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Carbon sequestration and performance enhancement of lime mortar modified with natural additives for sustainable construction

ABSTRACT

This study investigated the potential of lime mortar modified with starchy extract and Terminaliachebula to enhance carbon capture and utilization (CCU) for sustainable construction applications. The modified mortars were evaluated using compressive strength testing, X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR), thermogravimetric analysis (TGA), and BET pore structure analysis to assess mechanical performance, carbonation behavior, and microstructural evolution. Results showed that starchy extract-modified mortar improved carbonation by 39.7% and mechanical performance by 15.2% due to enhanced pore refinement and moisture retention, reducing pore volume to 0.068 cc/g compared with the reference mortar (0.075 cc/g). Chebula-modified mortar exhibited the lowest pore volume (0.061 cc/g), indicating a denser microstructure formed through tannin-Ca²⁺ interactions, promoting cohesion, water retention, and formation of amorphous calcium carbonate (ACC). The combined additive mortar exhibited the highest pore volume (0.080 cc/g), creating an optimized pore network that enhanced CO₂ diffusion and maintained matrix stability, resulting in 35% higher compressive strength and 3.46% greater CCU efficiency than the reference mortar. Microstructural analysis confirmed enhanced calcite formation in starchy extract mortar, ACC-rich matrices in Chebula mortar, and balanced crystalline-amorphous carbonate phases in the combined system. The findings demonstrate that natural organic additives significantly improve carbonation, carbon sequestration, and mechanical performance of lime mortar for sustainable non-load-bearing construction applications.

Keywords: Lime mortar, carbon sequestration, carbon capture and utilization (CCU), sustainable material

1. INTRODUCTION

The continuous rise in atmospheric carbon dioxide (CO₂) concentration has become one of the most critical global environmental concerns. In 2025, the building and construction sector in India remained a major contributor to national carbon emissions due to rapid urbanization and large infrastructure demand. Although atmospheric CO₂ concentration is measured globally rather than by country, global levels reached about 425-426 ppm in 2025, representing one of the highest concentrations recorded [1]. Within India, the buildings and construction sector contribute roughly one-sixth of the nation's total greenhouse gas emissions, mainly due to the intensive use of carbon-intensive materials such as cement, steel, and bricks.

Cement production alone contributes approximately 6-7% of India's total CO₂ emissions [2], while the overall buildings sector accounts for nearly 25% of national emissions, including operational and embodied carbon from construction materials. The rapid expansion of urban infrastructure and housing demand continues to increase emissions from the construction industry, highlighting the need for low-carbon materials and sustainable construction technologies to reduce the sector's environmental impact.

To address this challenge, researchers have extensively explored alternatives to conventional Portland cement in order to reduce embodied carbon in construction materials. Various supplementary cementitious materials (SCMs) such as fly ash, ground granulated blast furnace slag, silica fume, mine tailings, red mud, and kaolin-based materials have been incorporated into concrete as partial replacements for cement [3,4]. These materials can improve mineral carbonation

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and reduce CO₂ emissions during cement production. However, although SCMs reduce emissions associated with manufacturing, they do not directly contribute to the capture of atmospheric CO₂ that has already been released into the environment.

In this context, lime has emerged as a promising alternative binder due to its comparatively lower embodied energy and its potential to reabsorb atmospheric CO₂ during its service life. Lime has been widely used as a primary building material in ancient constructions across many civilizations. Unlike Portland cement, lime-based materials harden mainly through the carbonation process, in which atmospheric CO₂ reacts with calcium hydroxide (Ca(OH)₂) to form calcium carbonate (CaCO₃) [5]. This mineralization process enables lime-based materials to act as potential carbon capture and utilization (CCU) systems [6]. The resulting carbonation products, including calcite and other polymorphic forms of calcium carbonate such as aragonite and vaterite, act as primary binding phases and contribute to the structural stability of lime mortars [7]. Furthermore, production of lime requires significantly lower calcination temperatures (approximately 900 °C) compared to Portland cement (around 1450 °C), resulting in reduced energy consumption and lower greenhouse gas emissions [7]. Although lime theoretically has the ability to reabsorb nearly the same amount of CO₂ released during its production, the actual sequestration capacity depends on several environmental and material-related factors, including relative humidity, temperature, pore structure, and exposure surface area, which influence CO₂ diffusion and carbonation depth [8,9].

Since ancient times, various organic additives have been incorporated into lime mortars to enhance their mechanical properties, durability, and workability. Historical and archaeological studies indicate the use of plant extracts, herbs, animal-based products, oils, and fermented organic materials in traditional construction. Examples include blood, animal fats, egg whites, tung oil, sticky rice, olive oil, and linseed oil [10,11]. In the Indian subcontinent, traditional building practices frequently incorporated fermented organic extracts derived from locally available plants and herbs to improve the performance of lime mortars and plasters [12,13]. Sticky rice is an organic additive for lime mortar systems that improves water retention, workability, pore refinement, and carbonation efficiency, leading to increased ambient CO₂ uptake through CaCO₃ production [14]. While traditional additives like Terminaliachebula, Terminaliabelirica, and amla

aid in CO₂ sequestration by providing bioactive compounds that enhance crystallisation, moisture retention, and microbial mineralisation, direct studies quantifying CO₂ capture in sticky rice-modified lime are still limited. However, the utilisation of a single additive often results in constraints: sticky rice enhance cohesion and reduce permeability at elevated dosages; chebula may impede setting due to interactions rich in tannins; belerica may modify pore connectivity; and amla may introduce an excessive level of organic acidity that impacts early carbonation [15]. A balanced multi-additive system provides synergistic benefits, reducing the drawbacks of individual additives while enhancing CO₂ absorption, carbonation efficiency, and long-term durability in sustainable lime mortars [16].

Despite the historical use and growing research interest in organic-modified lime mortars, significant knowledge gaps remain regarding the quantitative influence of organic additives on carbonation behavior and carbon sequestration capacity. In particular, the relationship between organic additive incorporation, pore structure development, carbonation kinetics, and overall CO₂ sequestration potential has not been systematically investigated. Understanding these interactions is essential for optimizing lime-based materials as sustainable construction systems capable of enhanced carbon capture.

Previous studies on lime-based materials have extensively explored the use of natural biopolymers and plant-derived additives, including sticky rice, proteins, herbal extracts, and oils, primarily to improve workability, durability, and mechanical performance. Sticky rice-derived polysaccharides have been shown to enhance moisture retention, pore structure refinement, and carbonation efficiency, whereas Terminaliachebula contains tannins and polyphenolic compounds that contribute to mineralization and durability enhancement. However, existing research has largely focused on these additives individually, and their combined effects on carbonation behaviour, carbon sequestration, microstructural evolution, and strength development remain unexplored. Therefore, the novelty of this study lies in investigating a hybrid bio-modified lime mortar incorporating both starchy extract and Terminaliachebula. The combined additive system is expected to provide synergistic benefits by improving moisture retention, promoting carbonation, facilitating mineral formation, and refining the pore network. Unlike previous studies that mainly emphasized mechanical and durability improvements, the present work quantitatively evaluates CO₂ sequestration and examines the

interrelationship between porosity, carbonation, and carbon capture, thereby contributing to the development of sustainable lime mortars for heritage conservation and low-carbon construction applications.

Most of the studies have focused on the effects of individual additives on workability, strength, and durability, whereas their contribution to carbonation enhancement and atmospheric CO₂ sequestration has received comparatively little attention. Moreover, the interaction between organic additives with pore structure development, carbonation, and carbon capture efficiency remain insufficiently understood. Existing investigations have generally examined sticky rice and traditional herbal additives separately, with limited information available on their combined influence on carbonation behaviour and microstructural evolution. In addition, few studies have quantitatively correlated pore characteristics, carbonation products, and CO₂ uptake using a comprehensive analytical approach. Therefore, further research is required to understand these interactions and to develop optimized lime-based materials with improved carbon sequestration potential for sustainable construction and heritage conservation applications.

