LEAN-CLINKER MORTARS WITH RECYCLED CEMENT TOWARDS THE PRODUCTION OF LOW-CARBON CONCRETE FOR MODULAR CONSTRUCTION

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Abstract

The construction sector is a major contributor to CO_2 emissions, largely due to clinker-intensive cement. This study explores low-carbon cementitious matrices by incorporating recycled cement (RC), calcined clays (CC), and limestone filler (LF), particularly in LC3 formulations, to develop eco-efficient modular concrete. An extensive experimental program assessed fresh-state properties, mechanical performance, and durability. Results demonstrated that LC3 + RC mixtures achieved up to 80% clinker reduction while maintaining strength and enhancing durability, proving their potential for sustainable construction.

1 Introduction

The construction industry is a significant contributor to global carbon emissions, primarily due to the reliance on conventional Portland cement. Numerous studies have highlighted the environmental costs of cement production and its significant role in global greenhouse gas emissions [1, 2]. In addition, the rapid population growth and prevailing social inequalities have created an urgent and pressing demand for adequate housing in contemporary society [3]. Innovative solutions are critical in addressing these challenges, particularly through the development of eco-efficient alternatives to traditional construction materials.

Cement production accounts for approximately 8% of global CO_2 emissions, largely driven by energy-intensive calcination processes and limestone decomposition [4, 5]. With increasing regulatory pressure and societal demand for sustainable practices, the construction sector is shifting towards greener alternatives. Materials that utilize industrial byproducts, recycled resources, and alternative binders can significantly reduce emissions, while conserving natural resources. This paradigm shift aligns with the broader goals of circular economy principles and climate change mitigation efforts.

Modular construction offers a promising pathway toward sustainability in the built environment. Studies have shown its capacity to reduce construction waste and improve energy efficiency [1, 3]. This approach also provides an opportunity to integrate innovative low-carbon materials into modular components, enhancing their overall environmental performance. Modular construction's scalability and adaptability further strengthen its potential as a cornerstone of sustainable urban development [6].

Recent studies have explored various strategies to reduce the carbon footprint of concrete. These include the utilization of recycled cement, calcined clays, and bottom ash as supplementary cementitious materials. Research has demonstrated that these alternatives can partially replace clinker in cement, which is responsible for the majority of its CO_2 emissions [2, 6, 7], without compromising performance. For instance, thermoactivated recycled cement, derived from waste concrete, has shown promising mechanical and durability properties in concrete, making it a viable eco-efficient material [8, 9]. Similarly, limestone calcined clay cements (LC3) leverage the synergy between calcined clay

and limestone to achieve high performance with significantly lower clinker content [9, 10]. Additionally, LC3 technology is able to offer a reduction in CO_2 emissions of up to 30% compared to conventional Portland cement [11]. Savings of over 60% in CO_2 emissions are reported for recycled cement [12].

Innovations in material science are paving the way for more sustainable construction. Bottom ash, a byproduct of coal combustion, has been utilized as a low-cost aggregate or pozzolan in concrete mixtures, reducing reliance on natural resources [13]. As mentioned, recycled cement, produced through thermal activation of construction waste, provides another avenue for incorporating circular economy principles into concrete production [9, 14 - 16]. These advancements reflect the growing potential of alternative binders to revolutionize concrete manufacturing. In fact, several alternative binders are gaining attention for their ability to replace traditional clinker.

The main challenge in modular construction is related with the fact that low-carbon concretes involve lower binder contents and high supplementary cementitious materials dosages, which is against the requirement of fluid concretes with high early strength. The integration of low-carbon concrete into modular construction represents a transformative step toward sustainable infrastructure. This study makes part of a research project exploring the synergy between various eco-efficient materials, such as recycled cement, calcined clays, and bottom ash, to achieve low-carbon binders of reduced clinker factors, as low as less than 20%, without significantly affect their mechanical and durability performance. In this case, the combination of these materials was analysed for the first time, considering the production and mechanical and durability characterization of mortars. Modular construction's efficiency and compatibility with these materials further amplify its potential as a solution for eco-efficient development. This convergence of innovative materials and methods underscores the urgency and feasibility of transitioning to a greener built environment.

2 Experimental Program

2.1 Materials

For this study, CEM I 52.5R (PC), recycled cement (RC), calcined clay (CC), bottom ash (BA), fly ash (FA), commercial limestone filer (LF), were selected to develop and produce a new generation of truly low-carbon binders. These binders were incorporated in mortars also produced with natural silicious sand, which properties are presented in Table 1. A polycarboxylate-based superplasticizer was also used to adjust fresh mortars to the same workability range.

