6/062007

NT Build 492. (1999). *Nord test method: Chloride Migration Coefficients from Non-Steady-State*. https://www.nordtest.info/wp/1999/11/21/nt-build-492/

Pérez, A., & Martirena-Hernandez, J. F. (2020). Influence of Limestone Content and PSD of Components on Properties of Clinker-Calcined Clay-Limestone Cements Produced by Intergrinding. *Proceedings of the International Conference of Sustainable Production and Use of Cement and Concrete: ICSPCC 2019*, 31–37. https://doi.org/10.1007/978-3-030-22034-1_4

Pillai, R. G., Gettu, R., Santhanam, M., Rengaraju, S., Dhandapani, Y., Rathnarajan, S., & Basavaraj, A. S. (2019). Service life and life cycle assessment of reinforced concrete systems with limestone calcined clay cement (LC3). *Cement and Concrete Research*, *118*, 111–119. https://doi.org/10.1016/j.cemconres.2018.11.019

Poon, C.-S., Lam, L., Kou, S. C., Wong, Y.-L., & Wong, R. (2001). Rate of pozzolanic reaction of metakaolin in high-performance cement pastes. *Cement and Concrete Research*, *31*(9), 1301–1306. https://doi.org/10.1016/S0008-8846(01)00581-6

R. Snellings, J. Chwast, ¨O. Cizer, N. De Belie, Y. Dhandapani, P. Durdzinski, J. Elsen, et al. (2018). RILEM TC-238 SCM recommendation on hydration stoppage by solvent exchange for the study of hydrate assemblages. *Mater. Struct.*, *51*(6), 1–4. https://doi.org/10.1617/s11527-018-1298-5

Ramezanianpour, A. A., Pilvar, A., Mahdikhani, M., & Moodi, F. (2011). Practical evaluation of relationship between concrete resistivity, water penetration, rapid chloride penetration and compressive strength. *Construction and Building Materials*, *25*(5), 2472–2479. https://doi.org/10.1016/j.conbuildmat.2010.11.069

Riahi Dehkordi, E., & GivKashi, M. R. (2024). Considerations for the Construction, Implementation and Economic Evaluation of Geopolymer Permanent Formworks (GPFs): A New Approach to Protect Concrete Structures Against Aggressive Environmental Factors. *Arabian Journal for Science and Engineering*, *49*(4), 4861–4875. https://doi.org/10.1007/s13369-023-08081-4

Richardson, M. G. (2002). *Fundamentals of durable reinforced concrete*. CRC Press. https://taylorfrancis.com

Scrivener, K., Martirena, F., Bishnoi, S., & Maity, S. (2018). Calcined clay limestone cements (LC3). *Cement and Concrete Research*, *114*, 49–56. https://doi.org/10.1016/j.cemconres.2017.08.017

Scrivener, K., Snellings, R., & Lothenbach, B. (2018). *A practical guide to microstructural analysis of cementitious materials*. Crc Press. https://api.taylorfrancis.com

Sharma, M., Bishnoi, S., Martirena, F., & Scrivener, K. (2021). Limestone calcined clay cement and concrete: A state-of-the-art review. *Cement and Concrete Research*, *149*, 106564. https://doi.org/10.1016/j.cemconres.2021.106564

Siline, M., & Mehsas, B. (2022). Effect of increasing the Blaine fineness of Metakaolin on its chemical reactivity. *Journal of Building Engineering*, *56*, 104778. https://doi.org/10.1016/j.jobe.2022.104778

Yaraghi, A. H. Y., Ramezanianpour, A. M., Ramezanianpour, A. A., Bahman-Zadeh, F., & Zolfagharnasab, A. (2022). Evaluation of test procedures for durability and permeability assessment of concretes containing calcined clay. *Journal of Building Engineering*, *58*, 105016. https://doi.org/10.1016/j.jobe.2022.105016

Zolfagharnasab, A., Ramezanianpour, A. A., & Bahman-Zadeh, F. (2021). Investigating the potential of low-grade calcined clays to produce durable LC3 binders against chloride ions attack. *Construction and Building Materials*, *303*, 124541. https://doi.org/10.1016/j.conbuildmat.2021.124541

Zunino, F., & Scrivener, K. (2020). Increasing the kaolinite content of raw clays using particle classification techniques for use as supplementary cementitious materials. *Construction and Building Materials*, *244*, 118335. https://doi.org/10.1016/j.conbuildmat.2020.118335