The present study addresses the gaps by evaluating two organic additives in Indian built heritage construction: Terminaliachebula and starchy extract. These natural additives have historically been incorporated into lime-based materials to enhance durability and mechanical performance. This research investigates their incorporation into lime mortar and examines their influence on carbonation rate, carbon sequestration capacity, mechanical strength, and microstructural characteristics, with particular emphasis on the relationship between porosity and carbonation behavior. Carbonation in lime mortar is strongly controlled by porosity because it depends on the diffusion of CO₂, the supply of moisture, and the reaction space within the network of pores. CO₂ enters through pores that are linked together, dissolves in the water in the pores, and mixes with Ca(OH)₂ to form CaCO₃. If pores are too small or closed, CO₂ can't get through and carbonation takes longer. If the porosity is too high, CO₂ may be able to move around easily, but a weak binder and poor moisture retention can make carbonate formation less effective. Therefore, an optimal pore structure is essential. During carbonation, newly formed CaCO₃ gradually fills pores, refines the microstructure, reduces porosity, and changes further CO₂ transport, making porosity and carbonation interdependent. The experimental methodology includes a combination of physical

testing and advanced analytical techniques. Carbonation behavior was monitored using weight change profile analysis, while mechanical performance was evaluated through compressive strength tests. Mineralogical and molecular characteristics were analyzed using X-ray diffraction (XRD) and Fourier Transform Infrared spectroscopy (FTIR), respectively. In addition, gas adsorption analysis was conducted to evaluate pore structure, and thermogravimetric analysis (TGA) was used to determine composition and carbon capture capacity. The findings aim to provide a quantitative understanding of how these organic additives influence carbonation mechanisms and improve the carbon sequestration potential of lime-based construction materials.

2. MATERIALS AND METHODS

2.1. Materials

Hydraulic lime was selected as the binder for the preparation of mortar specimens due to its superior workability and compatibility with traditional masonry substrates. The chemical composition of the hydraulic lime was determined using X-ray fluorescence (XRF) analysis. The lime was classified based on chemical compound as per IS 712-1984 as illustrated in Table 1 [17]. Natural river sand was used as the fine aggregate, having a specific gravity of 2.67. Prior to mixing, the sand was sieved through a 2.36 mm IS sieve to ensure uniform particle size distribution and to remove oversized particles, thereby improving packing density and consistency in the mortar matrix. Extracts of Terminaliachebula along with sticky starchy extract derived from *Oryza sativa*, were utilized as natural modifiers.

Table 1. Chemical composition of hydraulic lime

Minerals (%)	CaO	MgO	SiO ₂	Fe ₂ O ₃	Al ₂ O ₃	LOI
Lime	66.8	1.098	17.10	4.77	3.85	6.38

2.2. Material characterization

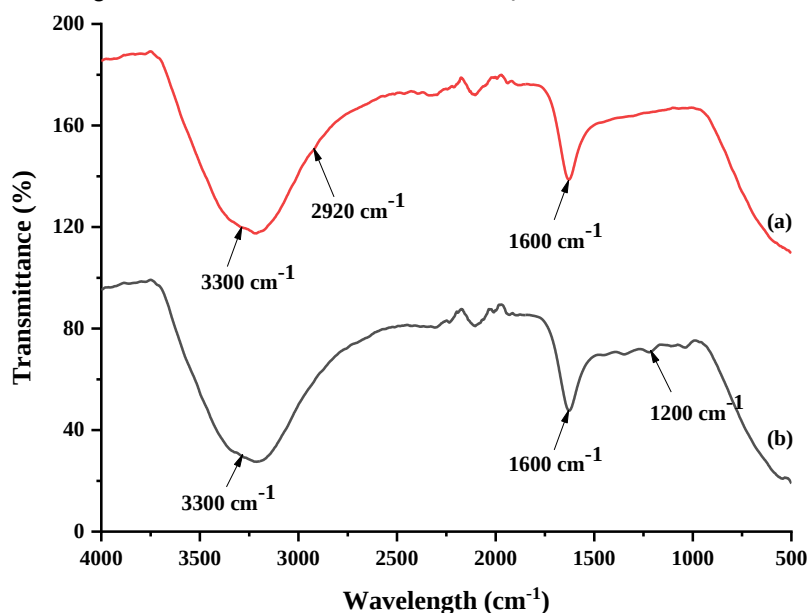
Terminaliachebula and starchy extract were selected as natural organic admixtures due to their potential to enhance carbonation, carbon sequestration, and microstructural development in lime mortar. Terminaliachebula contains bioactive compounds such as tannins, polyphenols, gallic acid, chebulagic acid, chebulinic acid, and flavonoids, which can interact with calcium ions and promote mineralization processes. Starchy extract obtained from cooked rice is rich in polysaccharides, primarily amylose and amylopectin, which improve moisture retention, refine pore structure, and facilitate carbonation by

enhancing CO₂ diffusion and calcium carbonate formation.

FTIR analysis confirmed the presence of functional groups associated with these compounds. Both extracts exhibited a broad absorption band in the range of 3200-3500 cm⁻¹, corresponding to O-H stretching vibrations of hydroxyl groups. The higher intensity observed in the starchy extract indicates the abundance of polysaccharides, which contribute to moisture retention and create favourable conditions for carbonation. The absorption band near 2920 cm⁻¹ corresponds to C-H stretching vibrations, confirming the carbohydrate-rich nature of the starchy extract. Terminaliachebula showed prominent bands in the regions of 1600-1700 cm⁻¹,

attributed to carbonyl, aromatic, and phenolic functional groups associated with tannins and polyphenols. In addition, peaks between 1200cm⁻¹ correspond to C-O and C-O-C stretching vibrations, indicating the presence of polysaccharide structures and phenolic compounds.

The FTIR results demonstrate that the starchy extract is predominantly rich in polysaccharides, while Terminaliachebula contains significant amounts of tannins and polyphenolic compounds. These functional groups collectively enhance moisture retention, promote calcium carbonate nucleation, refine pore structure, and facilitate carbonation reactions, thereby contributing to improved carbon sequestration and microstructural development in lime mortar.



2.3. Preparation of organic admixture

Terminaliachebula extract is prepared by adding 50 g of powdered Terminaliachebula in 1000 mL of water and allowed to ferment for 7 days at room temperature. The resulting fermented solution was filtered and used as the organic admixture.

The starchy extract was prepared by boiling rice in water at a rice-to-water ratio of 1:5 until complete gelatinization was achieved. The cooked mixture was filtered to separate the starch-rich liquid fraction. The extracted liquid was subsequently fermented for 24 h under ambient conditions before use. The starchy extract primarily contained soluble starch fractions composed of amylose and amylopectin released during the cooking process.

Based on preliminary trials and traditional practice, Terminaliachebula extract and starchy

extract were incorporated at dosages of 1% and 2%, respectively, by weight of lime. Three modified mortar mixtures were prepared: mortar containing only Terminaliachebula extract (LC), mortar containing only starchy extract (LR), and mortar containing a combination of both Terminaliachebula and starchy extract (MM). The combined system was designed to utilize the complementary effects of tannin-rich bioactive compounds and polysaccharides to enhance carbonation, carbon sequestration, and microstructural refinement in lime mortar.

2.4. Mortar preparation

The lime and sand were mixed thoroughly to achieve a uniform blend before the addition of water and the designated organic admixtures. The mixing liquid, consisting of water and the required dosage of organic extract, was added gradually and mixed until a homogeneous and workable

mortar was obtained. Table 2 illustrates the mix proportion of materials. The consistency and workability of the fresh mortar mixes were determined in accordance with IS 6932:1973 (Part VIII) [18]. The prepared mortars were then cast into

moulds and kept under laboratory conditions for 24 h. After demoulding, the specimens were cured at room temperature until the designated testing periods. Table 3 explains the number of specimens used for experimental test.

Table 2. Sample description

Sample code	Lime (%)	Sand (%)	W/B ratio	Terminalia chebula (%)	Starchy Extract (%)	Binder Ratio
LM	100	300	0.75	0	0	1:3
LC	100	300	0.75	1	0	1:3
LR	100	300	0.75	0	2	1:3
MM	100	300	0.75	1	2	1:3

2.5. Experimental methods

2.5.1. Mechanical behaviour

The mechanical behaviour of the prepared mortar samples was evaluated by conducting compressive strength test at 14, 28 and 56 days as per Indian standard IS 6932 - 2009 (Part VII) [19]. The low-capacity universal testing machine with loading rate of 0.5mm/min was used for testing. To minimize the standard deviation, values of six specimens of each mix was reported.

2.5.2. Weight loss profile

The carbonation behavior of the prepared mortar specimens was evaluated by monitoring changes in specimen weight over time. After demoulding, the specimens were stored under ambient laboratory conditions at room temperature (27 ± 2 °C) and relative humidity of $65 \pm 5\%$. The specimens were placed on open environment to allow free circulation of air and continuous exposure to atmospheric CO₂ throughout the carbonation period.

The weight of each specimen was measured using an electronic weighing balance with a sensitivity of 0.001 g at intervals of 3, 7, 14, 21, 28, 42, and 56 days after the initiation of carbonation. Before each measurement, loose surface particles were carefully removed to ensure consistency and accuracy. The changes in specimen weight during the exposure period were used as an indirect measure of carbonation progress, since the reaction between calcium hydroxide [Ca(OH)₂] and atmospheric CO₂ leads to the formation of calcium carbonate (CaCO₃). The carbonation rate and CO₂ sequestration capacity of the mortars was subsequently assessed from the weight change profile over the 56-day carbonation period [20].