Table 1 Natural aggregates properties.

Parameter	Standard	Coarse Sand	Fine Sand
Oven-dried particle density (kg/m3)	EN 1097-6 [17]	2599	2598
Water absorption at 24 h (%)	EN 1097-6 [17]	0.26	0.15
Loose bulk density (kg/m3)	EN 1097-3 [18]	1594	1507

Recycled cement production started with the preparation of original paste (OP), which was produced with a water-to-binder (w/b) ratio of 0.55, yielding an average compressive strength of approximately 35 MPa after 28 days. Following production, the OP underwent curing in a controlled environment with 95% relative humidity for seven days. Subsequently, the material was stored under standard laboratory conditions for over 90 days to ensure stabilization. After this time, OP was subjected to a series of mechanical processes, to reduce the materials particle size. Initially, the OP was broken into fragments roughly 7 cm in size using jaw crushers with variable openings. These fragments were then processed through a roller mill, producing particles smaller than 2 mm. The material was further refined in a horizontal steel ball mill for approximately two hours, resulting in fine particles that were then sieved through a 250 µm mesh to ensure uniformity. The thermal activation of this finely milled waste cement paste was carried out in a horizontal furnace. This procedure was based in previous works, where it was observed an effective dehydration of the OP particles without significant decarbonization [3, 4]. The process involved a controlled heating regimen, beginning with a temperature increase of 10°C per minute up to 150°C, where the material was held for one hour. This was followed

by further heating at the same rate until the target activation temperature of 650°C, where it remained for three hours to ensure complete activation. Once the heating cycle is complete, the binder is cooled gradually within the furnace to room temperature, preventing thermal shock and ensuring material stability. Table 2 presents the main properties of the utilized binders. The CC had a metakaolin content of 56%.

Table 2 Cement properties.

Parameter		Standard	PC	RC	CC	BA	LF	FA
Absolute density (g/cm^3)		-	3.10	3.00	2.65	2.23	2.61	2.3
BET surface area (m^2/g)		-	2.08	14.29	15.71	7.08	2.13	1.88
Activity index at 28 days (%)		EN 196-1 [20]	-	-	105	69.1	-	74.9
Activity index at 90 days (%)			-	-	-	84.1	-	-
Insoluble residue (%)		EN 196-2 [21]	1.45	<0.52	-	-	-	84.92
Loss on ignition (950 ◦C) (%)			2.55	8.41	-	5.34	-	2.69
SiO_2+Al_2O_3+Fe_2O_3 (%)			20.1+4.9 + 2.80	19.1+5.1 + 3.00	59.3+33. 5+ 3.22	59.4+20. 1+ 7.0	0.1+0.1+ 0.04	56.5+ 22.7+ 7
CaO+MgO (%)			62.9+1.7	60.8+1.8	0.1+0.2	1.9+1.7	55.4+0.3	2.9+1.6
Free CaO (%)		EN 451-1 [22]	-	13.94	-	-	98.9	-
Setting time (min)	Initial	EN 196-3 [23]	118	290	-	-	-	-
	Final		162	385	-	-	-	-

2.2 Composition and production of mortars

In this study, mortars were prepared with a cement-to-sand weight ratio of 1:3, following the guidelines outlined in EN 196-3 [23]. Fine and coarse sands were utilized with a 35:65 weight ratio. All mortars were produced using the same w/b ratio (0,45), with variations only in the superplasticizer dosage to achieve target flow values, according to EN 1015-3 [24], in the range of about 160±10 mm. Table 3 presents the compositions of the mixtures produced in the study. The equivalent w/c ratio is also indicated, considering only the content of cement used.

Approximately 27 different mixtures were developed, encompassing mortars with various combinations of cementitious matrices. These combinations were designed to assess the individual and combined contributions of each component. The selection of mixtures was based on experimental trials and previous studies [8, 10, 11, 14, 24, 25]. Thus, binary mixtures with different contents of LF (5, 10, 15, 20%), RC (20, 30, 40%), CC (10, 20, 30, 40, 45, 60%), and BA (20, 30, 40, 60%) were produced to evaluate the influence of each addition. Reference mortars with only PC (100% PC) and with 60% PC and 40% FA, were produced for comparison purposes. Mixtures with higher contents of CC and/or RC had increased water demand, necessitating higher superplasticizer dosages.

