Does lack of CO_2 control in environmental chambers influence the strength and stiffness of hydraulic lime mortars?

Miles Judd[1*], *Dylan* Squires[1], *Kutsi* Akcicek[1], *Marialuigia* Sangirardi[1], and *Sinan* Acikgoz[1]

[1]University of Oxford, Department of Engineering Science, OX1 3PJ Oxford, United Kingdom

Abstract. This study investigates the influence of carbon dioxide (CO2) control in environmental chambers, where hydraulic lime mortar specimens are typically cured before strength testing according to EN1015-11:2019. To do this, three sets of identical mortar samples were cast. Two sets were cured in an environmental chamber that offered the same temperature and relative humidity but different CO2 conditions. In the first chamber, carbonation reactions used the ambient CO2 trapped in the hermetically sealed chamber air. The second chamber was connected to an external carbon dioxide supply, which maintained a constant CO2 concentration, above ambient laboratory levels, during curing. A third set of samples was cured under laboratory conditions to serve as a reference. Following the 35-day curing period, the dynamic elastic modulus and flexural and compressive strengths of sample sets were obtained. Subsequent X-ray diffraction analysis gives additional insights into the material composition for each curing regime. Results indicate that controlling carbon dioxide to above ambient levels leads to noticeably higher stiffness and strength in samples. This suggests that implementing CO2 control as a part of standard curing conditions may enable more reliable estimates of strength and stiffness development in lime mortars, with respect to laboratory conditions.

1 Introduction

Lime mortar is a fundamental building material in many historic constructions. Curing, the process of maintaining an adequate environment (humidity, temperature, etc.) to facilitate the carbonation reactions (the transformation of slaked lime ($Ca(OH)_2$) to calcite ($CaCO_3$) in the presence of CO_2), is a critical determinant of the lime mortar's final mechanical characteristics. Understanding the effects of different curing environments on mortar properties is needed for optimising construction practices and ensuring long-term structural integrity. In addition, the possibility to incorporate elevated carbon dioxide (CO_2) levels during curing enables faster strength development in the laboratory [1] and may facilitate the development of sustainable carbon-sequestering mortars [2].

* Corresponding author: miles.judd@eng.ox.ac.uk

These proceedings are published with the support of EuLA.

Multiple studies have been conducted to investigate the factors affecting the curing process of mortars and their resulting properties [3], [4], [5]. In 1956, Zalmanoff [6] related higher CO_2 levels during the curing of lime putties to an increased carbonation rate and mechanical strength. Additionally, he reported a decrease in mechanical strength for dry environments [6] and temperatures above 30℃ [7]. Although several authors investigated the effect of curing lime mortar under elevated CO_2 levels on the compressive strength of the material [5], [6], [7], [8], [9], there is currently no evaluation of the effect of curing on mortar elasticity. While [10] related the advanced carbonation of lime mortars to increased strength and elasticity, the authors did not consider elevated CO_2 levels. In addition to improved mechanical properties, Moro *et al*. [2] investigated the economic and environmental benefits of curing by carbon capture. They indicated that CO_2-curing over the lifetime of the materials leads to improvements in the environmental and economic performance of mortar.

In this study, we investigate the impact of two distinct curing conditions on the mechanical properties of hydraulic lime mortar: curing in a hermetically sealed chamber with and without elevated levels of CO_2. Both chambers maintain the temperature and relative humidity recommended by the EN1015-11:2019 guidelines, in advance of mechanical testing. Although the standard does not specify whether the chamber should be hermetically sealed, many commercial chambers adopt this approach to ensure constant climate conditions. The curing environment is expected to influence the carbonation kinetics and microstructural development, which may have an impact on the mechanical performance of the mortar specimens. The results are further compared to samples cured under laboratory conditions, without active control of humidity, temperature, and CO_2. The strength properties under investigation are tensile and compressive strengths, assessed through flexural and compression testing, respectively. Additionally, we explore the dynamic elastic modulus of the mortar samples and the chemical composition of the materials.

2 Experimental study

Flexural mortar samples were produced using natural hydraulic lime with a nominal strength of 3.5MPa (NHL3.5) and cured under varying levels of CO_2 and at laboratory conditions. After 35 days of curing, the samples were subjected to mechanical tests to determine their dynamic elastic modulus, flexural strength, and compressive strength. Mechanical tests were complemented with X-ray diffraction (XRD) analysis to understand the chemical composition of the specimens investigated.