2.5.3. Quantification of CO₂

The fermentation behavior of Terminalia chebula extract, starchy extract, and their combination was evaluated by monitoring the concentration of carbon dioxide (CO₂) released during fermentation. Fermentation promotes the breakdown of complex carbohydrates into simple

sugars, which are subsequently metabolized by microorganisms to produce ethanol and CO₂. The CO₂ concentration was measured using a wireless infrared CO₂ sensor (PASCO PS-3208).

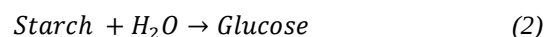
The sensor was placed in an airtight position over the fermentation vessel at 24 h intervals, and the CO₂ concentration (ppm) was recorded after 5 min once the readings stabilized. Measurements were continued throughout the fermentation period. The recorded CO₂ concentration was used as an indirect indicator of fermentation activity and the availability of fermentable substrates in the organic admixtures.

The biochemical reaction occurring during fermentation are illustrated in equation (1) - (3).

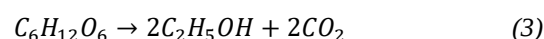
Hydrolysis of tannins in Terminalia chebula:



Hydrolysis of starch in starchy extract:



Fermentation of glucose:



The generated CO₂ can subsequently participate in the carbonation reaction of lime mortar (Equation 4):



The assumptions are as follows:

- Weight gain during exposure is primarily due to carbonation.
- Carbonation occurs uniformly throughout the exposed specimen.
- The stoichiometric conversion of absorbed CO₂ to CaCO₃ is complete.
- Moisture-related weight fluctuations are negligible compared with carbonation-induced mass gain.

2.5.4. Mineralogical analysis

X-ray diffraction (XRD) analysis was performed using a Bruker desktop diffractometer to determine

the mineralogical composition of the prepared mortar samples. The sample were powder and sieved through 63 microns were tested. The measurements were carried out using Cu K α radiation at a voltage of 9 kV and a current of 9 mA. The diffraction patterns were recorded over a 2θ range of 10° - 70° at a scanning rate of 1° min^{-1} . The obtained reflections were indexed using Diffrac Plus software (BrukerAXS) with reference to the JCPDS database. To quantitatively evaluate the carbonation products, the XRD patterns were subjected to Rietveld refinement using HighScore Plus software. The refinement was performed using the reference crystal structures of calcite (CaCO_3), vaterite (CaCO_3), and portlandite (Ca(OH)_2). A good fit was obtained between the experimental and calculated diffraction patterns, indicating accurate phase identification and quantification.

2.5.5. Infrared analysis

Fourier transform infrared spectroscopy (FTIR) was performed using a PerkinElmer 1000 spectrometer to identify the organic functional groups present in the hardened mortar samples. Spectra were recorded in the range of 400 - 4000 cm^{-1} using 32 scans at a resolution of 1 cm^{-1} . The measurements were carried out in attenuated total reflectance (ATR) mode. The analysis was done to identify the functional group present in the samples.

2.5.6. Morphological analysis

Scanning electron microscopy (SEM) was employed to examine the microstructure and morphology of the modified and reference lime mortar samples. SEM/EDS analysis was carried out using an FEI Quanta FEG 200 high-resolution scanning electron microscope. For analysis, a fractured mortar specimen of approximately 1 cm in

Table 3. Specimen description

Test	Specimen geometry (mm)	Sample code	No. of specimens	Curing days
Hardened state properties				
Compressive strength	$50 \times 50 \times 50$	LM, LC, LR, MM	3 each	14, 28, 56
Carbonation depth	$50 \times 50 \times 50$		3 each	3, 7, 14, 21, 28, 32, 56

3. RESULTS AND DISCUSSION

3.1. Chemical analysis of binder

The binder constituents, mainly CaO and MgO, account for nearly 68% of the composition, while clay minerals (SiO_2 , Al_2O_3 , and Fe_2O_3) constitute about 26%. These show that calcium and magnesium oxide carbonation is the main strength-building mechanism. However, little concentrations

size was sectioned from the cube and used as the representative sample. SEM images were acquired at multiple magnifications ranging from $500\times$ to $12,000\times$. Lower magnifications were used to examine the overall pore structure, pore connectivity, and matrix densification, whereas higher magnifications were employed to identify carbonation products, crystal morphology, and the distribution of carbonate phases within the mortar matrix.

2.5.7. Thermal analysis

Thermogravimetric analysis (TGA) was performed to evaluate the thermal decomposition behavior of the mineral phases present in the mortar samples. The analysis was conducted using a thermogravimetric analyzer (Netzsch STA 2500 Regulus) with Al_2O_3 crucibles under a purified air atmosphere over a temperature range of 30 - 1000°C at a heating rate of $20^\circ\text{C min}^{-1}$. The weight loss of the samples was recorded as a function of temperature.

2.5.8. Nitrogen adsorption analysis

The surface area and pore size distribution of hardened lime putty samples were determined using nitrogen adsorption-desorption analysis with a Quantachrome Autosorb iQ instrument. Approximately 1.5 g of 56-day cured samples ($<8 \text{ mm}$ size) were immersed in ethanol for 24 h to terminate hydration reactions. The samples were then dried in a vacuum oven at 100°C to remove residual solvent and moisture. Prior to testing, samples were degassed under vacuum at 40°C for 24 h. Nitrogen adsorption-desorption measurements were performed at 77.35 K . The specific surface area was calculated using the Brunauer-Emmett-Teller (BET) method, while pore size distribution was determined using the Barrett-Joyner-Halenda (BJH) method.

of clay minerals increase hydration processes, which strengthen the material along with carbonation.

3.2. Compressive strength

The compressive strength results (Figure 1) indicate that sample MM exhibited the highest mechanical strength at all curing ages, followed by LC and LR. At 14 days, the compressive strength

of LC and LR increased by 58% and 38%, respectively, compared to LM, while MM showed an increase of 88%. This trend continued up to 56 days, where MM exhibited a 35% increase in strength compared to LM, whereas LC and LR showed improvements of 23.8% and 15.2%,

respectively. The superior performance of MM can be attributed to the synergistic action of Terminaliachebula and starchy extract in enhancing carbonation, pore refinement, and microstructural densification.

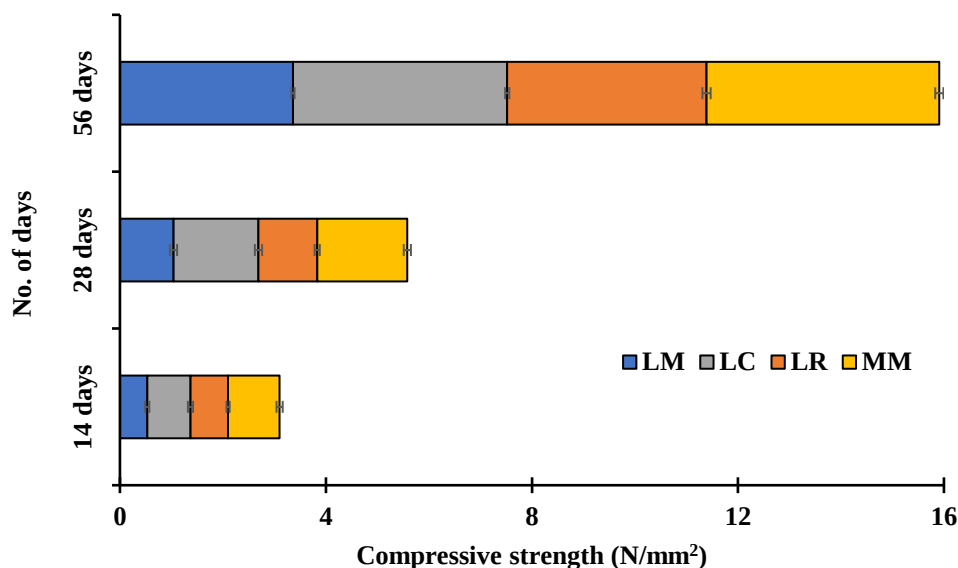


Figure 1. Compressive strength

Terminaliachebula contains tannins and polyphenolic compounds that interact with calcium ions (Ca^{2+}) released during lime hydration, forming calcium-organic complexes that act as nucleation sites for calcium carbonate precipitation and improve matrix cohesion [21], whereas starchy extract is rich in polysaccharides, which function as natural binders by improving adhesion, water retention, and moisture distribution within the mortar matrix [22]. The retained moisture facilitates the diffusion and dissolution of atmospheric CO_2 , thereby promoting prolonged carbonation and the continuous conversion of calcium hydroxide into calcium carbonate.