The simultaneous incorporation of CC and LF followed the philosophy developed in LC3 [11, 25, 27]. According to the literature, the preferred ratio between LF and CC should be approximately 1:2, anticipating clinker reductions of around 50%. In this study, solutions ranging from 1:2 to 1:3 and clinker replacements from 45% to 60% were considered. As the main innovation of this work, ternary and quaternary mortars were developed with an additional substitution of PC by RC, allowing for significantly higher reductions in clinker content, aiming for truly more sustainable mixtures. The objective is to achieve low-carbon solutions without excessively compromising the beneficial properties of clinker and the efficiency of pozzolanic additions, which require minimum dosages of the former to activate and develop hydration products similar to those of cement. Notably, "green" mortars were also produced, associated with 0% clinker content and 100% use of mineral additions and recycled products.

Table 3 Produced mortars composition (% by weight of binder).

Designation	w/c $_{equ}$	PC (%)	CC (%)	LF (%)	RC (%)	BA (%)	FA (%)
PC	0.45	100%	0%	0%	0%	0%	0%
5LF	0.47	95%	0%	5%	0%	0%	0%
10LF	0.50	90%	0%	10%	0%	0%	0%
15LF	0.53	85%	0%	15%	0%	0%	0%
20LF	0.56	80%	0%	20%	0%	0%	0%
20RC	0.56	80%	0%	0%	20%	0%	0%
30RC	0.64	70%	0%	0%	30%	0%	0%
40RC	0.75	60%	0%	0%	40%	0%	0%
10CC	0.50	90%	10%	0%	0%	0%	0%
20CC	0.56	80%	20%	0%	0%	0%	0%
30CC	0.64	70%	30%	0%	0%	0%	0%
40CC	0.75	60%	40%	0%	0%	0%	0%
45CC	0.82	55%	45%	0%	0%	0%	0%
60CC	1.13	40%	60%	0%	0%	0%	0%
30CC10LF	0.75	60%	30%	10%	0%	0%	0%
30CC15LF	0.82	55%	30%	15%	0%	0%	0%
40CC20LF	1.13	40%	40%	20%	0%	0%	0%
30CC10RC15LF	1.00	45%	30%	15%	10%	0%	0%
30CC20RC15LF	1.29	35%	30%	15%	20%	0%	0%
40CC20RC20LF	2.25	20%	40%	20%	20%	0%	0%
30CC40RC15LF	3.00	15%	30%	15%	40%	0%	0%
40CC40RC20LF	-	0%	40%	20%	40%	0%	0%
40FA	0.75	60%	0%	0%	0%	0%	40%
20BA	0.56	80%	0%	0%	0%	20%	0%
30BA	0.64	70%	0%	0%	0%	30%	0%
40BA	0.75	60%	0%	0%	0%	40%	0%
60BA	1.13	40%	0%	0%	0%	40%	0%

2.3 Test Methods

The mortars characterisation was divided in two phases: fresh state and hardened state. For the mortar's characterisation in the fresh state, tests of fresh density and flowability were performed. Flowability was accessed according to EN 1015-3 [24]. The fresh density was directly accessed in the moulds to guarantee the same compaction conditions. The mechanical characterisation of the mortars, by means of flexural and compressive strength tests, was performed in 160×40×40 mm specimens, according to EN 196-1 [20]. After 24 hours, the specimens were demoulded and cured in a wet chamber (95% RH) until testing age, namely 1, 3, 7 and 28 days. The capillary absorption was tested according to EN 1015-18 [28]. Three sawn specimens of 80×40×40 mm were wet cured for 7 days followed by 21 days at 65±5% relative humidity (RH) and then oven-drying at 60ºC until constant weight. The sawn surfaces of each specimen were immersed in a water layer of 5 ± 1 mm. The mass of the specimen was measured at 10, 20, and 30 minutes, and subsequently at 1, 2, 6, 24, 48 and 72 hours after the initial water contact. The capillary absorption coefficient (C_{abs}) was determined as the slope of the linear regression of the absorption data between $\sqrt{20}$ minutes and $\sqrt{6}$ hours.

Carbonation resistance was measured according to LNEC E 391 [29], using 80×40×40 mm specimens. Specimens were wet cured until 7 days, followed by 21 days at 65±5% RH. At 28 days, the top and bottom surfaces of each specimen were sealed with an epoxy-based paint to ensure that carbonation occurred exclusively through radial CO_2 diffusion. The specimens were then placed in an accelerated carbonation chamber, at 23 ± 3°C, 65 ± 5% RH, and a CO_2 concentration of 5 ± 0.1%. At 7, 28, 56, and 91 days, one specimen from each composition was removed from the chamber, divided into four parts, and the fractured surfaces were sprayed with a 0.2% phenolphthalein solution. The carbonation depth was determined as the average length of the uncoloured region, where the pH was below 9. Assuming stable test conditions over time, the carbonation coefficient (K_{ca}) was calculated as the slope of the linear regression of carbonation depth between 0 and $\sqrt{90}$ days.

