

Bond strength evolution of rendering mortars incorporating ladle furnace slag under natural outdoor exposure

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ABSTRACT

Ladle furnace slag (LFS), a byproduct of steelmaking with slow-reacting phases, has emerged as a potential supplementary binder to partially replace Portland cement (PC), although its in-service behavior remains poorly understood. This study investigates how bond strength develops in rendering mortars incorporating LFS under natural outdoor conditions. Two mortar systems were analyzed: lime–cement mixed mortars and admixture-based mortars. Mechanical, physical, and microstructural analyses were used to elucidate bonding behavior in rendering mortars. Lime-based systems demonstrated stable long-term performance, with LFS showing a gradual increase in mortar–substrate bond strength of approximately 9–19% between 90 and 180 days, reaching values comparable to the reference mixtures (up to 0.293 MPa). In contrast, admixture-based mortars showed reductions in bond strength of up to 17% over time when LFS was incorporated, indicating greater sensitivity to environmental moisture exposure. Microstructural analyses confirmed limited participation of LFS in hydration, with results strongly influenced by the binder's chemical composition and environmental exposure. The findings indicate that LFS can be safely used in lime–cement rendering mortars, whereas its incorporation in admixture-based systems requires careful control of moisture transport properties to ensure long-term performance, supporting its potential as a sustainable supplementary binder for rendering mortars exposed to natural environmental conditions.

1. Introduction

Concerns about climate change have highlighted two main challenges: reducing the extraction of natural resources and managing the waste produced by human activities [1,2]. In the construction industry, Portland cement (PC) production is responsible for approximately 8% of global anthropogenic CO₂ emissions [3,4]. The combination of high-temperature processes, energy-intensive chemical reactions, and the enormous demand for the material makes PC one of the most difficult sectors to decarbonize [5].

To address this burden, industrial by-products, such as steel slags, have been increasingly incorporated into concrete production, used as supplementary cementitious materials (SCMs). Among these, Ladle Furnace Slag (LFS) attracts attention, with an estimated generation of

about 0.02 tons per ton of steel produced, corresponding to more than 30 million tons worldwide [6–8]. LFS is rich in calcium oxide (CaO) and calcium hydroxide (Ca(OH)₂), in addition to oxides of aluminum, silicon, and iron [9,10]. Its reactivity is strongly influenced by the cooling method [11]. Slowly air-cooled slags tend to exhibit lower hydraulic reactivity due to their predominantly crystalline structure, whereas rapid water cooling promotes the formation of more amorphous and reactive phases [12]. Consequently, the engineering performance of LFS cannot be inferred solely from its bulk oxide composition, since the mineralogical form of these phases, governed by the cooling history, largely determines their reactivity and long-term behavior [13,14]. Furthermore, the thermal behavior is also influenced, as more reactive slags generally exhibit greater mass loss during thermal decomposition and higher loss on ignition (LOI) [14,15].

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Previous research has highlighted its potential as a partial replacement of PC in paste, mortar, and concrete [16–21]. Its effectiveness, however, largely depends on fineness, chemical variability, and processing methods [11,12,20]. In cement pastes, LFS can partially replace PC, typically at 5–25% by weight. It participates in hydration reactions, although generally at a slower rate than clinker, thereby affecting early strength development [9,22]. In concrete, studies have investigated LFS at PC mass substitution levels of 5–30%, indicating that levels of 10–15% produced mechanical performance comparable to or superior to that of the reference mixtures [23,24]. In general, although the reported optimal levels vary with the incorporation strategy, most studies converge on an upper limit of approximately 30% LFS to preserve mechanical performance [13,24,25].

Mortars also represent an important application area, as they are produced in large volumes for masonry and rendering [26]. This application is a particularly promising target for incorporating LFS, as it imposes fewer mechanical requirements than concrete [21]. In line with findings for concretes, replacement levels of around 30% by mass of PC can also be adopted in mortars without compromising mechanical performance [16,17]. In rendering mortars, the lower stiffness associated with reduced compressive strength may even be beneficial, as it facilitates deformation accommodation and reduces shrinkage and cracking without impairing serviceability [27]. Furthermore, the use of air-cooled LFS may reduce the risk of deleterious expansion compared with more reactive steel slags, owing to its lower hydraulic reactivity and greater mineralogical stability [12,22,24]. However, to fully enable broader use of LFS in mortars, further research is needed to evaluate long-term durability.

It is important to compare and benchmark mortars containing LFS with other conventional binders, such as lime, whose interactions can significantly affect their properties. Lime has traditionally enhanced workability, water retention, bond strength, and shrinkage control [28, 29], but its effects in LFS-containing mixtures depend on binder ratios, curing conditions, and exposure environments [30]. Conversely, chemical admixtures increasingly replace lime as the primary means of adjusting rheology and water retention [31]. Viscosity modifiers, most commonly cellulose ethers, enhance flow behavior and stability, reducing the traditional reliance on lime for workability control [29,32]. This shift is critical for evaluating SCMs, as their performance must be assessed in both lime-based and admixture-driven mortar systems [31, 33].

In addition to rheological control, LFS performance can be further enhanced by combining it with reactive SCMs. As LFS exhibits limited pozzolanic activity and slower hydration kinetics, pozzolanic materials such as Metakaolin (MK) can complement its behavior by promoting the formation of additional calcium silicate hydrates (C-S-H) [24]. When combined with LFS, MK can promote microstructural development, contributing to overall mechanical and bond performance [21,30]. The incorporation of SCMs into lime-based mortars has been widely investigated to improve performance through pozzolanic reactions while maintaining the compatibility required for rendering applications [34–36]. Zhang et al. [37] incorporated ground granulated blast-furnace slag and silica fume into natural hydraulic lime mortars, compressive strength increases of up to 227% at 28 days, together with enhanced sulfate and acid resistance, which were attributed to the formation of additional C-(A)-S-H phases and a denser microstructure. Similar benefits have also been reported for air lime systems incorporating metakaolin, in which the pozzolanic reaction promoted the formation of C-S-H, C-A-H, and C-(A)-S-H phases, increasing the 28-day compressive strength from 13.77 to 21.06 MPa and the flexural strength from 4.17 to 5.26 MPa compared with natural hydraulic lime mortars [38]. These studies demonstrate the potential of SCMs to improve the performance of lime-based mortars. However, the use of LFS as an SCM in lime-based rendering mortars remains underexplored [11,24].

To enable the practical use of LFS, durability analysis should not be limited to mechanical strength and deformation mechanisms (shrinkage

and cracking), but also include bond strength to the substrate, which is especially important in rendering mortars. It is essential not only to protect the substrate against aggressive environments but also to ensure the watertightness of the building envelope, maintaining the overall functionality of the construction system [39]. Insufficient bond strength can accelerate deterioration under environmental exposure, highlighting the need to carefully evaluate this property when incorporating LFS into mortar formulations. This requirement becomes crucial for external-rendering mortars, which are subjected to wetting–drying cycles, carbonation, and chloride or sulfate ingress [39–41].

Despite its importance, the bond strength of rendering mortars with LFS remains insufficiently understood. Table 1 summarizes representative studies on the use of LFS in mortars, highlighting the properties evaluated, bond strength assessment, and exposure conditions.

Only Rodríguez et al. [42] and Borges Marinho et al. [30] directly evaluated the bond strength of LFS-containing mortars, and both investigations were conducted exclusively under laboratory conditions. This limitation has also been recognized in recent review papers. Najm et al. [24] highlighted that bond performance still requires systematic assessment when LFS is incorporated into cement-based systems. Wu et al. [11] emphasized the need for studies addressing long-term performance under realistic service conditions. Among the available experimental studies, Borges Marinho et al. [30] reported bond strength increases of up to approximately 65% in mixed mortars containing LFS, attributing this improvement to the filler effect of finely ground LFS and the formation of additional hydration products. Taken together, these findings indicate that LFS may affect interfacial performance; however, further research is needed, particularly under realistic exposure conditions.

Natural humidity cycles, temperature fluctuations, and other environmental stressors strongly affect bond strength and long-term durability. These effects are not fully captured in laboratory conditions [44]. The evolution of bond strength under outdoor exposure depends not only on the mortar's intrinsic properties but also on changes at the mortar–substrate interface [45,46]. Repeated wetting–drying cycles promote moisture transport and differential dimensional changes between the substrate and the rendering layer, while carbonation progressively alters the interfacial microstructure by consuming Ca(OH)_2 and precipitating CaCO_3 [47]. In addition, daily temperature fluctuations generate cyclic stresses due to differential thermal movements between the mortar and the substrate [48,49]. The combined action of these mechanisms progressively alters the mortar–substrate interface throughout service life, effects that are not fully reproduced under conventional laboratory curing conditions [50,51].

Although the influence of these mechanisms is recognized, research on LFS-based rendering mortars under natural outdoor conditions remains scarce, with a notable lack of studies addressing bond strength evolution. Such exposure conditions have been widely investigated for mortars incorporating other residues or partially replacing PC, highlighting the need for field-based evaluations [32,52–55]. Santos et al. [54] applied lime-based mortars to masonry panels and combined laboratory tests with *in situ* bond strength measurements, demonstrating that bond strength evolves differently under field conditions than under laboratory curing. The analysis also incorporated ultrasonic pulse velocity (UPV) tests as a complementary indicator of mortar quality, as it can reflect densification processes, stiffness development, and potential degradation [56]. Similarly, Ruviano et al. [55] investigated PC-based rendering mortars with recycled aggregates applied to an exterior wall for 1059 days. Their results showed that prolonged field exposure induces chemical and microstructural changes that are difficult to detect under controlled laboratory conditions. Thermogravimetric analysis (TGA) played a central role in identifying these transformations, providing direct evidence of progressive carbonation, revealing marked reductions in Ca(OH)_2 and substantial increases in CaCO_3 , and exposing microstructural densification processes that mechanical tests alone could not elucidate.

Table 1

Summary of representative studies on the use of ladle furnace slag (LFS) in mortars, highlighting bond strength assessment and exposure conditions.

Reference	LFS application	Material	Bond strength	Exposure/curing condition	Main contribution / Remaining gap
Manso et al. (2011) [16]	30% of cement replacement	Masonry mortar	No	Laboratory	Long-term laboratory durability of masonry mortars; no bond strength evaluation.
Rodríguez et al. (2013) [42]	100% of fine aggregate replacement	Rendering (plaster) mortar	Yes	Laboratory	Bond strength of rendering mortars under laboratory curing only.
Herrero et al. (2016) [17]	5, 10, 15 and 20% of cement replacement	Masonry mortar	No	Laboratory	Mechanical and durability performance; no bond strength evaluation
Marinho et al. (2017) [30]	100% of lime replacement	Rendering and masonry mortars	Yes	Laboratory	Bond strength of rendering mortars under laboratory curing only.
Najm et al. (2021) (Review) [24]	—	Mortars and concrete	—	—	Review of LFS in mortars and concrete; highlights the lack of bond strength assessment and long-term field performance.
Sultana et al. (2022) [43]	5, 10, 15, 20, 25, and 50% of cement replacement	Mortar	No	Laboratory	Mechanical performance under different curing regimes; no bond strength evaluation.
Wu et al. (2023) (Review) [11]	—	Cementitious materials	—	—	Review of LFS utilization; identifies the need for long-term performance assessment under realistic service conditions.

Based on the literature summarized above, it is evident that understanding the bond strength behavior of mortars containing LFS under natural exposure is essential for their practical application. Therefore, this study presents a systematic evaluation of bond strength development in rendering mortars incorporating LFS under natural outdoor exposure, combining mechanical testing with UPV, TGA, and scanning electron microscopy (SEM) to elucidate the governing microstructural mechanisms.

2. Materials

The ladle furnace slag (LFS) by-product used in this study was supplied by a steel plant in the Piracicaba region of Brazil, where LFS is produced under a slow air-cooling process. Due to its low intrinsic reactivity and heterogeneity, the LFS was pre-processed before use. First, it was sieved through a 30-mesh sieve (0.6 mm) to remove impurities and metallic residues. It was then ground in a ball mill. Grinding was carried out in 10-minute cycles at a steady rotation speed of 200 rpm, processing about 400 g of material per cycle, using a Marconi MA500 ball mill with an alumina ceramic vessel. This process followed procedures previously reported by Silva et al. [12].