The enhanced strength development is supported by microstructural evidence. XRD analysis (Figure 2) revealed higher calcite peak intensities in the modified mortars, particularly in MM, indicating a greater degree of carbonation compared to the reference mortar. The appearance of a vaterite peak in the modified mortars further suggests that the organic additives influenced the carbonation pathway by promoting the initial formation of metastable calcium carbonate phases, which subsequently transformed into stable calcite. This phase transformation contributes to the

development of a denser and mechanically stronger matrix [23]. After 56 days of curing the quantitative analysis of mortar sample are illustrated in Table 4.

Table 4. Quantitative analysis of lime mortar samples

Phase	LM (%)	LC (%)	LR (%)	MM (%)
Calcite	42.6	55.4	59.8	68.7
Portlandite	37.8	24.3	20.5	11.8
Vaterite	0	4.63	2.41	5.11

The increased calcite content and reduced portlandite content observed in MM demonstrate that the combined incorporation of Terminaliachebula and starchy extract significantly enhanced the carbonation process. The polysaccharides present in the starchy extract improved moisture retention and facilitated CO_2 diffusion within the pore network, whereas the tannins and polyphenolic compounds in Terminaliachebula promoted calcium-ion complexation and provided nucleation sites for calcium carbonate precipitation. These synergistic effects accelerated the conversion of calcium hydroxide into calcium carbonate, leading to

greater carbon sequestration and microstructural densification. Furthermore, the presence of vaterite suggests that the organic additives altered the carbonation pathway by promoting the formation of metastable carbonate phases that subsequently transformed into stable calcite.

The SEM results section has been expanded to provide a more detailed interpretation of pore refinement and carbonate formation. The micrographs reveal that the reference mortar (LM) exhibits a relatively porous structure with visible portlandite crystals and limited carbonate precipitation. In contrast, the modified mortars (LC, LR, and MM) show increased formation of carbonate phases, reduced pore connectivity, and a denser matrix. Fine granular carbonate deposits and ACC-like phases were observed in LC and LR, while MM exhibited the highest degree of pore filling and calcite precipitation. These observations indicate enhanced carbonation and microstructural densification resulting from the incorporation of Terminaliachebula and starchy extract.

SEM observations (Figure 3) confirmed the presence of densely packed calcite crystals in the modified mortars. The precipitation of calcium carbonate within capillary pores and interparticle voids reduced pore connectivity and enhanced particle bonding. As carbonation progressed, the newly formed calcite crystals filled available pore spaces, resulting in a compact microstructure with improved load-transfer capacity. This pore-filling

effect reduced internal defects and increased the effective load-bearing area of the mortar. The formation of amorphous calcium carbonate (ACC) in the modified lime mortars is attributed to the interaction of calcium ions with organic compounds present in Terminaliachebula and starchy extract during carbonation. Polyphenols, tannins, and polysaccharides can stabilize transient ACC phases by inhibiting immediate crystal growth and delaying the transformation to crystalline calcite. SEM images revealed fine granular and crystalline carbonate deposits, characteristic of ACC-like phases [24].

Furthermore, TGA analysis demonstrated a higher carbonate content in the modified mortars, as evidenced by the greater mass loss associated with calcite decomposition. The increased carbonate content confirms enhanced CO₂ uptake and a higher degree of carbonation. Therefore, the combined effects of calcium-organic complex formation, enhanced carbonation, calcite precipitation, pore refinement, and microstructural densification are responsible for the significant improvement in compressive strength observed in MM. These findings demonstrate that the incorporation of starchy extract and Terminaliachebula not only improves carbon sequestration but also contributes to the development of a stronger and more durable lime mortar matrix [25].