The chloride penetration resistance was tested by the Rapid Chloride Migration Test (RCMT), according to NT Build 492 [30]. Three samples of Ø100×50 mm, sawn from Ø100×250 mm cylinders, were tested per mixture. The specimens were cured in the wet chamber until 7 days and then at 65 ± 5% RH until testing, at 28 and 90 days. Before testing, the specimens were vacuum saturated in a calcium hydroxide solution for 24 hours. During the test, an external electrical potential (U, V) was applied for a specified duration (t, hours), driving chloride ions from the anodic solution into a specimen of thickness L (mm). Following the test, the specimen was split into two halves, and a 0.1 M silver nitrate solution was sprayed onto the fractured surfaces to determine the chloride penetration depth (X_d (mm)). Finally, the chloride migration coefficient ($Dcl_{RCMT} \times 10^{-12}$ m²/s) was calculated according to NT Build 492 [30].

3 Results and discussion

The average results of main properties analysed in this study are presented in Table 4. The study of the evolution of strength over time and durability is based on just a few selected mixtures capable of reflecting the influence of each of the additions studied.

3.1 Compressive strength

The mechanical strength was not significantly affected up to 10% LF, and then decreased proportionally with LF content as a result of clinker dilution, as also reported by Matschei [31].

Recycled cement demonstrated excellent hydration capacity, allowing replacements of up to 40% without significant changes in compressive strength. The relative strength compared to PC varied with the incorporation level, showing a slight reduction for 20% RC (-3%) and 40% RC (-8%), while an increase was observed for 30% RC (+7%). In fact, only the mortar with 30% RC exhibited a void content similar to PC mortar (Table 4). In this case, 30% RC mortar also benefited from the use of high SP content, which facilitated RC dispersion and subsequent hydration. Conversely, the 40% RC mortar proved high-water demand, making effective compaction more difficult. Nevertheless, the suitability of RC as a direct substitute for PC is confirmed, as observed in other studies [8, 32].

Replacing PC with up to 30% CC led to strength increases of up to 5%, in line with the activity index above 1.0 shown in Table 2. This confirms the high pozzolanic reactivity of the CC used in this study, with performance comparable to cement, at least after 28 days. For replacement levels above 40%, results were affected by clinker dilution. However, the strength reduction for 40% CC (-7%) and especially for 45% CC (-19%) was unexpectedly high, given that at 60% CC the strength was similar to that of the 45% CC mortar. One reason for this reduction is likely related to the lower compactness of 45% CC mixture, where the void content exceeded 7%.

Among the non-inert additions, FA and BA were the least reactive, leading to the greatest strength reductions. Its low initial reactivity explains the low activity indices. In this study, the strength losses were lower than those suggested by the activity indices, with reductions of 20%, 30%, and 53% for 30%, 40%, and 60% PC replacement by BA, respectively. A reason for this apparently better performance was the higher compactness achieved in these mortars, which presented lower void content. As suggested by the differences in activity index, the reactivity of FA was slightly higher than that of BA, resulting in only 25% reduction for 40% FA.

Table 4 Produced mortars composition (% by weight of binder).