The cement used was Brazilian CP II-F (strength class 32 MPa at 28 days), which contains an inert limestone filler and is comparable to ASTM Type II cement [57]. MK was obtained from a stabilized commercial product and is characterized by high pozzolanic reactivity, attributable to the controlled calcination of kaolinite. Air lime (AL) was a commercial hydrated lime, classified as CH III according to ABNT NBR 7175 [58], commonly used in rendering mortars to improve workability and water retention. Quartz sand (QS) was a naturally occurring siliceous aggregate of local origin used as the fine aggregate in all mortar formulations. The sample had a characteristic maximum size of 0.60 mm and a fineness modulus of 1.06, indicating a fine aggregate mostly made up of particles smaller than 0.30 mm, which is typical for sands used in

rendering mortars. The average particle density was 2.64 g/cm³ and water absorption was 0.20%, indicating a dense, low-porosity material. This contributes to dimensional stability and helps control water demand. The water-retaining admixture (AD) used was a cellulose-based polymer, synthesized from hydroxypropyl methylcellulose (HPMC). When in contact with water, this AD forms a transparent, stable gel that improves water retention and workability by temporarily immobilizing some of the mixing water within the fresh mortar matrix. Table 2 presents the chemical compositions of the binders used in this study and compares LFS with literature data, highlighting its variability and mineralogical features.

The batch of LFS used in this study has a high CaO content (54.6% by weight), similar to values reported in the literature for LFS, and relatively low SiO₂ and Al₂O₃ levels, with significant amounts of Fe₂O₃, MgO, and MnO. The basicity index, calculated as (CaO + MgO)/(SiO₂ + Al₂O₃), is about 5.7 [62], indicating a strong presence of basic oxides and confirming the slag's highly alkaline nature. According to Wu et al. [11], the Fe₂O₃ content over 5% by weight places the material in the CaO–SiO₂–Al₂O₃–FeO–MgO system (type IV LFS). Although the lower SiO₂ and Al₂O₃ levels suggest limited intrinsic hydraulic reactivity compared with PC, the high CaO content and alkalinity support the use of this LFS as an SCM. The relatively high LOI (12.0 wt%) of the LFS used in this study, compared with the values reported in the literature [59, 61], does not necessarily indicate the presence of free CaO or MgO. Instead, it may also reflect the presence of hydrated and carbonated phases formed during cooling, storage, and handling of the air-cooled slag, which contribute to mass loss during heating [22]. AL contained 38.7 wt% CaO and 27.3 wt% MgO, indicating a magnesium-rich composition that differs from those reported for hydrated limes used in previous studies on rendering mortars [63,64].

Table 3 summarizes the key physical parameters of the binder materials, and Fig. 1 presents their particle-size distribution curves. These parameters are relevant to interpreting fresh-state behavior and packing

Table 2

Chemical composition and LOI index of the binders used in this study, and comparative data for LFS reported in the literature.

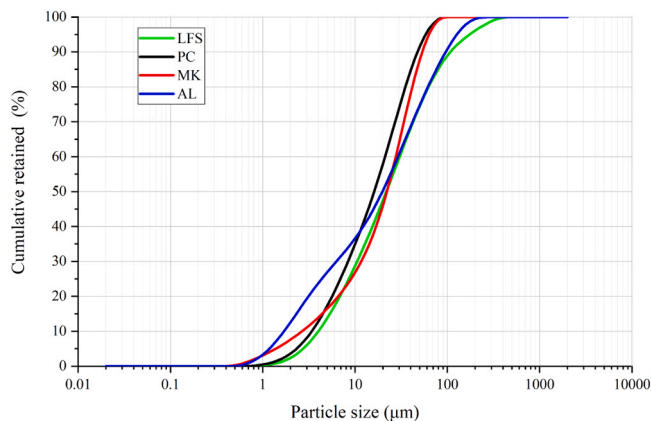
Material	Oxide (wt%)											
	Na ₂ O	CaO	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MgO	MnO	K ₂ O	SO ₃	P ₂ O ₅	TiO ₂	LOI*
PC	0.12	57.5	15.3	3.62	2.32	4.28	< 0.10	0.72	2.65	0.10	0.19	12.5
AL	-	38.7	1.01	0.15	0.25	27.3	0.03	0.04	0.04	< 0.01	0.02	32.4
MK	0.05	0.13	50.8	35.9	3.43	0.7	-	3.12	0.1	0.07	2.11	3.36
LFS	0.06	54.6	9.78	0.80	7.59	5.79	7.29	0.16	0.60	0.09	0.19	12.0
LFS in literature												
Islam et al. [19]	1.27	46.14	31.42	2.95	0.88	2.13	1.49	0.07	-	-	0.79	
Singh et al. [59]	0.72	65.85	1.73	20.45	3.08	2.44	-	-	1.44	-	-	2.01
Václavík et al. [60]	0.06	47.6	12.1	5.08	21.1	3.65	3.01	0.03	0.89	0.61	0.26	
Thwe et al. [61]	-	58.9	25.2	7.7	0.3	4.4	-	-	1.9	-	-	1.1

* LOI: Loss on ignition.

Table 3

Key physical parameters of the raw materials used in the mortar formulations.

Material	d(0.1) (μm)	d(0.5) (μm)	d(0.9) (μm)	Specific surface area (m^2/g)	Bulk density (kg/m^3)	Specific gravity (g/cm^3)
MK	0.71	2.669	53.345	0.888	729.85	2.73
LFS	1.57	4.003	107.959	0.588	722.34	2.75
PC	0.86	3.405	47.398	0.733	974.72	3.01
AL	1.47	1.702	95.959	1.140	695.56	2.50

**Fig. 1.** Particle size distribution curves of the starting materials (LFS, PC, MK, and AL) used in the mortar formulations.

efficiency, and to their implications for in-service performance.

Fig. 2 shows the X-ray diffraction (XRD) patterns of the binder materials used in this study, revealing their main crystalline phases and offering insight into their fundamental mineralogical properties.

PC consists mainly of clinker phases, particularly alite (C_3S) and belite ($\beta\text{-C}_2\text{S}$), confirming its role as the primary reactive component and the primary source of early calcium release in the system. In contrast, AL exhibits diffraction peaks characteristic of an air lime with a complex mineralogical composition. To support its mineralogical classification, quantitative phase analysis by Rietveld refinement indicated approximately 28.5 wt% brucite, 27.2 wt% calcite, 23.0 wt% portlandite, 11.4 wt% dolomite, 6.3 wt% periclase, and 3.5 wt% quartz. These values represent the relative proportions of the identified crystalline phases, normalized to 100 wt%, and therefore do not account for any

amorphous content. Based on the chemical composition (**Table 2**) and the quantitative mineralogical analysis, the material was identified as a dolomitic hydrated lime in accordance with the terminology adopted in EN 459-1 [65]. However, this mineralogical assemblage differs from those reported for hydrated limes in the rendering mortar literature. In their respective studies, Palomar et al. [64] and González-Sánchez et al. [63] employed calcitic hydrated limes, and interpreted the development of their binders mainly in terms of portlandite carbonation and, when pozzolanic additions were present, the consumption of Ca-bearing phases. Therefore, the significant amounts of brucite and dolomite identified in the AL used in the present study may alter the kinetics of hydration and carbonation, since Mg-bearing phases are generally less reactive than Ca-bearing phases [65].

Within this alkaline environment, MK and LFS exhibit contrasting behaviors. MK exhibits limited crystallinity, primarily comprising quartz, muscovite, and residual kaolinite, while its most reactive fraction remains largely amorphous and thus not fully detectable by XRD. Conversely, LFS exhibits a predominantly crystalline mineralogy comprising periclase, brucite, olivine, anorthite, and minor calcium silicates, with no prominent amorphous halo. Nevertheless, this does not preclude its participation in hydration reactions in multicomponent binder systems, where its contribution is generally slower than that of clinker [12,22]. Hence, despite the relatively high contents of CaO , MgO , Fe_2O_3 , and MnO reported in **Table 2**, the predominance of crystalline phases suggests that the behavior of the LFS is governed primarily by its mineralogical composition and cooling history rather than by its bulk oxide composition alone.

In addition to chemical, physical, and mineralogical characterization, the morphology of the raw binder materials was analyzed using scanning electron microscopy (SEM) to provide further insight into particle shape and surface texture. The micrographs obtained are shown in **Fig. 3**.

The LFS particles (**Fig. 3a**) are mostly angular and irregular, with rough surfaces and signs of fragmentation, consistent with mechanical grinding following air cooling. PC particles (**Fig. 3b**) display an irregular morphology with angular edges and a relatively wide range of particle sizes, typical of ground clinkers. AL (**Fig. 3c**) consists of finer particles with a plate-like, agglomerated morphology, consistent with the hydroxide phases identified by XRD (**Fig. 2**). MK particles (**Fig. 3d**) exhibit lamellar and flaky morphologies characteristic of thermally treated kaolinite, with thin, plate-like structures and occasional clustering.

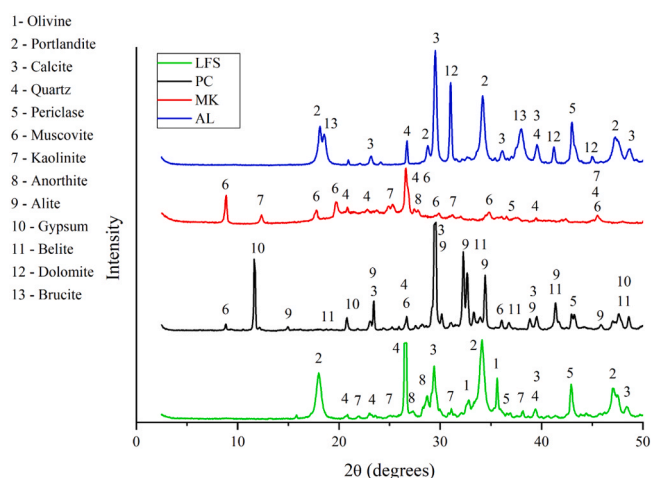
3. Experimental program

3.1. Preparation of rendering mortars

Rendering mortars were prepared by partially replacing PC with LFS at 30% by mass, based on previous studies that identified this replacement ratio as both technically feasible and environmentally beneficial [16,21,66,67]. Reference formulations were defined using normalized mass ratios, with PC as the baseline unit ($\text{PC} = 1$), corresponding to conventional volumetric ratios of 1:2:6 (PC:AL:QS) for lime-based mortars and 1:6 (PC:QS) for lime-free mortars (including AD). MK was added at 10% of the PC (or PC+LFS) mass in half of the formulations to improve pozzolanic activity. For mixes containing the AD, a dose of 0.6 g of AD per kg of dry materials was used, based on initial flow table tests. The water content of each mixture was adjusted to achieve a target flow consistency of 260 ± 5 mm, measured using a standard flow table test in accordance with NBR 13276 [68], equivalent to ASTM C1437 [69].

Eight mortar formulations were produced: four with AL and four with AD. The full mix proportions are shown in **Table 4**.

The calculated w/b ratio differs inherently between the AL and AD systems due to their mixture proportions. In the AL series, both PC and AL were considered as binders, whereas in the AD series, only PC constituted the binder fraction. Accordingly, the mortars were

**Fig. 2.** X-ray diffraction (XRD) patterns of the binder materials used in this study: ladle furnace slag (LFS), Portland cement (PC), metakaolin (MK), and air lime (AL), with the main crystalline phases identified.