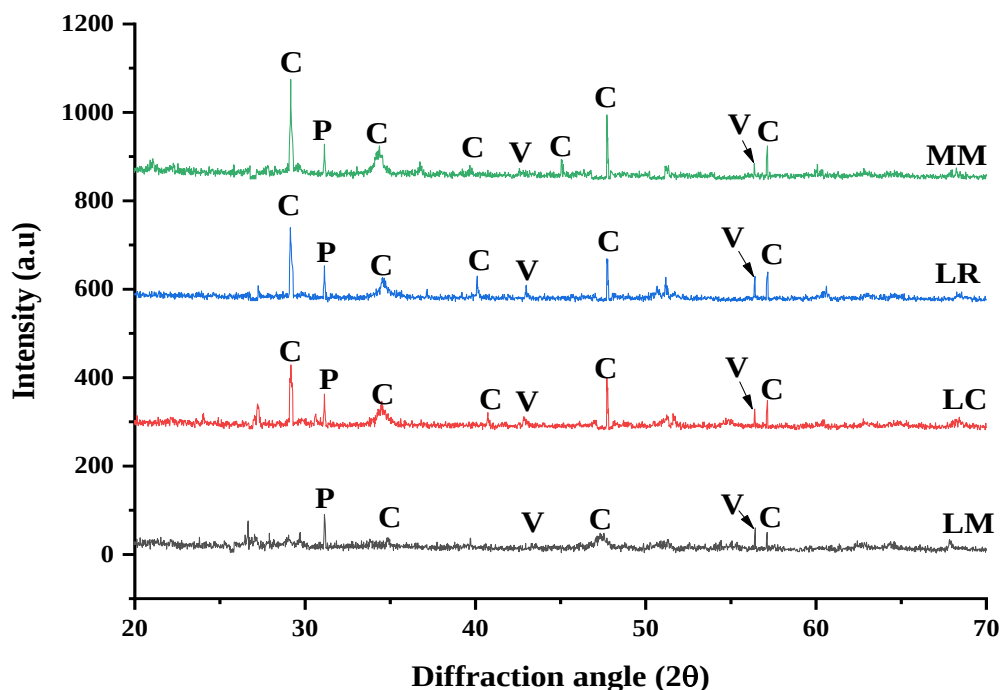


Figure 2. Mineralogical analysis through XRD

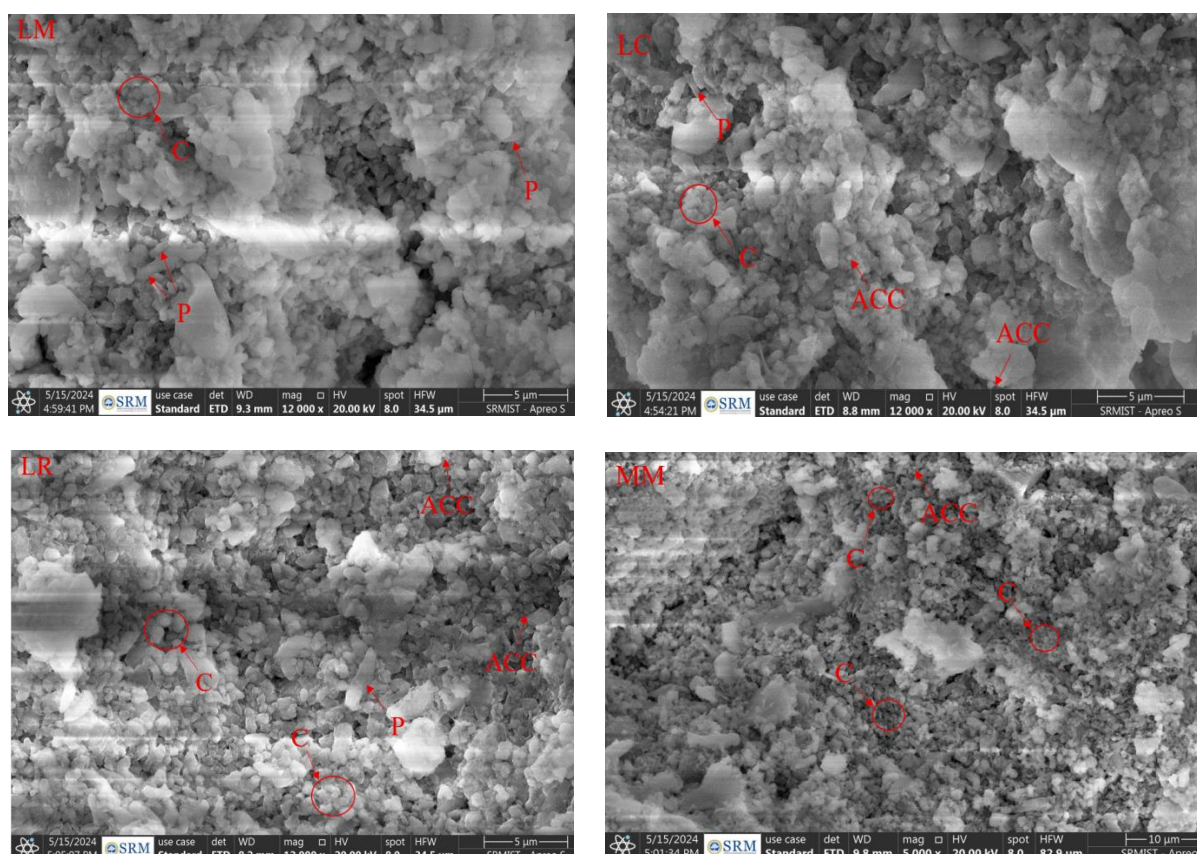


Figure 3. Morphological analysis through SEM

FTIR analysis (Figure 4) was carried out to identify the functional groups and carbonation products present in the reference and modified lime mortars. All samples exhibited characteristic absorption bands associated with calcium carbonate formation. The strong absorption band observed at $1400\text{--}1450\text{ cm}^{-1}$ corresponds to the ν_3 asymmetric stretching vibration of the carbonate ion (CO_3^{2-}), while the peaks near 875 cm^{-1} and $712\text{--}715\text{ cm}^{-1}$ are attributed to the ν_2 out-of-plane bending and ν_4 in-plane bending vibrations of calcite, respectively, confirming the formation of calcium carbonate during carbonation [26,27].

A broad band observed in the region of $3400\text{--}3600\text{ cm}^{-1}$ is assigned to O-H stretching vibrations associated with adsorbed water and residual calcium hydroxide [28]. The intensity of this band was relatively higher in the modified mortars, indicating improved moisture retention due to the presence of organic additives. Weak absorption bands around 2920 cm^{-1} correspond to aliphatic C-H stretching vibrations originating from the organic constituents of Terminalia chebula and starchy extract [29]. Furthermore, bands in the range of

$1600\text{--}1700\text{ cm}^{-1}$ are associated with C=O stretching vibrations of tannins, polyphenols, and polysaccharides present in the organic admixtures [30]. These functional groups can interact with calcium ions and influence nucleation and growth of calcium carbonate crystals.

Compared with LM, the modified mortars exhibited sharper and more intense carbonate bands, particularly in MM, indicating a higher degree of carbonation. The slight broadening of carbonate-related bands in LC suggests the influence of tannins and polyphenolic compounds on crystal growth and mineralization processes. In MM, the enhanced peak intensity and resolution indicate increased calcite formation and improved crystallinity. These observations are in good agreement with the XRD results, which showed higher calcite contents and lower residual portlandite contents in the modified mortars. Therefore, the FTIR analysis confirms that the incorporation of Terminalia chebula and starchy extract promoted carbonation, enhanced calcium carbonate formation, and contributed to microstructural refinement and carbon sequestration in the lime mortar system.

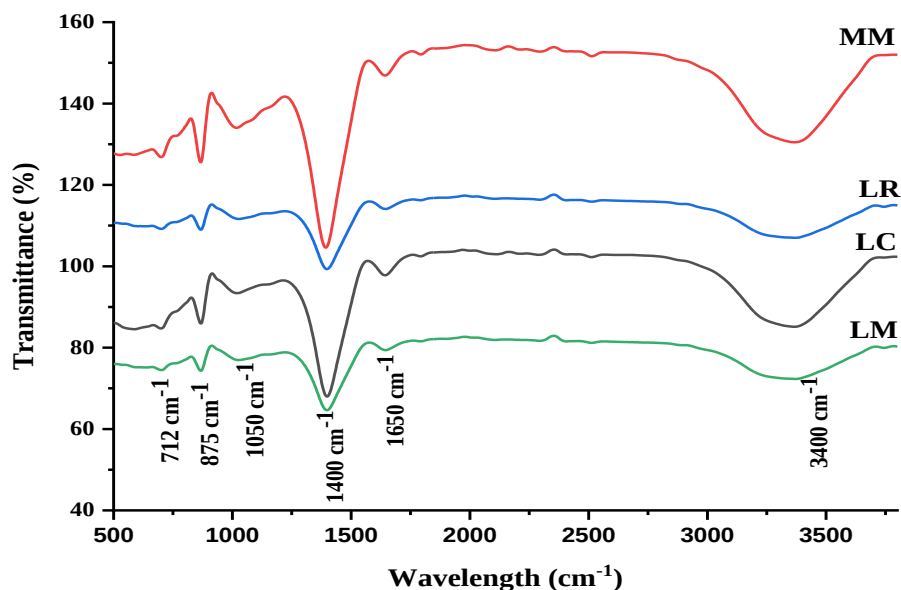


Figure 4. FT-IR analysis

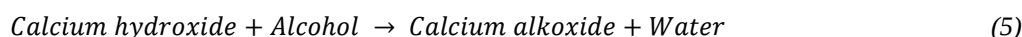
3.3. Carbonation rate

The early hardness and strength development of lime typically comprises of two phases - moisture evaporation and setting. In the first 3 to 7 days, excess water and moisture in the specimens evaporate, leading to weight reduction. Subsequently, carbonation occurs when the specimens absorb CO_2 from the atmosphere, resulting in an increase of weight and an enhancement of strength [31]. The change in weight profile serves as an indirect measure of the carbonation rate in the specimens, measured by monitoring the average percentage rise in weight of six specimens compared to the weights of those cured for three days, as shown in Figure 5 [32].

The reference sample (LM) exhibited a limited increase in weight of 1.47g within 56 days, resulting in carbonation rate of 26.25 mg/day. This indicates an overall weight increase of 15.02%, with a coefficient of determination of 0.954, confirming the strong correlation between the model and the dependent variable. The LR and LC modified mortar samples exhibited a slight reduction in the carbonation rate compared to the reference mortar. Specifically, LR and LC showed decreases of 10.17% and 3.6%, respectively. Despite the reduced carbonation rate, both samples demonstrated a noticeable increase in weight, with LR and LC recording weight gains of 13.32% and 14.08%, respectively. The coefficient of determination (R^2) values of 0.927 for LR and

0.959 for LC indicate a strong correlation between carbonation progression and weight gain. The formation of hydration products such as calcium silicates and aluminates inhibited the carbonation process compared to LR, as confirmed by the mineralogical evidence discussed in the XRD section[33].

The modified mortar (MM) shows an excellent carbonation of 42.5 mg/day, with the overall increase in weight of 18.93% and R^2 of 0.96. In lime mortar modified with chebula and starchy extract, the organic compounds released during soaking and fermentations such as alcohols, sugars, and polysaccharides enhance the carbonation process[34]. Alcohols formed in the mixture facilitate the transport of CO_2 into the inner pore structure of the mortar, improving carbonation throughout the specimen[35]. These alcohols can also react with calcium hydroxide to form reactive intermediates such as calcium alkoxides, which subsequently convert to calcium carbonate during carbonation as mentioned in equation 5&6[36]. Additionally, polysaccharides present in chebula and starchy extract retain moisture within the matrix, promoting the formation of hydration products while slightly delaying early carbonation. This controlled moisture retention improves internal curing and ultimately contributes to better microstructural development and durability of the lime mortar [37,38].