Designation	SP (%)	Spread (mm)	Fresh Density	Volume of Voids	Mechanical Strength								Durability			
			ρ fresh (kg/m^3)	Vv (%)	$f_{c,1d}$ (MPa)	$f_{ctm,1d}$ (MPa)	$f_{c,3d}$ (MPa)	$f_{ctm,3d}$ (MPa)	$f_{c,7d}$ (MPa)	$f_{ctm,7d}$ (MPa)	$f_{c,28d}$ (MPa)	$f_{ctm,28d}$ (MPa)	C_{abs} ($mm/min^{0.5}$)	K_{ca} ($mm/year^{0,5}$)	Dcl_{RCMT_28d} ($x10^{-12} m^2/s$)	Dcl_{RCMT_90d} ($x10^{-12} m^2/s$)
PC	0.1	150.7	2220	3.9	32.5	5.12	47.1	6.2	47.4	6.5	59.1	7.4	175.4	17.10	11.0	10.7
PC5LF	0.4	176.7	2205	4.3							59.5	7.4				
PC10LF	0.4	158.0	2204	4.2							59.6	7.6				
PC15LF	0.2	155.3	2183	5.0							53.2	7.2	206.6	17.70	12.9	
PC20LF	0.2	156.7	2189	4.6							51,8	6.6				
20RC	1.4	167.7	2168	5.7							57.2	7.3	94.8	15.30	11.4	11.0
30RC	1.6	154.3	2197	4.3							63.2	7.5				
40RC	2.6	160.0	2123	7.2	27.4	4.90	44.3	6.4	42.9	6.2	54.5	6.7	35.7	15.60	10.3	9.2
10CC	0.5	157.3	2196	4.5							61.4	7.4				
20CC	1	148.7	2192	4.3	22.7	4.52	35.8	5.4			62.0	7.7	165.7	18.00	3.4	2.8
30CC	1	146.7	2181	4.5							61.9	7.0				
40CC	1.5	154.3	2148	5.6							54.8	6.3	120.5	39.70	3.0	1.7
45CC	1.9	180.0	2102	7.4							48.2	6.4	124.6	58.90	4.7	
60CC	2.6	161.3	2156	4.4							47,0	6.0	183.5	106.60	4.7	4.3
30CC10LF	0.9	157.7	2178	4.4							60.2	8.1				
30CC15LF	0.9	177.0	2159	5.1							55.3	8.1	121.3	62.28	3.2	2.1
40CC20LF	0.7	149.7	2152	5.0							52.1	8.1	182.4	74.13		1.5
30CC10RC15LF	1.4	148.3	2136	5.9							54.2	7.4				
30CC20RC15LF	2.1	167.0	2154	4.9							54.8	8.4	127.8	68.59		1.0
40CC20RC20LF	3.1	165.3	2122	5.7							40.6	6.9	140.2	93.61		1.2
30CC40RC15LF	3.7	174.0	2132	5.4	4.4	1.18	27.9	4.3	46.1	6.3	52.5	6.7	101.3	55.47	1.0	0.8
40CC40RC20LF	3.8	158.3	2105	6.1							40.4	6.0	278.7	81.81	2.7	1.4
40FA	0.2	189.0	2184	3.2							44.5	6.3	280.8	55.58	11.2	7.2
20BA	0.1	148.0	2215	2.9							54.8	7.2	174.6	20.78	10.1	7.4
30BA	0.2	170.7	2196	3.0							47.5	6.6				
40BA	0.2	160.3	2180	3.1	15.2	3.52	26.1	4.4	33.2	4.5	41.3	6.5	280.8	51.25	11.6	11.5
60BA	0	159.0	2161	2.8							27.7	4.4	499.0	109.00	17.0	10.7

These mortars were produced with a low clinker factor, for addition contents of 40 to 100%. LC3 mixtures, replacing PC with 30% CC and 10% to 15% LF resulted in similar (+2%) to slightly lower (-7%) strengths compared to the reference mortar with 100% PC. In this study, the best results were obtained for 30% CC and 10% LF (3:1), although the differences were small. The strengths were similar to those of mortars containing only 10–15% LF or 30% CC, indicating that the synergy between both additions allowed for a reduction in the clinker factor without compromising mechanical performance. When the CC and LF content was increased to 40% and 20%, respectively, while maintaining the same 2:1 ratio, an additional 6% reduction in strength occurred. Nevertheless, compared to the reference mortar, only a 12% reduction in strength was observed, while achieving a 60% reduction in cement content.

To further develop more sustainable mortars with lower clinker factors, PC was replaced with 10% to 40% RC in LC3 mixtures. In the 30CC15LF mortar, replacing up to 20% of PC with RC did not affect compressive strength. Only at 40% RC a 5% strength reduction was observed, likely due to the increased difficulty in compacting this mortar (Table 4). These results are highly promising, as it was possible to achieve solutions with only 35% and 15% cement, for strength reductions of just 7% and 11%, respectively, compared to PC mortar. In fact, based on these results, it can be concluded that incorporating RC into LC3 does not negatively affect strength. Assuming that the mechanical performance of LC3 is comparable to that of PC mixtures, it is possible to produce mortars with over 80% clinker reduction, while maintaining mechanical performance.

The same approach was applied to mixtures with 40% CC and 20% LF, revealing that replacing PC with 20–40% RC led to an additional 22% reduction in strength. This reduction was identical for both 20% and 40% RC. In this case, a smaller strength reduction would have been expected, in line with what was observed for 30CC15LF series. This discrepancy may be related to the greater difficulty in dispersing the binder, as well as challenges in homogenization and compaction of these mixtures, which contain high levels of CC and RC, both with high water demand.