2.1 Specimen preparation

In accordance with the manufacturer's guidelines, a mixture comprising 1 kg of water, 1.285 kg of NHL 3.5 lime, and 3.21 kg of sand was prepared. Subsequently, a flow test conducted as per EN 459 [11], yielded an overall flow value of 180.5mm. The resultant mixture was then cast into steel moulds, creating nine specimens of consistent dimensions measuring 160mm x 40mm x 40mm. After a period of three days, the specimens were extracted from the moulds. In accordance with EN 1015-11 guidelines [12], mortar samples were stored within airtight plastic bags together with wet cloths for the first seven days to uphold a minimum relative humidity (RH) of 95%.

After seven days, three sets of three specimens each were subjected to curing under the following conditions: 1) in a hermetically sealed chamber without external CO_2 supply (low-CO_2 chamber), 2) in a hermetically sealed chamber with elevated levels (nominally 2000 ppm) of CO_2 (high-CO_2 chamber) and 3) at laboratory conditions (lab-cured). Both environmental chambers (Memmert ICH110L and ICH110C respectively) were maintained

at a constant temperature of 20℃ and a RH of 65%. The air inside the chambers was circulated using built-in fans. Although slight fluctuations in temperature and humidity can occur in the laboratory, observations indicated a reasonably stable environment around 18℃ and 50% RH. Given the specimens were placed in a plastic box, however, it is likely they experienced humidity and CO_2 values varying from laboratory conditions. A comparable batch of mortar samples, cured inside a loosely closed plastic box, demonstrated considerably higher humidity (90% RH) and reduced CO_2 levels (500 ppm) in contrast to the ambient conditions of the laboratory (50% RH, 700 ppm CO_2) in which the box was placed.

The curing conditions are outlined in Table 1. Specimens remained in their curing environment and were tested after a total of 35 days of curing.

Table 1. Different curing environments. Results are presented in the format Mean±Standard deviation.

	Low-CO_2 chamber	**High-CO_2 chamber**	**Laboratory***
Relative Humidity [%]	65.6±3.8	65.7±3.3	~50
Temperature [℃]	20.0±0.2	20.0±0.4	~19
CO_2 [ppm]	394.4±41.5	2273.1±511.2	400–900

* Conditions describe general laboratory conditions as opposed to the local curing conditions in the box

2.2 Mechanical testing

First, the dynamic elastic modulus was estimated using the non-destructive Impulse Excitation of Vibration (IEV) technique. Second, a three-point bending test was conducted to assess the tensile strength of the cured materials. Finally, a compressive test was used to compare the compressive strength of the mortar samples.

2.2.1 Dynamic modulus of elasticity

IEV was performed per ASTM E 1876-01:2002 [13]. By exciting the flexural and torsional modes of the flexural specimen and measuring the dynamic response using the Polytec PDV-100 laser vibrometer, the resonant frequencies of the samples were calculated. Factoring in the mass, density, and geometry of the specimen, these natural frequencies were then used to determine the dynamic modulus of elasticity of the material. Average natural frequencies were estimated for both flexural and torsional modes across five repeat tests for each specimen.

Fig. 1 demonstrates that the high-CO_2 chamber shows a systematically higher dynamic elasticity compared to its low-CO_2 counterpart. The increase of ambient CO_2 results in the dynamic elasticity to more than double. At the same time, the lab-cured samples indicate the highest dynamic elasticity, slightly exceeding the samples in the high-CO_2 chamber.

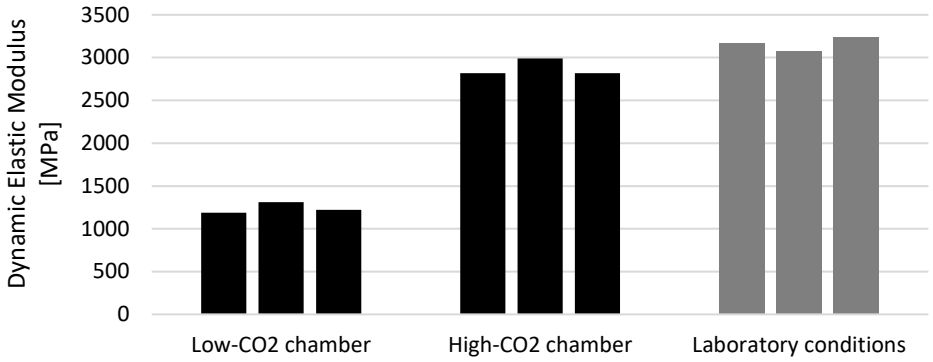

Fig. **1.** Dynamic elasticity modulus [MPa] for each curing regime estimated through IEV 35 days after casting.