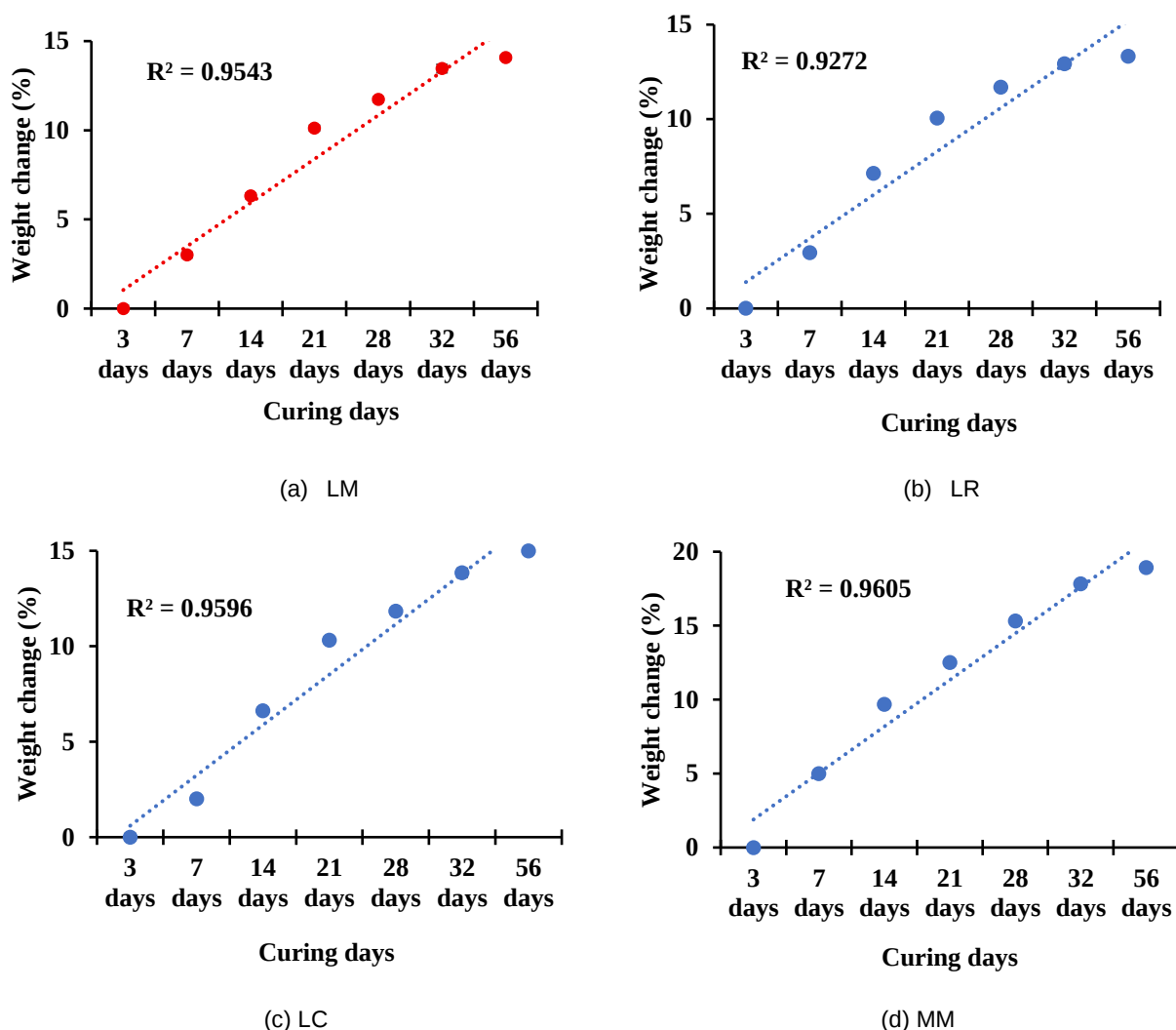


Figure 5. Weight change profile

3.4. Quantification of CO_2

The emission of carbon dioxide during the fermentation of *Terminalia chebula* extract, starchy extract, and their combination was monitored, and the results are presented in Figure 6 and 7. *Terminalia chebula* contains hydrolysable tannins, including chebulagic acid, chebulinic acid, and gallic acid, along with minor amounts of carbohydrates and proteins [39]. During fermentation, these tannins undergo hydrolysis to produce glucose-containing intermediates and phenolic compounds, which are subsequently metabolized by microorganisms to generate ethanol and CO_2 through the reactions represented in Equations (1) - (4). Starchy extract, in contrast, is rich in amylose and amylopectin, which are readily hydrolysed into fermentable sugars such as

glucose and maltose, thereby enhancing microbial activity and CO_2 production [40].

The fermentation of *Terminalia chebula* produced a maximum CO_2 concentration of approximately 10,000 ppm after 7 days, whereas starchy extract generated about 12,000 ppm after only 1 day due to its higher content of readily fermentable carbohydrates. The combined admixture exhibited substantially greater CO_2 evolution, reaching nearly 50,000 ppm. This significant increase indicates a synergistic interaction between the two organic additives. The starchy extract supplied abundant fermentable sugars that supported microbial metabolism, while *Terminalia chebula* provided bioactive compounds and micronutrients that enhanced microbial growth and fermentation efficiency [41].

The increased CO_2 evolution is beneficial for lime mortar carbonation because the released CO_2 reacts with calcium hydroxide to form calcium carbonate, the principal binding phase responsible for strength development and carbon sequestration. Enhanced carbonation in the modified mortars was confirmed by XRD analysis, which showed increased calcite peak intensities compared to the reference mortar. The presence of vaterite peaks in the modified samples further indicates that the organic additives influenced the carbonation pathway by promoting the formation of intermediate calcium carbonate phases that subsequently transformed into stable calcite.

The microstructural evidence obtained from SEM analysis revealed densely packed calcium carbonate crystals filling capillary pores and interparticle voids in the modified mortars. This pore-filling effect contributed to microstructural densification and improved particle bonding. Furthermore, TGA results demonstrated a higher carbonate content in the modified mortars, confirming increased CO_2 uptake and a greater degree of carbonation. Consequently, the combined use of Terminaliachebula and starchy extract enhanced carbon sequestration capacity, and microstructural development, resulting in improved mechanical performance and durability of the lime mortar system.

Although the study evaluated carbonation behavior, carbon sequestration, and microstructural development up to 56 days, lime-based materials continue to undergo carbonation over extended periods under natural exposure conditions. The increased calcite formation observed through XRD, higher carbonate content identified by TGA, and dense microstructure revealed by SEM suggest that the modified mortars possess the potential for continued carbonation and strength development beyond the testing period. As carbonation progresses, additional calcium carbonate may precipitate within the pore network, further refining the microstructure and enhancing durability.



Figure 6. Experimental setup for CO_2 quantification

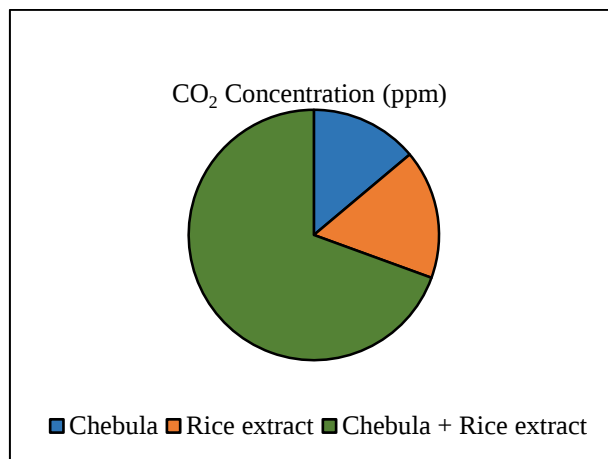


Figure 7. Quantification of CO_2 liberation

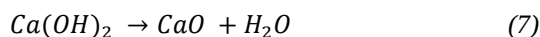
3.5. TGA

TGA-DTA analysis (Figure 8) was performed to evaluate the thermal decomposition behavior of the hydration and carbonation products present in the lime mortars. Four distinct weight-loss regions were identified based on the decomposition temperatures of the different phases.

The first weight-loss region ($<200^\circ\text{C}$) corresponds to the evaporation of free water, physically adsorbed moisture, and weakly bound water present within the pore structure. The weight loss in this region increased from 2.24% in LM to 5.11% in MM, indicating enhanced moisture retention in the modified mortars. This behavior is attributed to the hygroscopic nature of the polysaccharides present in starchy extract and the tannins contained in Terminaliachebula, which improve internal moisture storage and support continued carbonation reactions.

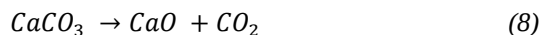
The second region ($200\text{--}400^\circ\text{C}$) is associated with the decomposition of calcium–organic complexes and the release of chemically bound water from hydration products. The weight loss values ranged from 4.47% in LM to 4.78% in MM, suggesting increased interaction between calcium ions and the organic constituents in the modified mortars.

The third region ($400\text{--}600^\circ\text{C}$) corresponds primarily to the dehydroxylation of residual calcium hydroxide ($\text{Ca}(\text{OH})_2$), according to equation (7):



The higher weight losses observed in LR (3.29%) and MM (3.43%) indicate the presence of hydrated phases undergoing transformation during curing.

The fourth region ($600\text{--}800^\circ\text{C}$) represents the decomposition of calcium carbonate (CaCO_3), which is the principal carbonation product (Equation 8).



This region is directly related to the degree of carbonation and CO₂ sequestration. The modified mortars exhibited higher carbonate-related weight losses, with MM showing the highest value of 6.22%, followed by LR (6.16%), LC (5.78%), and LM (6.01%) as illustrated in Table 5. The greater weight loss in this temperature range indicates increased calcite formation and enhanced carbonation in the modified mortars.