Nevertheless, it is noteworthy that mortars achieving strengths above 40 MPa (for 32% reduction in strength), were produced with total absence of clinker. This represents a significant step forward to the production of truly green concretes, with substantial CO_2 reductions. Even disregarding the fact that both RC and CC require less thermal energy during production (about half the processing temperature), the non-decarbonation of these raw materials results in more than a 60% reduction in CO_2 emissions compared to cement [12, 26]. Under these conditions, clinker-free mixtures with 20% LF, 40% CC, and 40% RC can lead to an approximately 70% reduction in CO_2 emissions.

As previously mentioned, it is confirmed that mixtures containing BA exhibit a slower strength development due to their lower pozzolanic reactivity at early ages. Only at long term strength is expected to approach that of reference mortar. On the other hand, calcined clay showed a faster strength development, particularly after 1–3 days of age [25, 33]. In fact, the strength ratio at 3 days compared to 28 days was identical in both the reference mortar and that with 40% CC.

As reported in previous studies [34, 35], the mortar with 40% RC exhibited slower strength development up to 1 day of age, accelerating afterward until 3 days. Finally, the 30CC40RC15LF mixture showed slower strength development up to 7 days due to the delayed reactions occurring in CC and RC. This is particularly evident at 1 day of age, when CC and RC are just beginning their hydration reactions.

3.2 Flexural Strength

In general, the same trends observed for compressive strength are confirmed, particularly regarding the reduction in strength for PC replacement levels above 10% LF, 30% CC, and the incorporation of FA and BA. Compared to compressive strength, the differences between compositions were smaller, which aligns with the fact that tensile strength varies approximately with the square root of compressive strength. The strength loss in mortars with 40% BA and FA was lower, remaining below 15% and 17%, respectively, compared to the reference mortar with PC. In the case of CC replacement, even for 60% incorporation, the strength loss was less than 19%. The anomalous results obtained for 40–45% CC are confirmed. On the other hand, only for 40% PC replaced with RC, there was a slight reduction in strength (10%). The high reactivity of RC is confirmed, with even a 2% increase in strength for 30% RC.

For ternary and quaternary mixtures, the results were generally consistent with those observed for compressive strength. Notably, LC3 mixtures performed better than the PC mortar, both for 30% and

40% CC. The additional incorporation of up to 20% RC had little impact on strength, always ensuring values higher than those of the reference mortar. This means that a 65% reduction in cement content was achieved without negatively affecting the mechanical performance of mortars. As found in compressive strength, the simultaneous incorporation of RC, 40% CC, and 20% LF resulted in slightly lower performance. Nevertheless, it was possible to produce mixtures with only 15% PC or even without PC, with strength reductions of just 9% and 19%, respectively, compared to reference PC mortar. These results, along with the compressive strength findings, are highly promising as they validate the production of fully cement-free structural solutions.

3.3 Capillary Absorption

Capillary absorption is mainly affected by the effective water-to-binder ratio and the quantity, distribution, and connectivity of the pores.

The replacement of PC with 15% LF led to an expected increase in the absorption coefficient (18%), as it resulted in the dilution of the clinker content and the volume of hydration products formed. Replacing PC with RC progressively reduced capillary absorption by 46% and 80% for 20% and 40% RC, respectively. A similar trend was observed by Carriço [32] in mortars with a water-to-binder ratio of 0.6 and up to 50% RC. This phenomenon is explained by the fact that the RC matrix is associated with a more refined microstructure, consisting of a higher volume of smaller pores and, consequently, lower capillary absorption [35]. In other words, although the total porosity remains similar, it is distributed among a greater number of fine pores [34].

As observed in the previous chapter, after 3–7 days, CC exhibits high reactivity, enabling the development of additional hydration products. This leads to the formation of more refined structures associated with pores of greater tortuosity and lower connectivity [36]. A progressive decrease in water-accessible porosity and capillary absorption rate is confirmed for PC replacement levels up to 45% CC. However, at 60% CC, the absorption rate reverses its trend, reaching values 5% higher than that of the PC mortar. This confirms the limitation of additional hydration development for replacement rates above approximately 40%, as previously discussed. In this case, there is an excess of CC that cannot be satisfied by the deficient amount of PC, resulting from the dilution of clinker content. The greatest reduction in capillary absorption (31%) was achieved with 40% CC.