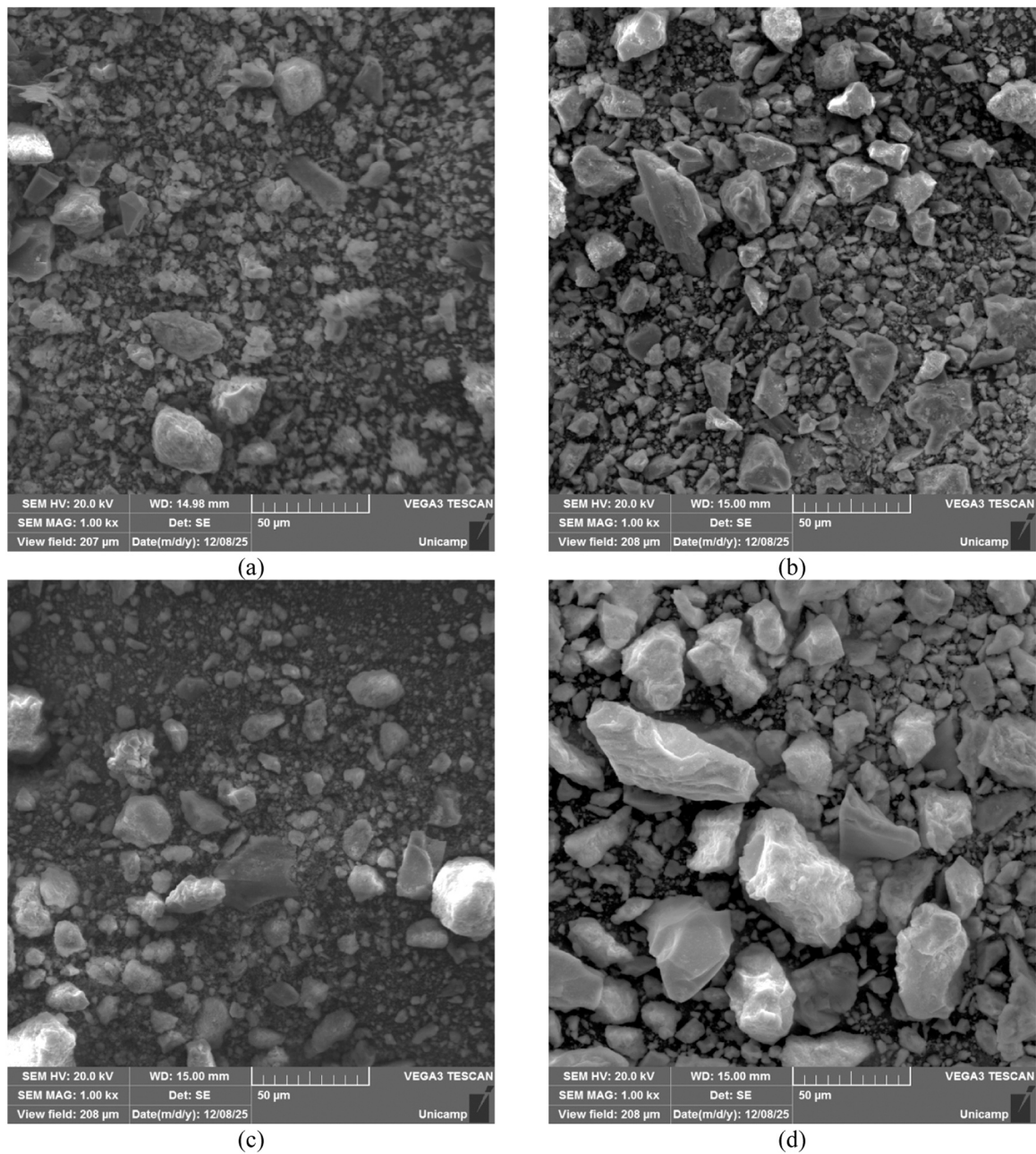


Fig. 3. SEM micrographs of the raw binder materials: (a) ladle furnace slag (LFS), (b) Portland cement (PC), (c) air lime (AL), and (d) metakaolin (MK).

Table 4

Theoretical mortar constituent contents obtained from the mix design using the absolute-volume method, including LFS, MK, and a water-retaining admixture (AD). The admixture was added at a fixed dosage of 0.6 g/kg of dry materials.

Series	Binders (kg)			Aggregate (kg)	Addition (kg)	Admixture (g/kg)	w/b ratio
	PC	AL	LFS	QS	MK	AD	
AL-REF	149.78	220.18	0.00	1425.94	0.00	-	0.87
AL-MK	145.87	214.42	0.00	1388.65	11.67	-	0.93
AL-LFS	102.49	215.22	33.67	1393.80	0.00	-	0.99
AL-MK-LFS	101.75	213.68	33.43	1383.81	11.63	-	1.00
AD-REF	164.52	0.00	0.00	1566.23	0.00	0.6	2.14
AD-MK	162.93	0.00	0.00	1551.10	13.03	0.6	2.17
AD-LFS	112.16	0.00	49.67	1525.31	0.00	0.6	2.29
AD-MK-LFS	111.63	0.00	49.44	1518.18	12.76	0.6	2.29

proportioned to achieve a common target consistency (260 ± 5 mm) rather than a fixed w/b ratio, reflecting the practical objective of designing rendering mortars representative of conventional applications. Therefore, differences in the w/b ratio resulted from both the adopted mixture design and the water demand required to achieve the target consistency.

These differences in water demands were associated with the variations in binder composition and particle characteristics. In the AL series, lime was included in the binder fraction and, because of its very fine particles and high specific surface area, increased the amount of water needed for workable consistency. The presence of LFS further raised water demand in these mixes because the milled slag contains angular particles with a wider size distribution and higher absorption capacity. In the AD series, the cellulose-based AD immobilized part of the mixing water, requiring additional water to maintain flow. When combined with LFS, this effect was more pronounced, resulting in the highest water content among all formulations. Consequently, the fresh- and hardened-state properties reflect the combined effects of the adopted binder composition and the corresponding water demand of each formulation.

After mixing, the mortars were split into two sets of specimens. The first set was used for laboratory characterization under controlled curing conditions, while the second set was prepared for performance assessment under natural weather exposure. Laboratory characterization tests were performed at 28 days, representing the standard reference age for mortar characterization under controlled curing conditions [70,71]. Although hydration, carbonation, and other microstructural processes continue beyond this age, particularly in lime- and SCM-containing systems, the 28-day results provide a standardized baseline for comparison among the mixtures rather than the final stage of microstructural development. Therefore, the laboratory characterization was used as a reference for interpreting the subsequent performance of the outdoor panels. In contrast, the panels exposed to natural weathering were evaluated after 90 and 180 days to capture the time-dependent evolution of bond strength and microstructural properties under real environmental conditions.

3.2. Characterization of rendering mortars

The characterization set of mortar samples was evaluated in both the fresh and hardened states. In the fresh state, measurements were taken of consistency, density, entrained air content, and water retention. For hardened-state characterization, prismatic specimens ($40 \times 40 \times 160$ mm) were prepared and cured under controlled laboratory conditions at 20 ± 2 °C and 65% $\pm$ 5% relative humidity. At 28 days, the mortars were tested for compressive strength, flexural strength, water absorption by capillarity, bond strength, and the dynamic modulus of elasticity. For statistical reliability, all tests were performed with at least three repetitions, except for the bond strength test, which used 12 specimens to better account for the greater variability inherent in the mortar–substrate interface. Table 5 summarizes all tests conducted,

Table 5
Summary of characterization tests and corresponding standards.

Property / Test	Standard	State of mortar
Preparation of mortar mix	NBR 16541 [72]	Fresh
Consistency	BS EN 1015-3 [73]	Fresh
Fresh density	BS EN 1015-6 [74]	Fresh
Entrained air content	NBR 13278 [75]	Fresh
Water retention	NBR 13277 [76]	Fresh
Bulk density	ASTM C138M-16A [77]	Hardened
Compressive strength	BS EN 1015-11 [78]	Hardened
Flexural strength	BS EN 1015-11 [78]	Hardened
Water absorption by capillarity	NBR 15259 [79]	Hardened
Bond strength (pull-off)	BS EN 1015-12 [80]	Hardened
Dynamic modulus of elasticity	NBR 15630 [81]	Hardened

indicating the corresponding standards and the evaluated material state.

3.3. Performance evaluation of rendering mortars under natural weathering conditions

For external testing, masonry panels were prepared using concrete blocks with nominal dimensions of 390 mm in length, 190 mm in width, and 190 mm in depth. The blocks exhibited a characteristic compressive strength of approximately 4 MPa, with a measured average of 4.3 MPa. Water absorption by capillarity was limited, with an average value of 6.6%, indicating moderate porosity compatible with plaster applications.

Each panel was assembled by stacking two concrete blocks with a horizontal mortar joint between them, resulting in composite panels with final dimensions of approximately 390 mm in height and 390 mm in width. Prior to mortar application, the panel surfaces were pre-moistened by brushing clean water onto the substrate to reduce its initial water absorption. The mortar was then applied immediately while the surface remained damp but free of standing water. No cementitious bonding layer was employed to assess the mortars' intrinsic bond strength. A leveled rendering layer with a nominal thickness of 10 mm was then applied to both faces of each panel. Three panels were produced for each mortar mix, and samples were subsequently collected from both faces of each panel to ensure representative bond strength and ultrasonic measurements. Fig. 4a shows the prototype prism before mortar application, while Figs. 4b and 4c illustrate the arrangement of the panels after rendering and prior to outdoor exposure.

The panels were positioned vertically to ensure uniform exposure to environmental cycles, following methods described in this kind of analysis [55,82]. Fig. 5 shows the layout of the panels in an uncovered, open-air environment in the experimental field at the School of Civil Engineering, Architecture, and Urban Design (University of Campinas, Brazil). During the exposure period, the mean air temperature was 24.1 °C, with a range of 9.5–36.1 °C. The mean relative humidity was 75.3% (range: 21.4%–100%), and cumulative precipitation totaled 380.7 mm, according to meteorological data from the CIIAGRO database [83]. These environmental conditions exposed the material to rainfall, solar radiation, wind, and daily fluctuations in temperature and humidity throughout the exposure period.

The ultrasonic pulse velocity (UPV) methodology followed the approach proposed by Amaral et al. [84], which presents a detailed ultrasonic testing protocol for structural concrete blocks and establishes correlations between UPV and compressive strength, considering geometric variations and material properties. Fig. 6a illustrates the positioning of the transducers used in the tests, ensuring that the ultrasonic wave passes through both the mortar and the block via direct transmission.

To assess mortar bond strength, the panels were coated in accordance with BS EN 1015-12 [80], and the mortar was applied vertically onto the panels using a trowel. After mortar hardening, the samples were prepared by cutting the rendering layer with a 50 mm-diameter core drilling machine, penetrating approximately 2 mm into the substrate. The pull-off tests were performed using a hydraulic adhesion tester equipped with a load cell rated at approximately 20 kN (1.999 ton). Specimens that showed visible damage during cutting were discarded. The general setup of the exposed panels and the pull-off test configuration is illustrated in Fig. 6b.

Finally, chemical and microstructural characterization was carried out using thermogravimetric analysis (TGA/DTG) and SEM. The TGA measurements were performed using a NETZSCH STA 449F3 thermal analyzer, using approximately 11 mg of powdered sample placed in an open alumina crucible (85 μ L). The tests were conducted in a nitrogen atmosphere (50 mL/min) from 20 to 1000 °C at a heating rate of 10 °C/min. This technique was used to identify portlandite consumption, carbonation progress, and other thermal events associated with hydration and microstructural evolution under extended outdoor exposure.