2.2.2 Three-point bending test

Three-point bending tests were conducted according to EN 1015-11:2019 [12]. A Controls flexural testing device was used to apply loads uniformly and accommodate potential geometric imperfections of the specimen across contact areas. A constant displacement rate of 2 μm/s was employed on an INSTRON Universal testing machine, resulting in failure within a 60–90s timeframe.

The load-crosshead displacement curves derived from flexural testing are depicted in Fig. 2. Curing in the low-CO_2 chamber yielded the smallest average flexural strength (0.17 ± 0.02 MPa). In contrast, specimens subjected to high-CO_2 environments consistently demonstrated significantly greater average flexural strength (0.49 ± 0.04 MPa), nearly threefold higher than those from low-CO_2 conditions. Additionally, specimens cured in the laboratory exhibited the highest flexural strength (0.55 ± 0.09 MPa) relative to those cured in either chamber setting.

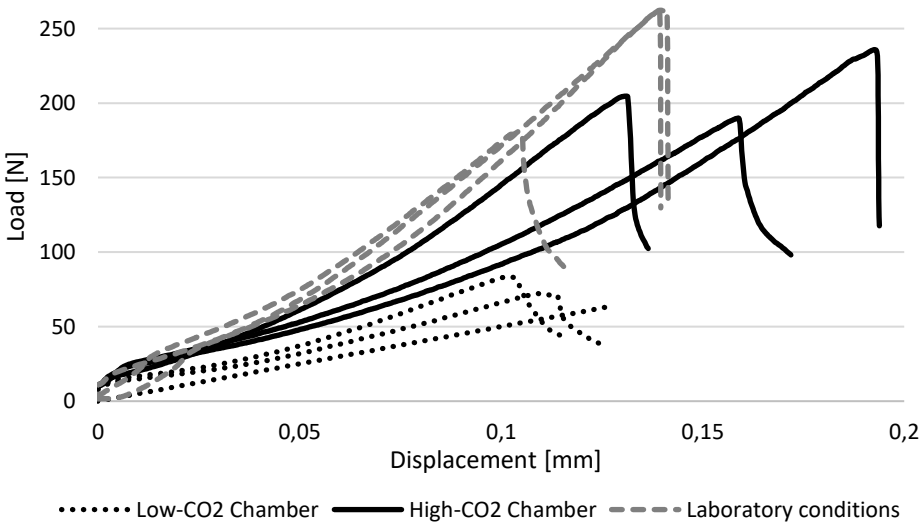

Fig. **2.** Load-displacement curves from flexural tests obtained 35 days after casting.

2.2.3 Compressive test on flexure halves

The compressive strength of the specimens was evaluated using the remaining flexural halves resulting from the three-point bending tests. A testing apparatus, compliant with EN 196 [14] was used to conduct the compression tests, to ensure as uniform loads as possible. A cyclic regime, involving three loading and unloading cycles from 0.1 MPa to 0.5 MPa, was adopted at the beginning of the test. This was used to estimate the static elastic modulus of the samples inspired by the BS EN 14580-2005 guidelines, but the results are not presented here. Following the completion of the final cycle, the specimen underwent loading until failure. A loading rate of 0.025 MPa/s was applied. Fig. 3 displays the compressive strength of all flexural halves grouped according to their respective curing environment. The average compressive strength values recorded for the three curing groups are 0.81 ± 0.07 MPa (low-CO_2 chamber), 1.98 ± 0. 14 MPa (high-CO_2 chamber) and 2.62 ± 0.11 MPa (lab-cured). Consistently, the specimens from the low-CO_2 chamber exhibit the lowest compressive strength, failing at approximately 40% of the average compressive strength observed in the high-CO_2 chamber samples. Lastly, specimens curing in the laboratory once again exhibit the highest compressive strength, failing at around 2.6 MPa.