In LC3 mixtures, both with and without recycled cement, a general and expected reduction in the absorption rate was observed, especially when combining 30% CC and 15% LF. Despite the lower PC and CC content, the absorption coefficient of the 30CC15LF mixture was similar to that of 40CC and PC15LF, suggesting a synergistic effect of these additions. Additionally, replacing PC with RC (30CC20RC15LF and 30CC40RC15LF) maintained or slightly improved performance, increasing by 5% for 20% RC and decreasing by 16% for 40% RC. Once again, RC demonstrated its ability to further refine the microstructure. It is important to note that CC and RC introduce different types of refinement: CC promotes refinement by forming additional hydration products within the existing matrix, whereas RC refines the microstructure by initially reducing the interparticle space [35].

Overall, the 30CC40RC15LF mixture exhibited the best performance, reducing the absorption rate by 42% compared to the reference mortar with PC, while using only 15% cement.

As observed for compressive strength, mortars with 40% CC and 20% LF showed poorer performance. The excessive dilution of clinker content, with too high CC/clinker ratio, as well as the lower compaction achieved in the fresh state can explain this reduction in performance, particularly in the 40CC40RC20LF mixture. Nevertheless, it is noteworthy that this mixture, despite having 59% higher capillary absorption, was produced without PC.

Regarding the use of BA, capillary absorption increased with its incorporation, especially at replacement levels above 20%. This indicates that, at the test age (approximately 45 days), BA had not yet developed sufficient reactivity to compensate for high clinker dilution, resulting in increased capillary absorption. In this case, the absorption coefficient was 1.6 to 2.4 times higher than that of the reference mortar with PC (Table 4). Specifically, it is confirmed that the reactivity of 60BA is compromised by the low clinker content in the system.

3.4 Chloride migration

The diffusion coefficient at 28 and 90 days ranged from 1.0 to 12.9 × 10^{-12} m²/s and 0.8 to 11.0 × 10^{-12} m²/s, respectively. These values classify the concretes within the range of moderate (10–15 × 10^{-12} m²/s) to extremely high resistance (<2.5 × 10^{-12} m²/s) to chloride penetration, according to the classification by Gjorv [37]. The Dcl_{RCMT} at 28 days for the reference mortar with PC was 11 × 10^{-12} m²/s,

consistent with values reported by Bogas [38] and fib 34 [39] for concretes with CEM I and a w/c ratio of 0.45 (10.7 and 11.2 × 10^{-12} m²/s, respectively). The partial replacement of PC with 15% LF increased the $D_{cl,RCMT}$ by 17%, in line with the absorption results.

The partial replacement of PC with up to 40% RC had a minimal impact on Dcl_{RCMT}. At 28 days, the diffusion coefficient increased by 3% for 20% RC and decreased by 7% for 40% RC. This can be explained by the fact that, unlike in the absorption test, very fine pores can contribute to the migration test. Since the RC matrix has similar porosity, but more refined, absorption decreases, whereas diffusion, which mainly depends on the volume and interconnectivity of pores rather than their size [40, 41], remains little changed. The minor variations observed were mainly due to differences in the compaction achieved after casting and issues with binder dispersion in drier mixtures.

The incorporation of up to 60% CC significantly reduced the diffusion coefficient, placing the mortars in the category of "high resistance to chloride penetration" (<5 × 10^{-12} m²/s), according to Gjorv [37]. The greatest reduction was achieved for 40% CC, with Dcl_{RCMT} at 28 and 90 days being 77% and 84% lower, respectively, compared to the PC mortar. The importance of CC in microstructure refinement and durability enhancement is emphasized by Sabir [36] and Maraghechi [42].

On the other hand, the partial replacement of PC with FA and BA had a limited impact at 28 days but reduced Dcl_{RCMT} by up to 33% at 90 days. This is due to the lower pozzolanic activity index of BA and FA at 28 days, leading to a more significant long-term contribution. This is even more relevant given that mortars were cured for only 7 days in water. Therefore, for pozzolanic additions with slow reactions, it is more appropriate to conduct diffusion tests at 90 days. Nonetheless, even at 28 days, the action of BA and FA compensated for the increased porosity of these mortars, which was associated with lower mechanical strength and higher absorption at early ages. As per CC, these results are not only attributed to pore refinement, but also the composition of pore solution.