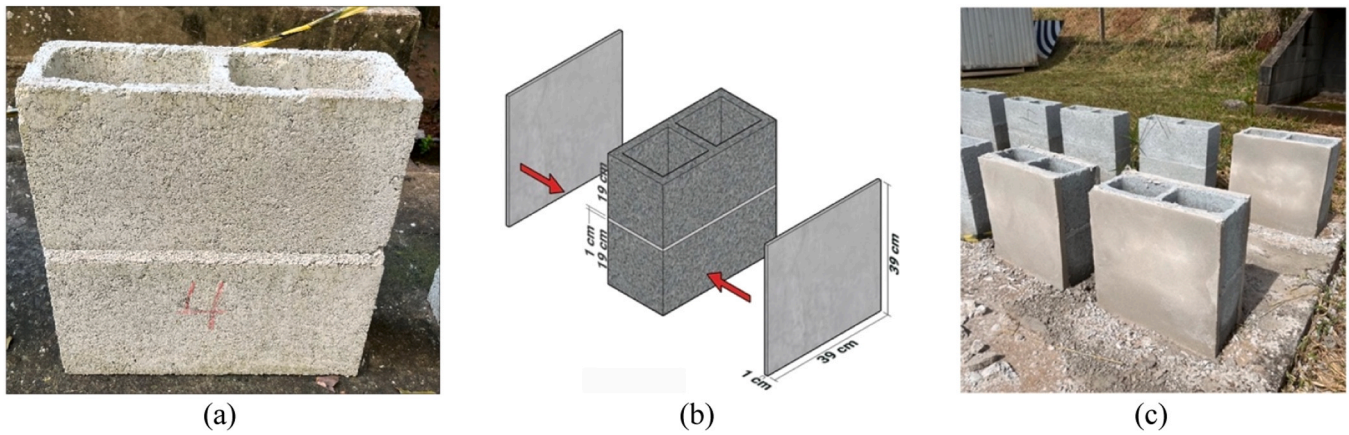


Fig. 4. (a) Prototype of the prism used to produce the rendering panels, (b) mortar-coated panel prepared for outdoor exposure, showing mortar applied on both faces (not to scale), and (c) panels after mortar application.

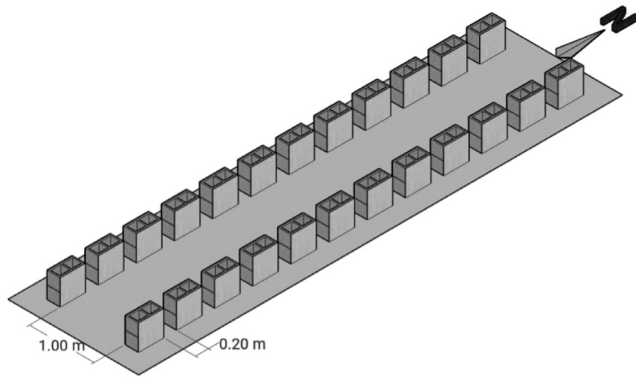


Fig. 5. Schematic layout of the mortar-coated panels in the outdoor exposure field, showing their orientation, relative spacing, and north direction (not to scale).

For quantitative analysis, mass losses were measured over the main temperature intervals associated with the dehydration of C-S-H, Aft, and $\text{Mg}(\text{OH})_2$ (100–400 °C), the dehydroxylation of $\text{Ca}(\text{OH})_2$ (400–600 °C), and the decarbonation of CaCO_3 and other carbonate phases (600–900 °C). The corresponding phase contents were calculated according to the

stoichiometric relationships proposed by Lothenbach et al. [85], as given in Eqs. (1) and (2).

$$\begin{aligned} \text{Ca}(\text{OH})_2(\%) &= \Delta m_{400-600} \times \frac{M_{\text{Ca}(\text{OH})_2}}{M_{\text{H}_2\text{O}}} \\ &= \Delta m_{400-600} \times \frac{74.09}{18.02} \end{aligned} \quad (1)$$

$$\begin{aligned} \text{CaCO}_3(\%) &= \Delta m_{600-900} \times \frac{M_{\text{CaCO}_3}}{M_{\text{CO}_2}} \\ &= \Delta m_{600-900} \times \frac{100.09}{44.01} \end{aligned} \quad (2)$$

where $\Delta m_{400-600}$ and $\Delta m_{600-900}$ are the mass losses measured between 400–600 °C and 600–900 °C, attributed primarily to the dehydroxylation of portlandite and the decarbonation of calcium carbonate, respectively. $M_{\text{Ca}(\text{OH})_2}$ and M_{CaCO_3} are the molar masses of portlandite and calcium carbonate, respectively. $M_{\text{H}_2\text{O}}$ and M_{CO_2} are the molar masses of the water and carbon dioxide released during dehydroxylation and decarbonation, respectively.

SEM analyses were conducted using a VEGA3 scanning electron microscope (TESCAN, Czech Republic) equipped with secondary electron (SE) and backscattered electron (BSE) detectors. The instrument operates with a tungsten filament and a maximum acceleration voltage of 30 kV, offering a theoretical resolution of up to 3 nm under high

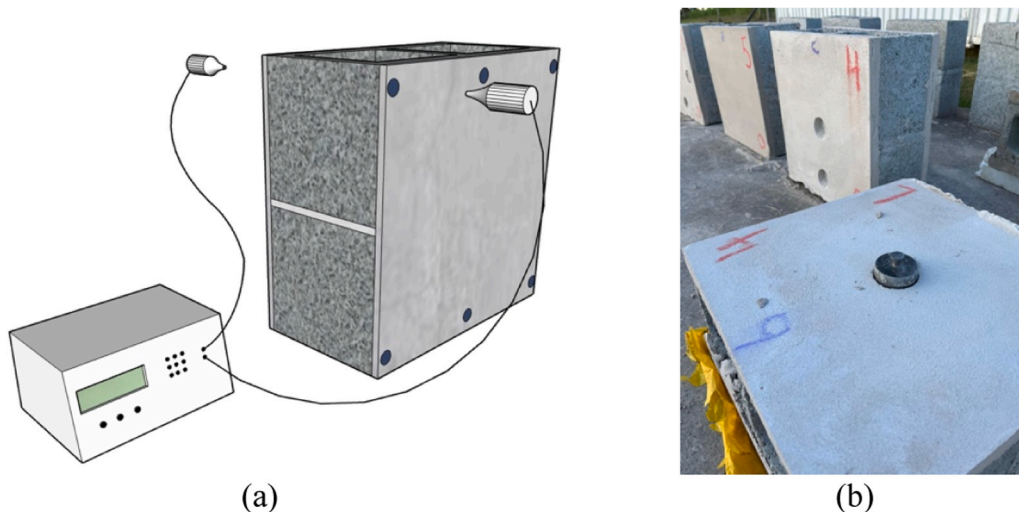


Fig. 6. (a) Schematic illustration of the ultrasonic pulse velocity test in direct transmission, showing the positioning of the transducers across the rendering layer and concrete block, and (b) mortar-coated panel after coring, showing the test areas prepared for pull-off adhesion testing.

vacuum conditions. For microstructural analysis, polished cross-sections of the mortars were prepared. Hydration reactions were initially stopped by solvent exchange with analytical-grade acetone, followed by drying. The samples were then embedded in acrylic resin, and the exposed surfaces were sanded and polished to produce a flat cross-section representative of the matrix and interfacial transition zones. Before imaging, the specimens were coated with a thin conductive carbon layer to prevent charging during electron beam exposure. Observations were primarily made in backscattered electron (BSE) mode to enhance compositional contrast between phases, facilitating the identification of unreacted particles, hydration products, voids, and interfacial features. SEM was also employed to examine surface morphology. This method is especially useful for phase contrast analysis in heterogeneous cementitious systems. SEM observations qualitatively supported interpretations of microstructural densification, the formation of secondary hydration products, and degradation features associated with open-air weathering cycles.

4. Results and discussion

4.1. Physical and mechanical characterization of mortars

Table 6 presents the fresh-state properties of the mortar mixtures, highlighting the effects of different binder combinations on fresh density, entrained air content, and water retention.

Mortars formulated with AL exhibited the highest fresh densities, particularly those containing MK, and the lowest air contents. This result is primarily attributed to the lower w/b ratios of these mixtures (Table 4), reflecting the combined influence of binder composition and water demand. The incorporation of MK further enhanced particle packing, leading to higher fresh densities and lower entrained air contents (Table 6), and consequently to a denser and more cohesive fresh matrix. Because of its fine particle size and lamellar shape, MK fills the voids between larger particles, resulting in a more compact fresh structure and higher bulk density [86,87]. This densification also reduced water retention in the AL-based mortars, probably because less free water remained available for suction and part of the mixing water was consumed during the early reactions associated with MK incorporation [88,89].

Alternatively, AD incorporation increased water retention capacity to about 87%, promoted air incorporation, and decreased fresh density. This weight-reducing effect is especially helpful for projection-applied rendering mortars, as it lessens gravitational effects during application, reduces initial slippage, and promotes a more stable early bond [90,91]. These trends are consistent with the higher w/b values required by the AD mixtures to achieve the target consistency (Table 4). For example, the AD-LFS mixture, which required one of the highest w/b ratios (2.29), also exhibited the highest water retention (87%) together with lower fresh density than the AL mixtures. The slightly higher value observed for AD-LFS relative to AD-REF is consistent with the overall

trend for the AD mixtures and indicates that incorporating LFS did not adversely affect the admixture's water-retention capacity. This observation may also be associated with the angular shape and rough surface texture of LFS particles (Fig. 3), which can contribute to physical adsorption of water and reduce bleeding [29].

AL-MK-LFS exhibited the highest fresh density and the lowest air content, reflecting the densification promoted by the combined incorporation of MK and finely ground LFS. These findings are in agreement with previous studies reporting that ground LFS improves particle packing and reduces voids [42,62,92].

Fig. 7 summarizes the mechanical and physical properties of the hardened mortars at 28 days.

In the lime-based series, adding LFS or MK did not cause statistically significant changes in flexural or bond strength. However, mortars with LFS exhibited lower dynamic modulus values and higher water absorption via capillarity, indicating a mortar matrix that allowed greater water transport at 28 days. This finding agrees with the lower hardened density and higher w/b ratios (Table 4). In the fresh state, the AL-LFS mixture also exhibited lower fresh density and slightly higher entrained air content than AL-MK-LFS (Table 6), indicating a less compact particle arrangement prior to hardening, which is consistent with the higher capillary absorption measured after curing [11,17,24, 89]. Herrero et al. [17] reported similar capillary absorption behavior for mortars incorporating LFS, which was associated with a less compact hardened matrix. Similarly, broader reviews on LFS behavior, highlight its heterogeneous mineralogy and delayed hydraulic reactivity compared with other steelmaking slags [24].

Overall, mortars with AL consistently exhibited higher dynamic modulus than AD-based mixtures, indicating a more continuous solid structure formed without polymer-induced air entrainment. Although bulk density varied among the mortars, water absorption by capillarity did not show a linear relationship with this property, indicating that differences in moisture transport are not governed by bulk density alone [93,94]. This is illustrated by the AD-REF and AD-MK mixtures, which exhibited similar densities but distinct capillary absorption values. The lower absorption measured for AD-MK is attributed to the incorporation of MK, which has been reported to improve matrix development through its pozzolanic activity [86,87]. AL-LFS showed absorption values similar to those of AL-MK mixtures, indicating that at 28 days the slag contribution was mainly due to a physical filler effect [42]. In addition, the higher water demand of the LFS-containing mixtures resulted in higher w/b ratios (Table 4), together with greater entrained air contents (Table 6), which collectively influenced the observed capillary absorption behavior.

When comparing binder systems, AD-based mortars generally exhibited lower absorption than lime-based mixtures, with AD-MK showing the lowest value, followed by AD-LFS. Consequently, during the evaluated curing period, the interaction between MK and the AD was more effective at reducing capillary water absorption than the lime-based system [87,95].

In terms of compressive strength, lime-based mortars showed the highest values, particularly the AL-REF mixture, indicating a more continuous and denser matrix. The incorporation of LFS reduced compressive strength, as reported in previous studies, LFS has a more pronounced negative effect on this property, especially at early ages [21, 24]. In contrast, the incorporation of MK partially compensated for this loss, consistent with the lower capillary water absorption measured in MK-containing mortars (Fig. 7a), indicating enhanced matrix densification. This increase is also consistent with the fresh-state characteristics discussed previously, which favored the formation of a denser hardened matrix. Conversely, AD-based mortars exhibited lower overall compressive strength, reflecting the combined effects of higher water demand and greater air incorporation associated with the polymer-modified mixtures.

A different trend was observed for flexural strength. Compressive and flexural strength are governed by distinct mechanisms. Compressive

Table 6

Fresh-state properties of mortars: fresh density, entrained air content, and water retention.