The TGA results demonstrate that the incorporation of starchy extract and Terminaliachebula enhanced moisture retention, promoted carbonation, and increased carbonate formation. These findings are consistent with the XRD results showing higher calcite contents and lower residual portlandite contents in the modified mortars. The enhanced carbonate formation contributed to improved carbon sequestration, microstructural densification, and strength development. Based on the TG-derived carbonate content and the XRF-determined CaO content (66.8%), the modified mortar MM exhibited the highest CO₂ capture efficiency, confirming the beneficial role of the organic additives in improving carbon capture and utilization (CCU) performance.

Table 5. TG analysis of mortar samples

Sample code	Weight loss (%)			
	<200 °C	200-400 °C	400-600 °C	600-800 °C
LM	2.24	4.47	3.04	6.01
LC	3.91	4.53	3.16	5.78
LR	3.41	4.65	3.29	6.16
MM	5.11	4.78	3.43	6.22

The amount of CO₂ captured from the atmosphere through carbonate formation in hardened lime mortar was quantified using the weight loss percentage obtained from TG analysis. The sample, MM showed the highest CO₂ capture of 3.46% followed by LR of 2.42% increase compared to the reference mortar. The improved carbonation, together with a well-developed microstructure and proper crystallization in the organically modified samples, contributed to enhanced strength and overall performance. In contrast, sample LC showed a 3.8% lower carbon capture capacity compared to the reference. However, the formation of hydration products, ACC in LC was greater in comparison to reference mortar which helps to contribute the improved early strength and better performance characteristics.

The theoretical CO₂ released during lime production was estimated to be approximately 619 g per kilogram of lime, calculated using the stoichiometric relationship between CaO and CO₂. The mortar sample reabsorbed 296.8 g of CO₂ per kg of lime, corresponding to a carbon sequestration of about 47.9 %. This partial CO₂ uptake highlights the environmental benefit of lime-based materials, particularly when modified with traditional organic additives, which enhance carbonation and improve CCU potential. Compared to Portland cement systems, which exhibit negligible direct CO₂ reabsorption, lime mortars demonstrate a notable capacity for atmospheric carbon sequestration during the carbonation process.

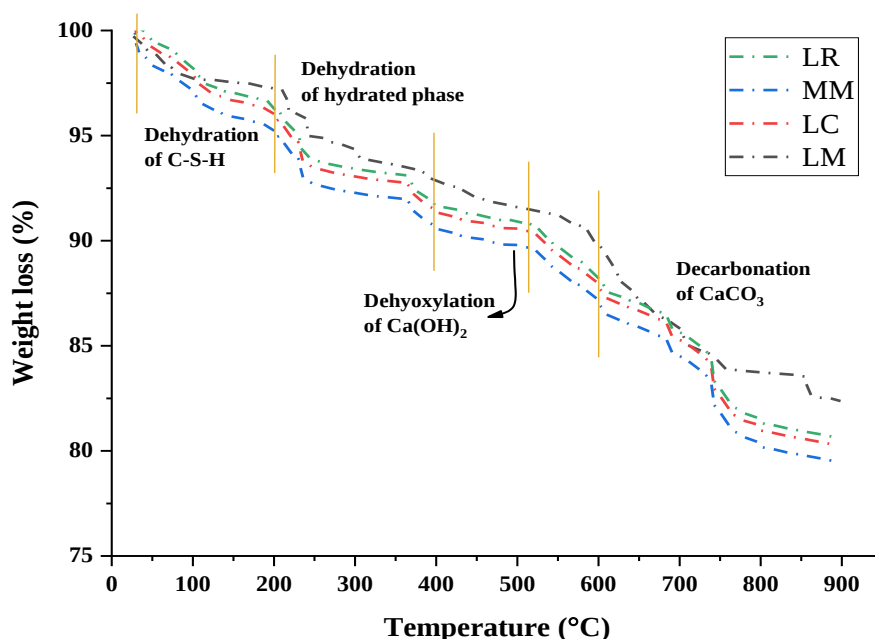


Figure 8. Weight loss in TG analysis

3.6. Porosity

The pore size distribution obtained from BET analysis exhibited multimodal characteristics with distinct peaks across all samples, indicating the presence of both microand mesoporous structures within the lime mortars. Major pore size ranges were observed between 3-10 nm and 20-30 nm. The presence of fine pores within the 3-10 nm range contributes to matrix densification and improved mechanical performance, whereas mesopores ranging from 20--100 nm facilitates moisture transport, CO₂ diffusion, and carbonation reactions [42,43]. Such pore structures are particularly important in lime-based systems

because carbonation depends on the simultaneous availability of atmospheric CO₂, moisture, and reactive calcium hydroxide [44].

The reference mortar (LM) exhibited a total pore volume of 0.075 cc/g (Figure 9), indicating an open and interconnected pore structure. This pore network provides adequate pathways for moisture movement and CO₂ transport, enabling moderate carbonation and strength development. However, the relatively higher proportion of coarse capillary pores may reduce matrix compactness and increase susceptibility to localized stress concentrations.

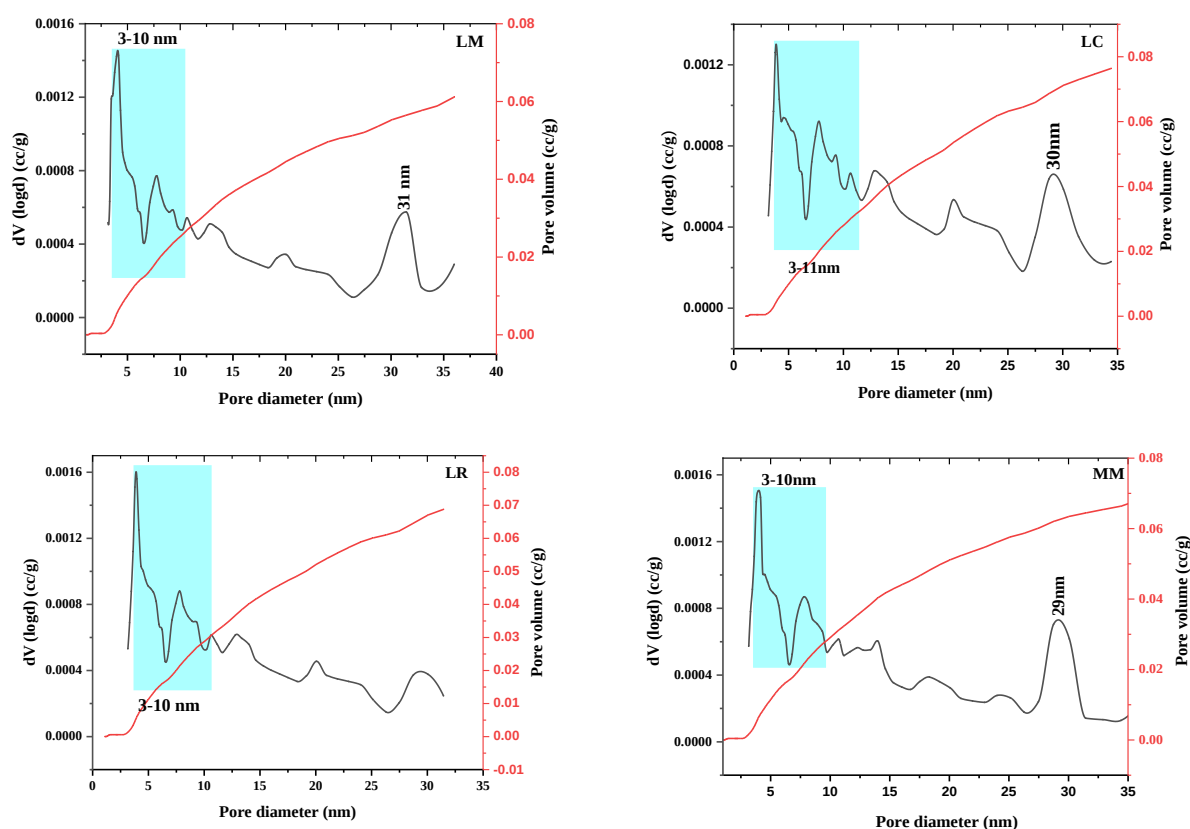


Figure 9. Pore volume and pore size distribution vs pore diameter from BET analysis

The starchy extract-modified mortar (LR) exhibited a reduced pore volume of 0.068 cc/g, indicating partial refinement of the pore structure. The polysaccharides present in the starchy extract likely promoted improved particle packing and the formation of an organic-inorganic network, reducing excessive capillary porosity while preserving sufficient mesoporosity for CO₂ ingress. This refined pore structure facilitates moisture retention and supports continuous carbonation, thereby promoting calcite formation and strength development.