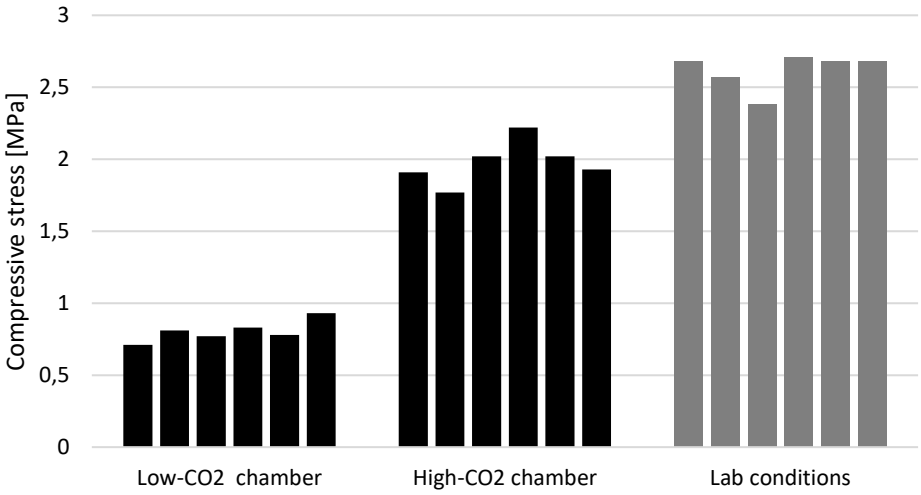

Fig. 3. Maximum compressive stress [MPa] of the broken flexural halves 35 days after casting.

2.3 X-ray diffraction analysis

XRD analysis was conducted to get insights into the chemical composition of the specimens investigated. A Rigaku Miniflex 600 was used with Cu-Kα radiation and a wavelength α_1 of 1.5406Å. The step size was set to $2\theta = 0.05^{\circ}$ with a scan step time of 0.1545 seconds. Measurements were performed approximately three months after casting the mortar prisms. One sample per curing regime was collected and analysed.

The normalised intensity signals from the XRD analysis are presented in Fig. 4. The peaks in the diffraction patterns indicate the presence of calcite (C), portlandite (P), and quartz (Q). Notably, portlandite is predominantly detected in the sample cured in the low-CO_2 chamber, suggesting that the carbonation of portlandite to calcite was less effective in this environment compared to the high-CO_2 chamber and lab-cured samples. Quartz peaks are observed across all curing regimes, corresponding to the consistent sand content, as all specimens were sourced from the same batch. Interestingly, the clearest calcite peaks are observed in the lab-

cured and low-CO_2 chamber samples. In contrast, the high-CO_2 chamber samples exhibit a generally low signal over a wide range of two theta positions, with a very prominent quartz peak at approximately $2\theta \approx 27°$.

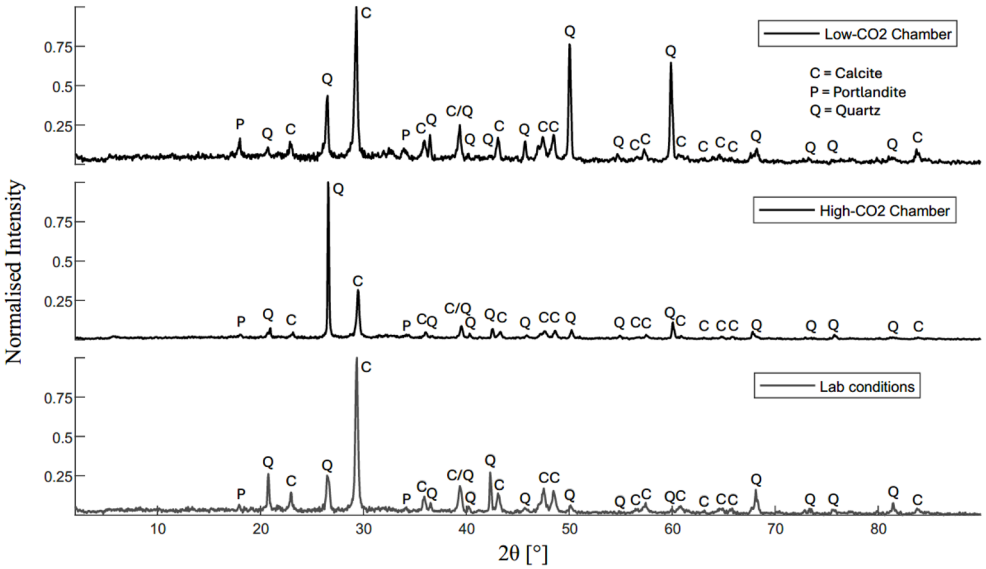

Fig. 4. Normalised intensity signal from XRD analysis for various angles of diffraction at their respective two theta positions, measured three months after casting.