Series	Fresh density (kg/m ³)	Entrained air content (%)	Water retention (%)
AL-REF	1666	21.47	83
AL-MK	1706	18.75	75
AL-LFS	1680	20.60	81
AL-MK-LFS	1727	18.35	79
AD-REF	1434	31.29	80
AD-MK	1446	30.63	82
AD-LFS	1456	30.08	87
AD-MK-LFS	1485	28.59	84

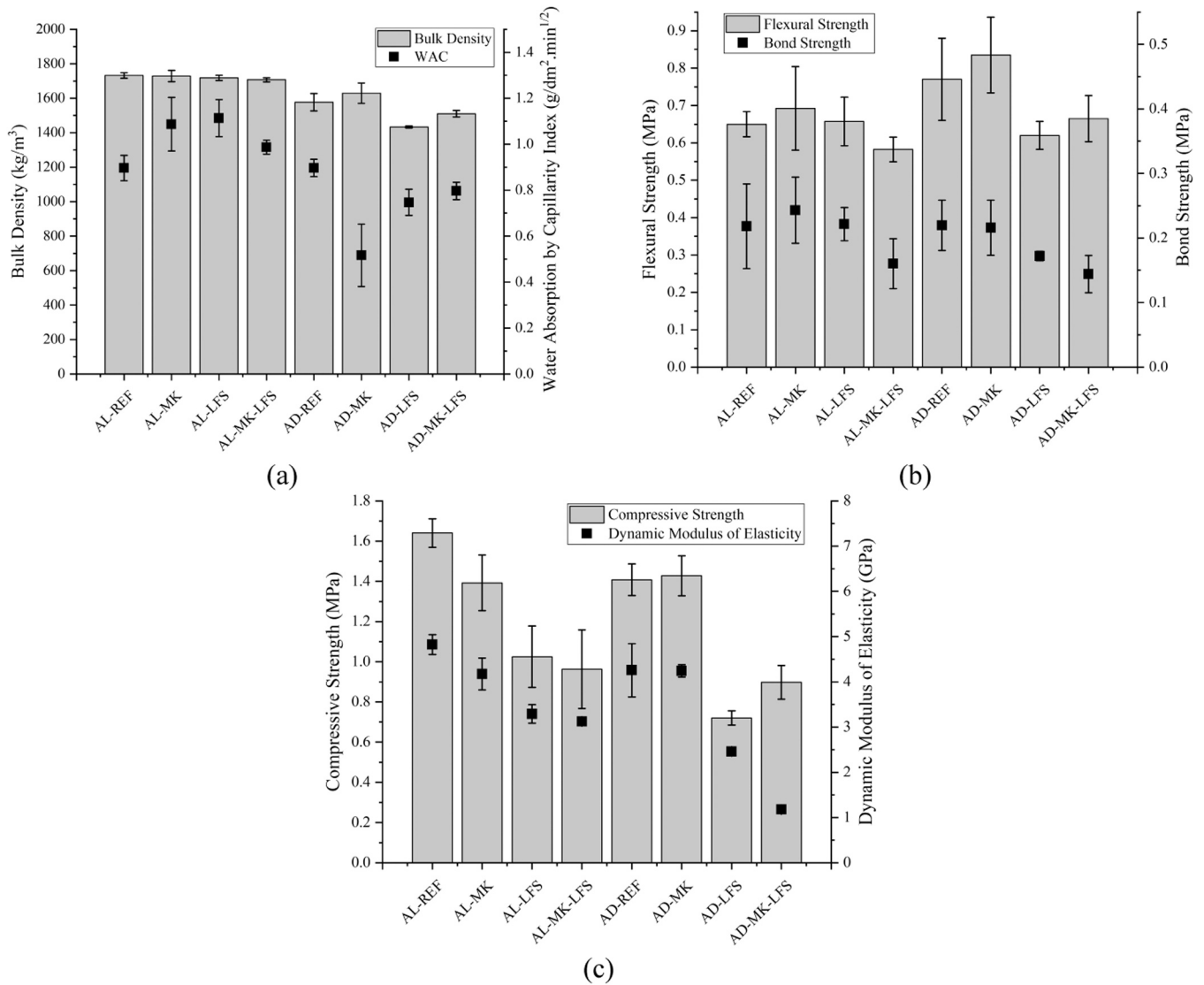


Fig. 7. Mechanical and physical properties of the mortars at 28 days: (a) bulk density and water absorption by capillarity, (b) flexural and bond strength, and (c) compressive strength and dynamic modulus of elasticity.

strength primarily depends on global matrix densification and hydration development, whereas flexural strength is more influenced by localized cracking and the properties of the interfacial transition zone [96]. Under these conditions, LFS primarily acts as a rigid filler at 28 days, contributing more to compressive than to flexural strength performance [14,97]. This behavior is reflected in the flexural results, which showed only limited variations among the AL-based mixtures. Consequently, adding MK to the AL-LFS mixture did not result in significant improvements in flexural strength at this age, indicating limited short-term synergy among AL, LFS, and MK [21]. However, this trend was not fully reflected in the bond strength results. The AD mixtures exhibited higher flexural strength than their AL counterparts; however, this was not always matched by proportional increases in bond strength. Within each binder system, flexural and bond strengths generally exhibited comparable patterns, as both are influenced by matrix cohesion and interfacial integrity. Overall, bond performance remained similar between the AL and AD systems, with some AL mixtures slightly outperforming their AD counterparts, suggesting that improvements in flexural strength did not necessarily translate into higher bond strength.

The dynamic modulus results are consistent with this interpretation. The lowest dynamic modulus values were observed for the AD-LFS and AD-MK-LFS mixtures, reflecting the combined influence of higher w/b

ratios and greater entrained air contents, which reduced matrix continuity at 28 days [21,27]. This effect was particularly evident for AD-MK-LFS, which exhibited the lowest dynamic modulus despite a slight recovery in compressive strength compared with AD-LFS. Accordingly, the improvement in compressive strength was not accompanied by a proportional increase in the overall stiffness of the hardened matrix at 28 days. In this context, the filler effect of LFS and the pozzolanic reaction of MK were insufficient to fully compensate for the slower early-age matrix development associated with clinker replacement during the evaluated curing period.

A proportional relationship between bond strength and capillary water absorption was observed in lime-based mortars and most AD mixtures, with reduced absorption associated with modest improvements in bond strength. However, this relationship was less consistent in LFS-containing AD mortars. These findings indicate that localized mechanical improvements associated with particle packing effects were not necessarily accompanied by an equivalent reduction in capillary water absorption (Fig. 7a), highlighting that strength development and capillary transport behavior are governed by different aspects of the pore structure, particularly at early ages (28 days) [98,99].

4.2. Bond performance and ultrasonic pulse velocity under outdoor exposure

Fig. 8 presents the ultrasonic pulse velocity (UPV) and bond strength of mortars after 90 and 180 days of outdoor exposure, allowing a comparative assessment of matrix stiffness and interfacial performance over time.

At 90 days, within the lime-based systems, AL-REF exhibited lower average UPV values compared to other AL mixtures, although overlapping error bars suggest a limited statistical difference at this age. The incorporation of MK and/or LFS tended to increase UPV, indicating improved microstructural continuity. Between 90 and 180 days, the UPV evolution became more closely aligned with the development of bond strength. The most significant increase occurred in AL-MK (residual = 1.31), indicating progressively increased matrix compactness. Conversely, mortars containing LFS showed smaller UPV gains, limiting uniform long-term densification, aligning with the previously discussed persistence of unreacted phases [11,18,21].

Across the two ages, most lime-based mortars exhibited concurrent increases in UPV and bond strength, indicating progressive matrix strengthening under outdoor exposure. This change is linked to carbonation processes, as evidenced by the continued presence of $\text{Ca}(\text{OH})_2$ in lime-rich systems, which promotes secondary reactions and the gradual activation of slag [100]. In addition, the progressive formation of carbonate phases likely contributed to matrix stiffening, which favors ultrasonic wave propagation and is reflected in the higher UPV values. By comparison, AD mortars containing LFS did not exhibit similar improvements, in line with the previously discussed limited reactivity of LFS and its reliance on the surrounding binder chemistry [101,102].

The influence of AD further emphasized the effects associated with LFS. In the AD series, UPV values remained relatively stable between 90 and 180 days and were consistently lower than those of the AL mortars, indicating more limited microstructural evolution under outdoor exposure. Although AD-REF showed a slight increase in UPV, the LFS-containing mixtures (AD-LFS and AD-MK-LFS) remained statistically similar to the reference mixture, indicating that replacing 30% of PC with LFS did not significantly affect ultrasonic wave propagation. No direct relationship was observed between water retention (Table 6) and UPV, suggesting that water retention influences stiffness indirectly through its interaction with entrained air and the evolving binder structure rather than as an isolated parameter [29]. Taken together, the limited evolution of UPV in the LFS-containing AD mortars is consistent with the lower reactivity of LFS in the polymer-modified system. Thus, it

indicates that the development of the hardened matrix was governed primarily by the characteristics of the binder environment rather than by PC content alone [103].

While Fig. 8 highlights the overall relationship between matrix continuity and bond strength performance, a clearer understanding of the influence of exposure time on each property individually can be obtained by examining the evolution of their residuals. Fig. 9 presents the residual ratios for bond strength (Fig. 9a) and UPV (Fig. 9b).

Regarding bond strength (Fig. 9a), lime-based mortars generally showed residual values above one, including the LFS-based ones, indicating consistent improvement from 90 to 180 days. The steady increase in bond strength reflects progressive interfacial consolidation [16,102]. Mortars combining LFS with AD exhibited residual values below 1, indicating that bond strength did not increase over time. This reduced time sensitivity aligns with their higher w/b ratios (Table 4) and increased entrained air content (Table 6), which contribute to lower matrix compactness, consistent with the water absorption by capillarity and dynamic modulus measurements at 28 days (Fig. 7). Additionally, the replacement of 30 wt% of PC with LFS reduced the clinker content and, together with the absence of AL, may have limited continued binder development during outdoor exposure, resulting in less pronounced late-age matrix development. Thus, this may explain why the AD-LFS and AD-MK-LFS mortars did not exhibit the bond strength gains observed in the lime-based systems (Fig. 8). Similar trends have been reported for slag-containing systems by Henríquez et al. [9] and Fang et al. [22].

A similar pattern was observed for UPV (Fig. 9b), in which lime-based mixtures consistently exhibited residual values above 1 (except for AL-REF), indicating progressive stiffening with increasing exposure time. These results support the well-known sensitivity of UPV to solid matrix densification processes [104,105]. Overall, the residual UPV values were generally consistent with the corresponding bond strength evolution, particularly for the LFS-containing mortars. Small differences between the residual ratios of the two properties were observed for some mixtures, indicating that UPV and bond strength do not necessarily evolve proportionally during outdoor exposure because they are sensitive to different aspects of matrix and interfacial development. For the AD-based systems, residual values remained close to 1, except for AD-MK, which showed a more noticeable reduction. Despite their lower nominal UPV than lime-based mortars, the 180/90 ratios close to 1 indicate that prolonged exposure did not significantly impair pulse velocity. The additional 90 days of outdoor exposure did not promote relevant leaching or internal degradation. Furthermore, the carbonation observed during this period can be considered beneficial in the current context, as it mainly affects the render mortar's superficial zone and contributes to matrix consolidation [55]. Since the studied materials are non-structural rendering mortars without embedded reinforcement, the associated decrease in alkalinity does not pose a corrosion risk. Instead, carbonation may improve surface consolidation and interfacial performance [40,53].

The extended exposure period generally promoted progressive microstructural stabilization, reflected by stable or slightly increased UPV values and the concurrent evolution of bond strength in most mixtures. This effect was particularly evident for the lime-based mortars, whereas the AD-based mixtures maintained relatively stable UPV and bond strength between 90 and 180 days, with only minor reductions observed in two formulations. This behavior indicates that, despite their initially more porous structure, the temperature and moisture conditions during outdoor exposure favored the continued evolution of the binder phases, contributing to the gradual development of the hardened matrix.

After establishing how each property evolved over time, the next stage was to conduct linear regression analyses separately for mortars with and without LFS at 90 and 180 days (Fig. 10).