The Terminaliachebula-modified mortar (LC) showed the lowest pore volume of 0.061 cc/g, indicating the densest microstructure among all mixes. The tannins, polyphenols, and organic acids present in Terminaliachebula can interact with Ca²⁺ ions released during lime hydration, forming calcium-organic complexes that reduce pore connectivity and limit the development of larger capillary pores. While this compact microstructure improves cohesion, durability, and crack resistance, the reduced pore connectivity may slightly restrict CO₂ penetration into the mortar

interior, resulting in a more gradual carbonation process.

In contrast, the combined mortar (MM) exhibited the highest pore volume of 0.080 cc/g. This result suggests that the combined action of starchy polysaccharides and tannin-rich compounds generated a balanced and well-connected mesoporous network rather than excessive porosity. Carbonation in lime mortars is strongly governed by pore size distribution because CO₂ transport occurs primarily through interconnected pores. Pores smaller than approximately 10 nm provide high moisture retention but may restrict gas diffusion, whereas mesopores between 20 and 100 nm facilitate efficient CO₂ transport and provide sufficient space for calcium carbonate precipitation [45]. The pore structure of MM therefore appears particularly favourable for carbonation, as the interconnected mesopores allow deeper CO₂ penetration while the retained moisture promotes CO₂ dissolution and subsequent reaction with calcium hydroxide.

The enhanced CO₂ diffusion characteristics of MM are consistent with the microstructural and mineralogical results obtained from XRD and TGA analyses. The higher calcite content determined by Rietveld refinement, together with the increased carbonate decomposition observed in TGA, confirms a greater degree of carbonation in MM. These findings demonstrate that not only total pore volume but also pore connectivity and mesopore distribution play a critical role in controlling CO₂ diffusion, carbonation kinetics, and long-term carbon sequestration in lime mortars. The synergistic interaction between starchy extract and Terminaliachebula produced an optimized pore structure that facilitated CO₂ diffusion and calcium carbonate formation while maintaining mechanical stability.

3.7. Practical Implications for Construction Applications

The practical implementation of starchy extract and Terminaliachebula in lime mortars is highly feasible owing to their natural abundance, low cost, and long history of use in traditional construction systems. Starchy extract is derived from rice, a readily available agricultural product, while Terminaliachebula is widely distributed across Indias, making both additives easily accessible for construction applications.

The preparation process involves simple boiling and fermentation procedures that require minimal energy input and no specialized equipment, thereby facilitating field-scale production. Furthermore, the relatively low dosage levels adopted in this study (2% starchy extract and 1% Terminaliachebula by binder weight) limit the

additional material cost and enable easy incorporation into conventional lime mortar mixing practices.

From a sustainability perspective, these bio-based additives offer an environmentally friendly alternative to synthetic admixtures while enhancing carbonation, CO₂ sequestration, microstructural development, and mechanical performance. Therefore, the developed mortar system shows strong potential for heritage conservation, restoration projects, and sustainable non-load-bearing masonry applications. Nevertheless, further field-scale investigations and long-term durability studies under diverse environmental conditions are necessary to establish its large-scale practical applicability.

3.8. Limitations and future scope of the study

The present study evaluated carbonation, carbon capture, and mechanical performance up to 56 days under controlled laboratory conditions. Future studies should investigate the long-term carbonation kinetics, CO₂ sequestration capacity, and durability behavior of the modified mortars under natural exposure conditions.

- The influence of environmental factors such as temperature, relative humidity, rainfall, and atmospheric CO₂ concentration was not considered. Further research should assess the performance of the modified mortars under diverse climatic conditions, including tropical, coastal, arid, and humid environments.
- Only a one dosage combination of starchy extract and Terminaliachebula was investigated. Optimization studies involving different additive proportions are required to determine the most effective composition for maximizing carbonation efficiency, carbon sequestration, and mechanical performance.
- The study focused primarily on laboratory-scale evaluation of non-load-bearing lime mortars. Large-scale field validation is necessary to assess the practical feasibility, performance consistency, and long-term behavior of the developed mortars in real construction applications.
- Although the modified mortars demonstrated enhanced carbonation and strength development, their applicability in heritage conservation remains to be fully validated. Future investigations should examine compatibility, durability, and long-term effectiveness in historic masonry and restoration projects.

4. CONCLUSION

The present study demonstrated that organic-modified lime mortars incorporating starchy extract

and Terminaliachebula significantly enhanced carbon capture and utilization (CCU) capacity while simultaneously improving key performance parameters, including workability, mechanical strength, carbonation behavior, and microstructural development. The findings confirm the effectiveness of traditional organic additives in promoting carbon sequestration and improving the suitability of lime mortars for sustainable non-load-bearing construction applications. The major conclusions from this study are as follows:

- The incorporation of natural organic additives significantly enhanced the carbonation behavior, carbon sequestration potential, and mechanical performance of lime mortars. Among all mixtures, the combined admixture containing 1% Terminaliachebula and 2% starchy extract (MM) exhibited the optimum overall performance.
- The starchy extract-modified mortar (LR) improved carbonation by 39.7% and compressive strength by 15.2% compared with the control mortar, while reducing pore volume from 0.075 cc/g to 0.068 cc/g through improved moisture retention and pore refinement.
- The Terminaliachebula-modified mortar (LC) developed the densest microstructure with the lowest pore volume (0.061 cc/g), owing to tannin-Ca²⁺ interactions that promoted matrix cohesion, water retention, and the formation of stable carbonate phases.
- The combined mortar (MM) exhibited an optimized pore network with a pore volume of 0.080 cc/g, facilitating effective CO₂ diffusion and carbonation while maintaining structural stability. This synergistic pore structure promoted enhanced calcite formation and microstructural densification.
- Compared with the reference mortar, the optimum mixture (MM) achieved 35% higher compressive strength and 3.46% greater CO₂ sequestration capacity, demonstrating that the combined use of Terminaliachebula and starchy extract is an effective strategy for developing sustainable lime mortars with improved carbon capture and utilization potential for heritage conservation and non-load-bearing construction applications.

Credit authorship contribution statement.

NishaSankar: Conceptualization, Formal analysis, Curation, Methodology, Data visualization, Investigation, Original draft-writing, **Ravi Ramadoss:** Conceptualization, Formal analysis, Data visualization, Data Curation, Writing-review & editing, all authors have read and agreed to the submitted version of the manuscript.

Declaration of competing interests

The authors declare that there are no potential competing interests.

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IZVOD

SEKVESTRACIJA UGLJENIKA I POBOLJŠANJE PERFORMANSI KREČNOG MALTERA MODIFIKOVANOG PRIRODNIM ADITIVIMA ZA ODRŽIVU GRADNJU

Ova studija je istraživala potencijal krečnog maltera modifikovanog ekstraktom skroba i terminalijahebulom za poboljšanje hvatanja i korišćenja ugljenika (CCU) za održive građevinske primene. Modifikovani malteri su procenjeni korišćenjem ispitivanja čvrstoće na pritisak, rendgenske difrakcije (XRD), infracrvene spektroskopije sa Furijeovom transformacijom (FTIR), termogravimetrijske analize (TGA) i BET analize strukture pora kako bi se procenili mehanički performanse, ponašanje karbonizacije i mikrostrukturna evolucija. Rezultati su pokazali da je malter modifikovan ekstraktom skroba poboljšao karbonizaciju za 39,7% i mehaničke performanse za 15,2% zbog poboljšanog rafiniranja pora i zadržavanja vlage, smanjujući zapreminu pora na 0,068 cc/g u poređenju sa referentnim malterom (0,075 cc/g). Malter modifikovan čebulom pokazao je najmanju zapreminu pora (0,061 cc/g), što ukazuje na gušću mikrostrukturu formiranu interakcijama tanina i Ca^{2+} , promovirajući koheziju, zadržavanje vode i formiranje amorfnog kalcijum karbonata (ACC). Kombinovani aditivni malter pokazao je najveću zapreminu pora (0,080 cc/g), stvarajući optimizovanu mrežu pora koja je poboljšala difuziju CO_2 i održala stabilnost matrice, što je rezultiralo 35% većom čvrstoćom na pritisak i 3,46% većom efikasnošću CCU u odnosu na referentni malter. Mikrostrukturna analiza potvrdila je poboljšano formiranje kalcita u malteru sa ekstraktom skroba, matrice bogate ACC-om u malteru Chebula i uravnotežene kristalno-amorfne karbonatne faze u kombinovanom sistemu. Rezultati pokazuju da prirodni organski aditivi značajno poboljšavaju karbonizaciju, sekvestraciju ugljenika i mehaničke performanse krečnog maltera za održive primene u nenosećim konstrukcijama.

Ključne reči: Krečni malter, sekvestracija ugljenika, hvatanje i korišćenje ugljenika (CCU), održivi materijal

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