3 Discussion

Results show a systematic difference in compressive strength, flexural strength, and the dynamic elasticity modulus when comparing the low- vs high-CO_2 environment. The low-CO_2 chamber shows the lowest values for all properties investigated. These findings agree with previous studies that related curing in a CO_2-rich atmosphere to increased mechanical strength [6]. Increasing the CO_2 concentration from atmospheric levels of around 400 ppm up to around 2200 ppm resulted in the dynamic elastic modulus more than doubling. Given the same humidity and temperature levels were used for both specimens, authors relate these findings to the accelerated carbonation process of natural hydraulic lime mortar in CO_2-rich environments [8].

Despite ambient CO_2 levels falling between those of the low- and high-CO_2 chambers, the specimens cured in the laboratory exhibited the highest dynamic elastic modulus, flexural strength, and compressive strength. Given the multitude of factors influencing curing, including temperature, humidity, CO_2 concentration, and their intricate interplay, explaining the mechanism behind the enhanced mechanical strength is challenging. Given the relatively minor temperature differences between the laboratory and the environmental chambers, the authors believe that variations in humidity, CO_2 levels, and air circulation may account for the observed differences. However, the scientific literature reports contradictory findings. Moorehead [5] identified 60% RH as favourable, with both excessively high (>75%) and low (<45%) RH levels linked to decreased carbonation. Additionally, Ergenç and Fort [8] confirmed decreased carbonation in a laboratory environment with low humidity (34%). However, Fusade and Viles [15] reported increased carbonation depth and compressive strength of NHL2 mortar after 28 days of curing at 85% RH and 15°C compared to standard conditions of 65% RH and 20°C. Similarly, Arizzi et al. [16] observed an increase in

compressive strength of NHL3.5 mortar from 60% to 90% RH. Notably, the authors stated that while carbonation is accelerated at 60% RH, the hydration reaction proceeds more slowly than at 90% RH.

Samples in the high-CO_2 chamber further experienced higher gas velocities due to the active fan, whereas the lab-cured specimens were placed in an unsealed box. Van Balen and Van Gemert [17] suggested that strong wind velocity promotes the drying process of fresh mortar, facilitating the initiation of carbonation. However, Moorehead [4] reported a minimal impact of forced air circulation on the rate of carbonation.

In a similar set of experiments, the authors recorded substantially higher RH, slightly lower CO_2-concentration and similar temperatures in the curing box compared to the laboratory conditions it was stored. It is assumed that the lower RH in the climate chambers in combination with the use of fans to circulate the air lead to the specimens drying out and the carbonation reaction ending prematurely. Despite lower CO_2-levels in the laboratory, the higher RH (exceeding EN1015-11:2019 recommendations) in the plastic box may have allowed for quicker hydration reactions, resulting in a higher strength at the end of the 35 day period.

These diverse findings underline the difficulty in explaining the causes of variations in mechanical properties across different curing conditions and highlight the need for further investigation in this area. Regardless, it is important to note that the results clearly demonstrate that lack of CO_2 control in the curing conditions specified by EN1015-11:2019 may be expected to lead to a notable underestimation of material stiffness and strength, in comparison to laboratory conditions.

4 Conclusion

This study investigated the influence of controlling CO_2 during the curing of natural hydraulic lime mortar on the mechanical properties of the material. The dynamic elasticity modulus, flexural strength, and compressive strength of mortar were compared for different curing conditions. A climate chamber at constant temperature and humidity, is typically used for curing lime mortars prior to testing, according to EN1015-11:2019 guidelines. Curing was conducted in a hermetically sealed chamber in this study, where the lack of an external supply of CO_2 led to reduced elasticity and strength properties. By controlling the CO_2 in the chamber, at an elevated level, mortar of significantly increased elasticity and strength properties was obtained. This mortar was also similar in strength to lab-cured mortar under ambient temperature, humidity, and CO_2 conditions.

This study demonstrated that the lack of CO_2 control during curing in current guidelines may lead to a significant underestimation of mortar stiffness and strength properties. A theoretical investigation is needed to obtain deeper insights into the relationship between curing conditions and mechanical parameters; the results from the literature provide contradictory indications of this relationship. A quantitative evaluation of carbonation depth may deliver further insight. Additionally, experimentally investigating the impact of curing under varying temperatures, humidity, and CO_2 levels on mechanical properties could offer valuable insights into hydration and carbonation kinetics. This could lay the groundwork for establishing new recommendations regarding CO_2 control during mortar curing and assist in the development of materials with improved carbon capture.

Acknowledgements: This research was conducted as part the UK EPSRC grant number EP/V048082/1 for the project on “MINT: Masonry in-situ testing and material identification”.

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