At 90 days, both groups exhibited a moderate positive correlation between UPV and bond strength, with coefficients of determination (R^2)

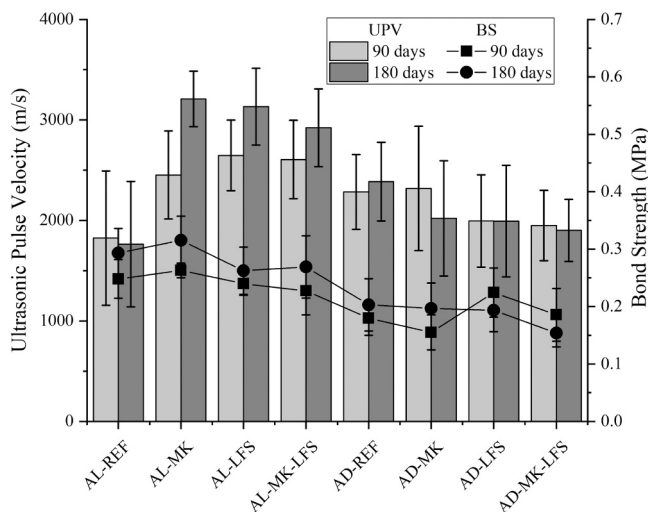


Fig. 8. Ultrasonic pulse velocity (UPV) and bond strength of mortars at 90 and 180 days of outdoor exposure.

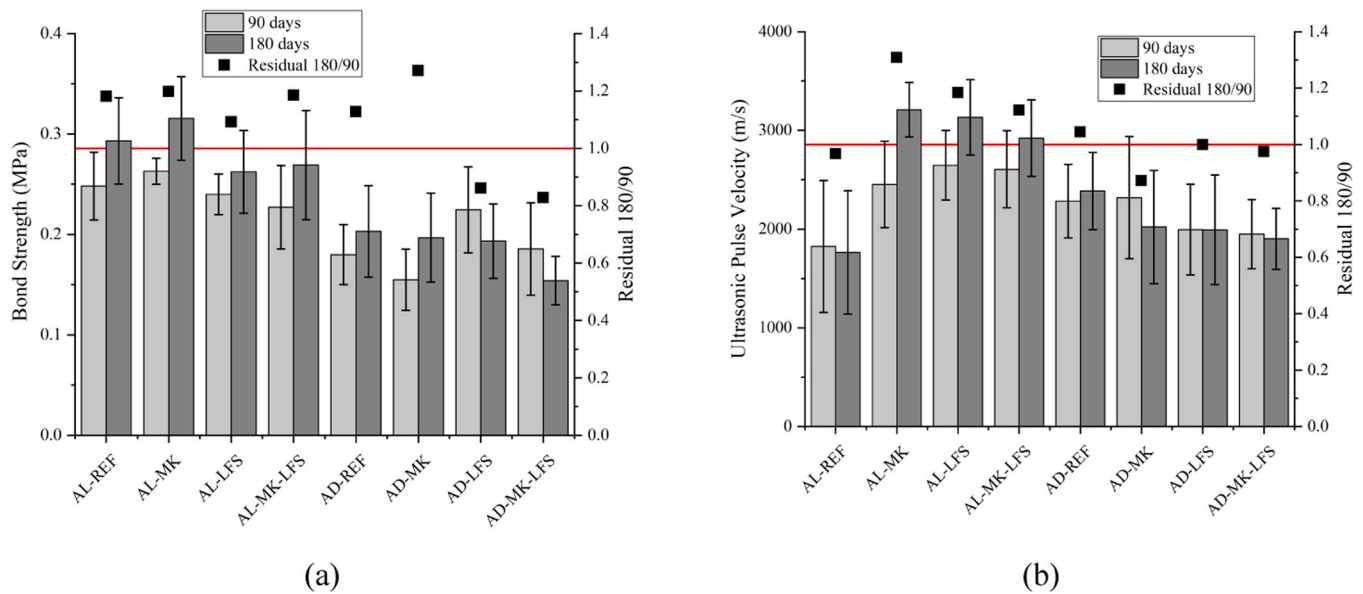


Fig. 9. (a) Bond strength at 90 and 180 days and corresponding residual ratio (180/90); (b) UPV at 90 and 180 days and corresponding residual ratio (180/90) for mortars exposed to natural outdoor conditions.

of 0.64 for mortars without LFS and 0.62 for mortars with LFS. These results indicate that, at early exposure ages, UPV was reasonably associated with bond strength performance, regardless of slag incorporation. At 180 days, mortars with LFS exhibited a strong correlation ($R^2 = 0.86$), suggesting that the development of long-term bonding in slag-containing systems is primarily driven by the gradual development of the solid matrix. This behavior confirms that mortars containing LFS hydrate more slowly, resulting in a stronger correlation between UPV and bond strength as matrix consolidation progresses. Mortars without LFS presented a lower coefficient of determination ($R^2 = 0.44$), indicating a weak correlation at 180 days. This may be associated with greater macro- and microstructural heterogeneity resulting from more rapid and less uniform hydration processes, which reduced the direct proportionality between UPV and bond strength. This result suggests that UPV monitoring is a potential practical tool for assessing the long-term bond strength performance of rendering mortars incorporating industrial by-products.

4.3. Thermogravimetric analysis (TGA) of mortars

To better understand the bond strength results observed in the panels after 180 days of natural outdoor exposure, thermogravimetric analyses (TGA/DTG) were conducted on samples from the eight mortar series. Fig. 11 presents the TG/DTG curves, while Table 7 summarizes the quantitative mass losses measured over the selected temperature intervals, together with the determined Ca(OH)_2 and CaCO_3 contents. The calculated Ca(OH)_2 contents are apparent values because brucite dehydroxylation occurs close to the 400 °C boundary adopted in this study. Consequently, a minor contribution from Mg(OH)_2 to the selected integration interval cannot be completely excluded [106,107]. The quantitative results provide a basis for interpreting the relationship between hydration, carbonation, bond strength development, and microstructural evolution during outdoor exposure.

Among the lime-based mortars, AL-MK exhibited the highest mass losses in all evaluated temperature intervals, reaching 4.73% (100–400 °C), 3.42% (400–600 °C), and 16.24% (600–900 °C). The corresponding calculated Ca(OH)_2 and CaCO_3 contents were 14.06% and 36.93 wt%, respectively. The higher mass loss measured in the 100–400 °C interval suggests a greater amount of hydrated products and retained water in the lime-rich binder system, consistent with the more prominent portlandite peaks observed in the XRD patterns (Fig. 2). Although Mg(OH)_2

may also contribute to the mass loss in this temperature interval, the similar values observed for the REF and LFS mortars indicate that its contribution was limited under the conditions investigated [60,108].

The dehydroxylation and decarbonation events further confirm the distinctive behavior of AL-MK. Compared with AL-REF, AL-LFS, and AL-MK-LFS, which exhibited calculated Ca(OH)_2 contents ranging from 5.69 to 6.57 wt%, AL-MK showed the highest calculated Ca(OH)_2 content and the most pronounced decarbonation step, indicating greater development of hydrated and carbonated phases. These quantitative results are consistent with the microstructural development associated with MK incorporation reported in the literature [21]. The increased decarbonation loss also suggests the coexistence of primary carbonates associated with the AL particles and secondary calcite formed during curing and outdoor carbonation. In comparison, AL-LFS and AL-MK-LFS exhibited intermediate decarbonation losses (7.25% and 7.97%, respectively), corresponding to calculated CaCO_3 contents of 16.49 and 18.12 wt%, indicating a moderate contribution of LFS to carbonate development within the blended matrix.

The reference mortar (AL-REF) displayed a distinct portlandite peak and moderate carbonate decomposition, indicating that some Ca(OH)_2 supplied by AL and PC remains unreacted after 180 days. The calculated Ca(OH)_2 and CaCO_3 contents for AL-REF were 5.69 and 11.27 wt%, respectively, confirming that PC and AL provide an alkaline environment rich in Ca(OH)_2 that remains available throughout curing. AL-MK exhibited substantially higher dehydration and decarbonation losses than AL-REF, indicating greater development of hydrated and carbonated phases. Accordingly, this is consistent with the progressive formation of C-(A)-S-H during curing, leading to a more consolidated matrix. As a result, AL-MK exhibited higher UPV values and continuous improvements in bond strength, in agreement with previous studies on MK-rich systems [87,92].

The AL mortars containing LFS (AL-LFS and AL-MK-LFS) exhibited intermediate thermal response. The TGA results showed total mass losses of 11.06% and 12.04%, respectively, which are intermediate between those of AL-REF (8.68%) and AL-MK (24.67%). The calculated CaCO_3 contents (16.49 and 18.12 wt%) were also higher than that of AL-REF (11.27 wt%) but lower than that of AL-MK (36.93 wt%). The increased decarbonation loss may reflect the coexistence of primary carbonates associated with the AL and LFS particles and secondary calcite formed during curing and outdoor carbonation. These findings indicate that the formation of hydrated and carbonated phases was less

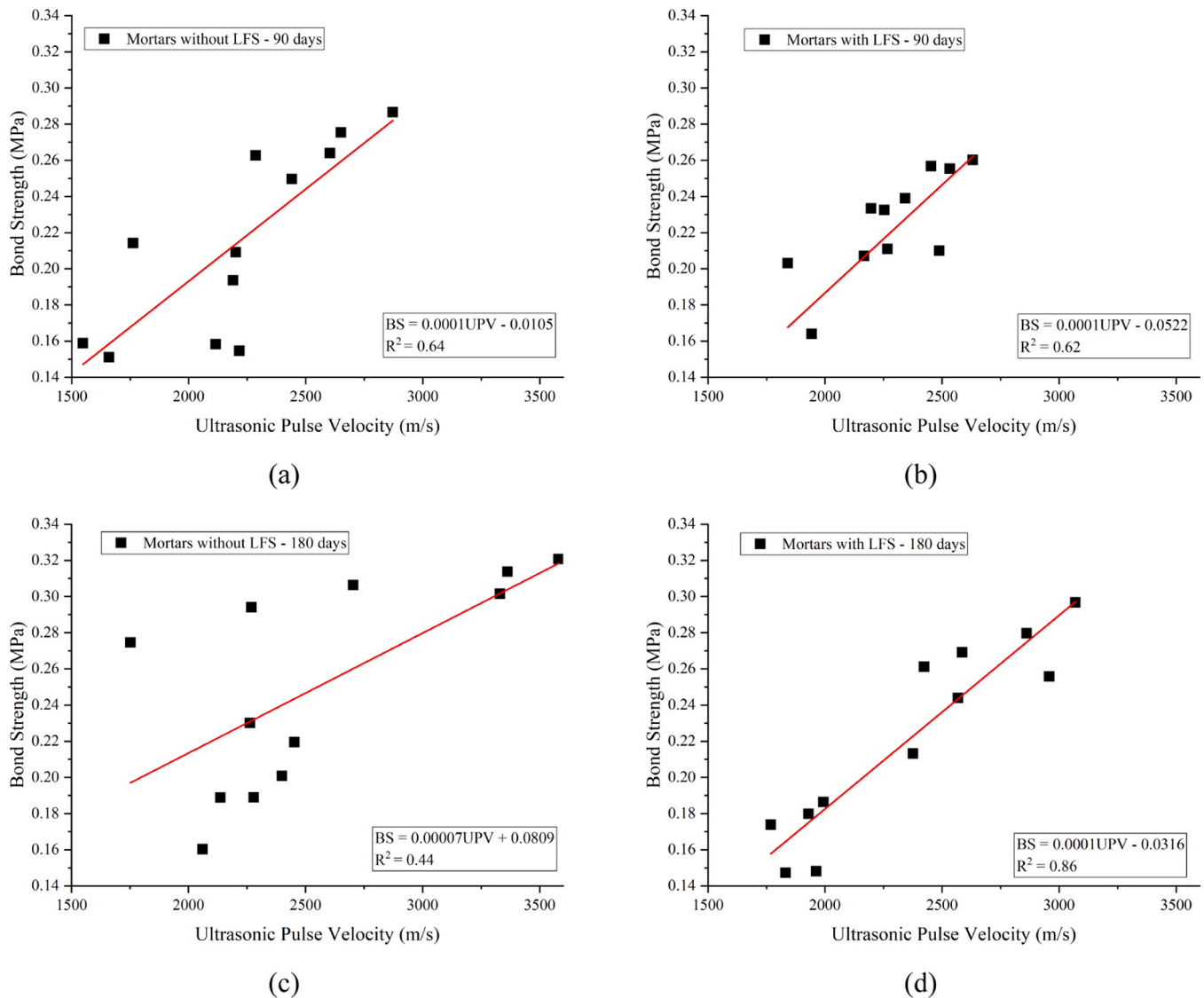


Fig. 10. Correlation between ultrasonic pulse velocity (UPV) and bond strength for rendering mortars exposed to natural weathering: (a) without LFS – 90 days; (b) with LFS – 90 days; (c) without LFS – 180 days; and (d) with LFS – 180 days. Linear regression lines, equations, and R^2 values highlight the influence of slag incorporation and exposure time on the relationship between microstructural stiffness and bond strength performance.

extensive than in the AL-MK mortar after 180 days of outdoor exposure.