LC3 mixtures exhibited the best performance, benefiting from the synergy between CC and LF in the additional and phased formation of hydration products, particularly carboaluminates and hemicarboaluminates [43]. Compared to the PC reference mortar, ternary mixtures with 30–40% CC and 15–20% LF achieved reductions of 81–87% at 90 days, showing a similar performance to the mortar with 40% CC. These mortars are classified as having "extremely high resistance to chloride penetration" (<2.5 × 10^{-12} m²/s), according to Gjorv [37]. As noted, the refined microstructure of LC3 mixtures leads to lower permeability and increased durability, reducing chloride penetration and increasing concrete resistivity [25, 44], besides the change in pore solution composition.

Finally, replacing PC with up to 40% RC further reduced diffusion properties in the LC3 mixture. In this case, diffusion coefficients were only 1.0 × 10^{-12} m²/s, one order of magnitude lower than in the reference mortar. The Dcl_{RCMT} varied insignificantly for CC between 30–40% and RC between 20–40%. Importantly, high-durability solutions against chloride penetration were achieved without clinker incorporation. This study confirms that recycled cement is a viable substitute of conventional cement, at least up to 40% incorporation, yielding similar mechanical strength and durability.

3.5 Carbonation

Depending on the type of binder mixture, the carbonation coefficient (K_c) varied significantly between 15.3 and 109 mm/year$^{0.5}$. It is possible to observe that the progression of carbonation is approximately proportional to the square root of time, which is consistent with the evolution of this mechanism in cementitious mixtures, according to Fick's first law, provided they are not significantly affected by moisture distribution [45].

As expected, the PC mortar exhibited one of the lowest K_c values (17.1 mm/year$^{0.5}$), due to its low w/c and high reserve of carbonatable material. The value obtained is similar to that reported in fib 34 [39], which indicates 17.2 mm/year$^{0.5}$ for Type I cement with w/c of 0.45. The incorporation of 15% LF led to a natural increase in K_c, as it caused a slight increase in porosity and dilution of the amount of carbonatable material. An important result was observed in the partial replacement of PC by up to 40% RC, where a slight reduction (~10%) in the carbonation rate was noted.

In mortars with CC, a significant increase in the carbonation rate was observed as the proportion of PC replaced by CC increased. The carbonation coefficient increased by 2.3 and 6.2 times for 40% and 60% CC, respectively, which limits the excessive use of this addition in certain applications. However, for replacements up to 20%, the carbonation rate increased by only 5%. As previously mentioned, the 45% CC mortar was also affected by a reduction in its compactness after production.

The addition of BA and FA resulted in a similar effect to CC, although achieving even higher carbonation rates. This is mainly due to their lower ability to refine the microstructure, allowing greater CO_2 diffusion. The increase in carbonation in concretes with fly ash has been widely reported in the literature [45, 46]. Compared to the PC reference mortar, the carbonation rate increased by 1.2, 3.0, and 6.4 times for 20%, 40%, and 60% BA, respectively. The FA mortar performed similarly to that with BA, for the same replacement level.

The LC3 mixtures (30CC15LF and 40CC20LF) exhibited reduced carbonation resistance, reaching carbonation rates approximately 3.6–4.3 times higher than PC mortar. The carbonation rate for 30CC15LF was even higher than that of 40CC, despite a similar effect on mechanical behaviour.

The partial replacement of PC with RC in LC3 mixtures did not always follow the same trend. In 40CC20LF mixtures, the incorporation of RC increased the carbonation rate by 10–26%. However, in 30CC15LF mixtures, Kc increased by 10% for 20% RC but decreased by 11% for 40% RC. These mortars achieved greater porosity refinement, especially at 40% RC, which likely compensated to some extent for the significant CH reduction.

4 Conclusions

The integration of low-carbon concrete into modular construction represents a transformative step toward sustainable infrastructure. By using alternative materials, such as recycled cement, calcined clays, and bottom ash, the construction industry can reduce its environmental footprint while meeting growing demands. Modular construction's efficiency and compatibility with these materials amplify its potential as a solution for eco-efficient development. Some of these 'green' concretes have sufficiently high strengths in the first few days to be used in prefabrication, without significantly affecting the traditional production method. Others have a slower resistance development and therefore their use in modular construction will require an adaptation in the production line.

RC has shown excellent potential as a partial replacement for clinker, maintaining mechanical strength while significantly refining the microstructure. Except for carbonation, the mixtures combining LC3 with RC demonstrated remarkable potential, achieving substantial clinker reductions without compromising its mechanical and durability performance. Concrete design with these binders must be adjusted regarding carbonation resistance. These findings highlight the viability of LC3 + RC formulations, with up to over 80% reduction in clinker content, as a key innovation in the development of truly sustainable cementitious materials.

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