Although LFS contains mayenite ($C_{12}A_7$), which may accelerate early-age reactions in cementitious systems [109], this effect appears to be limited during prolonged outdoor exposure. The predominantly crystalline nature of the air-cooled slag and the slower pozzolanic reactions of MK govern the long-term hydration behavior [21,24,61]. The DTG curves still exhibit noticeable hydroxide-related peaks, consistent with the persistence of Mg-bearing hydroxides from the slag. Similar behavior was reported by Yong et al. [110] for alumina-rich LFS systems, although in the present study it is mainly attributed to slag crystallinity and cooling history rather than to high Al_2O_3 content.

The AL-MK-LFS mixture also exhibited higher bound-water content below 400 °C and intermediate carbonate losses. The thermal results indicate that the Ca- and Mg-bearing phases of LFS progressively interacted with the calcium hydroxide supplied by PC and AL, as well as with the reactive aluminosilicates from MK, thereby promoting additional hydrate formation while maintaining moderate carbonation. Consequently, the blended mixture exhibited intermediate hydration and carbonation behavior [15]. This gradual microstructural development is also consistent with the progressive increase in bond strength observed between 90 and 180 days (Fig. 8).

For the AD series, the TGA and DTG curves exhibited very similar profiles, consistent with the quantitative results summarized in Table 7. Total mass losses ranged from 5.29% to 6.89%, while the calculated $Ca(OH)_2$ contents ranged from 4.18 to 4.62 wt% and the calculated $CaCO_3$ contents from 7.52 to 10.22 wt%, indicating only limited quantitative differences among the four mixtures. These mass losses, associated with C–S–H-type products and ettringite (100–400 °C), portlandite decomposition (400–600 °C), and carbonate decarbonation (600–900 °C), were relatively limited and significantly overlapped across all mixtures. Compared with the AL-based mortars, the lower total mass losses of the AD series indicate less extensive development of hydrated and carbonated phases after 180 days of outdoor exposure. In the absence of AL, the phase composition after 180 days of outdoor exposure is primarily determined by PC hydration and surface carbonation, and the addition of MK or LFS did not substantially alter the overall amounts of hydrated or carbonated phases formed.

Unlike AL-based mortars, the AD series exhibited only minor variations in the calculated $Ca(OH)_2$ and $CaCO_3$ contents and maintained similar DTG profiles across all mixtures. These results suggest that the pozzolanic reaction of MK and the hydration of LFS were less extensive in these systems than in the AL-based mortars. The water-retaining AD

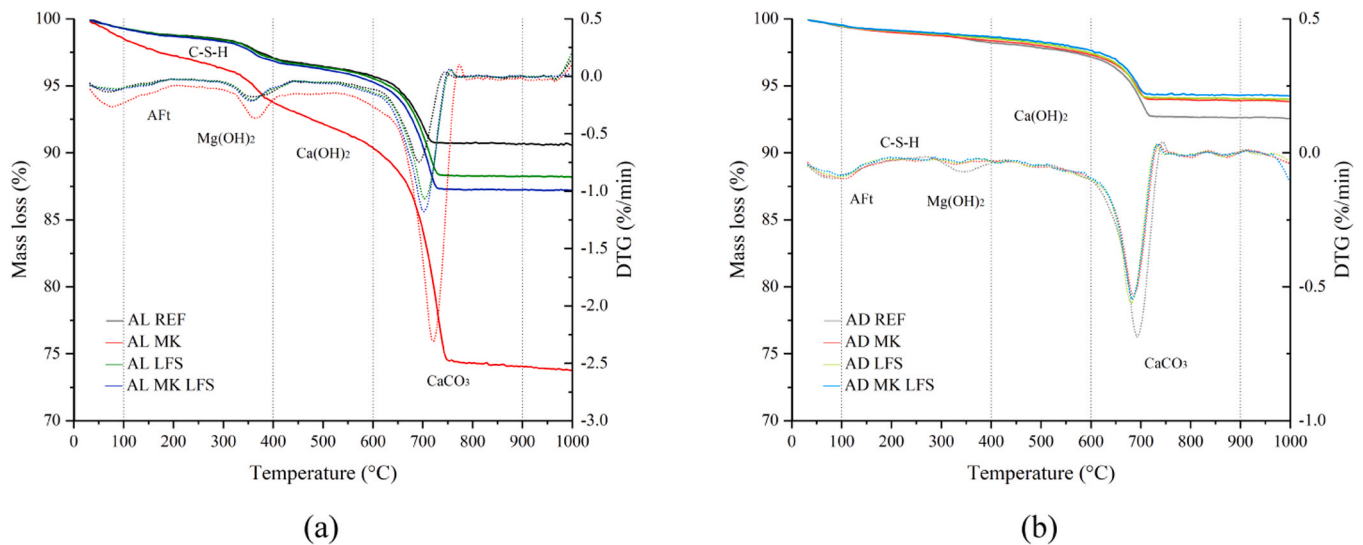


Fig. 11. Thermogravimetric (TGA) and derivative (DTG) curves of mortars after 180 days of natural outdoor exposure: (a) lime-based mortars and (b) AD-containing mortars. Main thermal events corresponding to dehydration of hydration products, dehydroxylation of hydroxides, and decarbonation of carbonate phases.

Table 7

Quantitative thermogravimetric results of the mortars after 180 days of outdoor exposure. Mass losses were determined over the selected temperature intervals, while the Ca(OH)_2 and CaCO_3 contents were calculated from the dehydroxylation (400–600 °C) and decarbonation (600–900 °C) intervals, respectively.

Series	TGA parameter					
	100–400 °C	400–600 °C	Calculated Ca(OH)_2 (wt%)	600–900 °C	Calculated CaCO_3 (wt%)	Total mass loss (20–1000 °C)
AL-REF	2.22	1.38	5.69	4.96	11.27	8.68
AL-MK	4.73	3.42	14.06	16.24	36.93	24.67
AL-LFS	2.27	1.49	6.13	7.25	16.49	11.06
AL-MK-LFS	2.42	1.60	6.57	7.97	18.12	12.04
AD-REF	1.23	1.09	4.48	4.49	10.22	6.89
AD-MK	1.10	1.09	4.48	3.40	7.73	5.65
AD-LFS	0.96	1.12	4.62	3.37	7.66	5.51
AD-MK-LFS	0.90	1.02	4.18	3.31	7.52	5.29

likely plays a key role in controlling moisture transport and microstructural development, thereby moderating differences in hydration behavior associated with the incorporation of SCMs. Similar observations were reported by Souza et al. [29], who found that cellulose-based water-retaining ADs primarily control initial moisture retention. Therefore, their influence appears to be more closely associated with the overall microstructural development of the mortar than with the formation of additional binder phases.

The persistence of hydroxide-related peaks together with the comparatively low calculated CaCO_3 contents and decarbonation mass losses reinforce the previously observed limited participation of LFS in hydration. These observations align with the known features of air-cooled low-calcium slag, including its heterogeneous mineralogy and the presence of slow-hydration phases such as $\gamma\text{-C}_2\text{S}$ and periclase [9,11,16]. Without lime, the combined effect of slag characteristics and increased porosity inhibits the formation of hydration products such as C–S–H, and carbonate products, resulting in limited development of a continuous solid skeleton. This behavior differs from systems in which low-calcium slag directly replaces lime or is chemically activated, which typically exhibit enhanced hydration and carbonate formation [12,24,30], underscoring the strong influence of the binder environment and activation level on slag reactivity.

4.4. Microstructural analysis by scanning electron microscopy (SEM)

Figs. 12 and 13 show backscattered SEM micrographs of the mortars after 180 days of natural exposure, highlighting the influence of LFS and

MK on particle packing, overall microstructural features, and the contact zones between the paste and aggregates.

The reference mix (AL-REF) exhibited a continuous lime–cement matrix surrounding the sand grains, with isolated voids and microcracks, which is consistent with its moderate stiffness and bond strength. The observed microstructure is typical of lime–cement mortars exposed to natural carbonation cycles, where partial densification occurs at the surface while the interior remains more porous. Compared with AL-REF, the AL-MK micrograph does not show marked morphological differences. However, the SEM observations, together with the lower capillary water absorption (Fig. 7a), higher UPV, greater 180-day bond strength (Fig. 9), and the TGA results, suggest a more consolidated matrix after prolonged outdoor exposure [86,87].

Introducing LFS fundamentally changed the microstructural pattern. In AL-LFS, the matrix became more irregular and discontinuous, with granular clusters and localized regions with visible voids adjacent to slag particles. These features are consistent with the observed heterogeneous behavior of LFS [12,15]. Although AL-LFS exhibited a high fresh density, the higher capillary water absorption measured at 28 days (Fig. 7a), together with the limited UPV development and lower bond strength observed after outdoor exposure (Fig. 9), indicates that this did not translate into a more consolidated hardened matrix. This interpretation is supported by the numerous voids and the less continuous ITZ observed by SEM. Similar relationships between interfacial microstructure and bond performance in slag-containing cementitious systems have also been reported in adhesive mortars [111].

The combined presence of MK and LFS (AL-MK-LFS) resulted in a

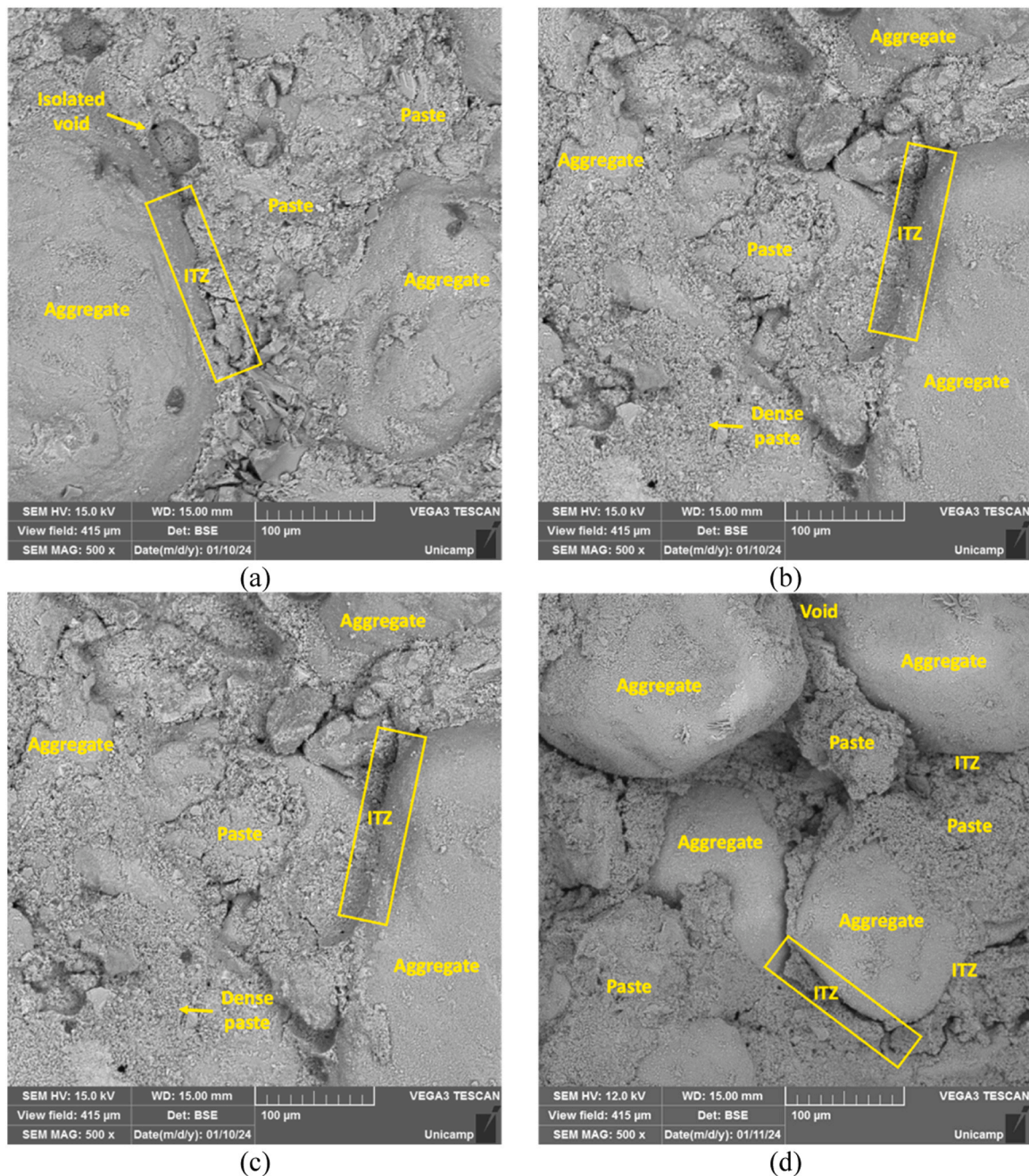


Fig. 12. SEM micrographs of lime-based mortars at 500 × magnification, obtained after 180 days of natural outdoor exposure, showing (a) AL-REF, the reference mortar with air lime; (b) AL-MK, the mortar containing metakaolin; (c) AL-LFS, the mortar containing ladle furnace slag; and (d) AL-MK-LFS.

denser yet notably more heterogeneous microstructure than AL-LFS. While MK was expected to promote localized densification [38], SEM images qualitatively indicate that larger voids remain around slag particles, suggesting that LFS may limit the full benefits of MK. Similar incompatibilities between MK and low-reactivity slags have been observed in mortars in which slag hydration is insufficient to support pozzolanic densification [21]. This uneven consolidation supports the experimentally identified intermediate behavior, particularly the moderate capillary absorption (Fig. 7a), intermediate UPV evolution, and lower bond strength (Fig. 9) compared with AL-MK.

Fig. 13 shows SEM micrographs of mortars containing the water-retaining AD at 500 × magnification.

All AD mortars showed a more open microstructure than their AL counterparts, reflecting their higher inherent air entrainment and water

demand. AD-REF exhibited numerous rounded voids and thin paste bridges, consistent with the lower bulk density of this mortar and with its comparatively higher capillary water absorption observed during laboratory characterization (Fig. 7a). This trend reflects the results from cellulose-based water-retaining ADs, which increase internal curing but also raise entrained-air levels and hinder densification [29]. MK partially compensates for this effect (AD-MK); however, void-rich regions remain visible throughout the matrix. This pattern matches the limited long-term increase in UPV of AD-MK. Although MK is expected to promote matrix development [87], prolonged exposure combined with retained moisture can compromise matrix continuity, consistent with the decrease in UPV observed between 90 and 180 days.

The presence of LFS in AD mortars (AD-LFS) increased microstructural instability. AD-LFS exhibited the most heterogeneous and

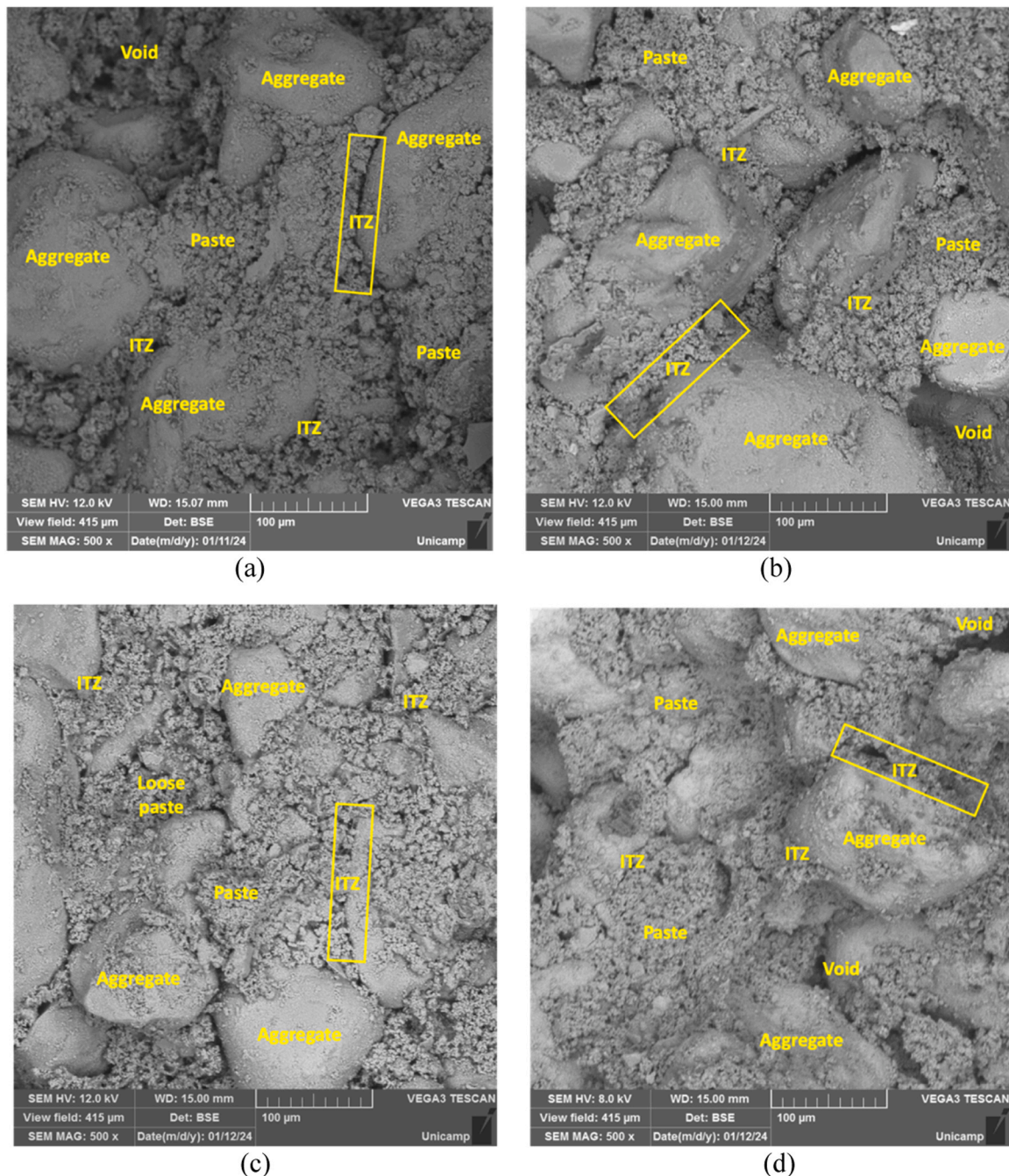


Fig. 13. SEM micrographs of AD-based mortars at 500 × magnification, showing (a) AD-REF, the reference mortar with water-retaining admixture; (b) AD-MK, the mortar containing metakaolin; (c) AD-LFS, the mortar containing ladle furnace slag; and (d) AD-MK-LFS, the mortar containing both metakaolin and ladle furnace slag.

discontinuous microstructure among all mixes: coarse slag particles were surrounded by a loose, porous paste with poorly defined interfaces. This morphology matches the behavior of LFS in systems with high water retention, where these effects reinforce one another [24]. The observed microstructure is consistent with the experimentally measured low stiffness, stagnant UPV development, and reduced bond strength. Similar observations have been reported for alkali-activated slag-based mortars, in which bond performance was strongly influenced by the microstructural development of the interfacial transition zone between the mortar and the substrate [112]. A similar microstructural pattern was observed for AD-MK-LFS. The matrix remained discontinuous around the slag particles, with weak interfacial bonding, confirming that MK cannot counteract the microstructural discontinuity generated by

LFS under AD-rich conditions. Silva et al. [21] reported similar findings when LFS hydration remained insufficient to support uniform matrix consolidation. Consequently, the experimental behavior of AD-MK-LFS was characterized by limited development of bond strength (Fig. 9a) and limited UPV evolution (Fig. 9b), despite the incorporation of MK.

Taken together, these results indicate that the binder system governs the long-term behavior of LFS-containing mortars under natural outdoor exposure. Three characteristic behaviors were identified. In lime-rich systems, the presence of Ca(OH)_2 provided a favorable alkaline environment, allowing the gradual interaction of LFS and promoting the development of bond strength over time. In contrast, systems containing AD exhibited higher water and air contents, leading to a less compact microstructure and limiting the contribution of LFS. Mixtures containing

both MK and LFS exhibited an intermediate response, reflecting the balance between matrix development promoted by MK and the limited reactivity of LFS. The combined interpretation of bond strength, capillary water absorption, UPV, TGA, and complementary SEM observations provides no evidence of deleterious expansion or instability associated with reactive CaO- and MgO-bearing phases under the outdoor exposure conditions investigated. These observations highlight the central role of the binder environment in governing LFS performance.

5. Conclusion

This study investigated the development of bond strength in rendering mortars incorporating LFS under natural outdoor exposure by integrating mechanical testing with UPV, TGA, and SEM analyses. The results demonstrate that the long-term performance of rendering mortars containing LFS is strongly governed by the binder composition and its interaction with environmental exposure rather than by the intrinsic reactivity of the slag.

Lime-based mortars incorporating LFS maintained bond performance comparable to the reference mixture after prolonged outdoor exposure, whereas mortars containing water-retaining admixtures consistently exhibited lower bond strength. The combined interpretation of UPV, TGA, and SEM results showed that the long-term behavior of LFS was dominated by filler effects and the carbonation of Ca-rich phases, with limited evidence of significant hydraulic reactivity. Notably, UPV closely followed the evolution of bond strength, particularly after 180 days of outdoor exposure, highlighting its potential as a complementary non-destructive technique for monitoring interfacial development.

Although this study was limited to a single source of air-cooled low-reactivity slag (LFS) and a fixed processing condition, it provides new insights into how binder composition influences LFS behavior in rendering mortars under real-world environmental conditions. By integrating laboratory characterization with extended field exposure, this work contributes to bridging the gap between controlled experimental results and actual in-service performance. Therefore, the results demonstrate that the effectiveness of LFS depends on the combined influence of binder composition, w/b ratio, entrained air content, and the resulting microstructural development rather than on any single parameter in isolation. These findings support the use of air-cooled LFS as a practical approach for clinker reduction and waste valorization within a circular economy framework.

CRediT authorship contribution statement

Riccardo Maddalena: Writing – review & editing, Validation, Supervision, Methodology, Formal analysis, Data curation. **Rafaela de Oliveira Amaral:** Writing – review & editing, Software, Methodology, Formal analysis, Data curation. **Gisleiva Cristina dos Santos Ferreira:** Writing – review & editing, Visualization, Validation, Supervision, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Eduardo Freire Mariano:** Writing – review & editing, Software, Resources, Methodology, Investigation, Formal analysis, Data curation. **Tayná Fracão da Silva:** Writing – review & editing, Visualization, Software, Resources, Investigation, Formal analysis, Data curation. **Everton de Freitas Cordova de Souza:** Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Resources, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

Data will be made available on request.

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