

Lime Based Construction Materials as a Carbon Sink

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Abstract. Lime is used in a variety of industrial sectors (e.g. construction materials, iron and steel industry, flue gas cleaning, etc.). By the thermal decomposition of limestone (CaCO_3), known as calcination, two products are obtained: CO_2 and quicklime, i.e. calcium oxide (CaO). There is a growing interest in quantifying and improving the potential of CO_2 absorption of lime containing products during their operational life.

The carbonation occurs during the lifetime of the lime application and it consists in the absorption of atmospheric CO_2 that closes the loop by forming calcium carbonate back. Thus, a portion of the CO_2 emitted during calcination is reabsorbed and stored in a permanent stable form.

A literature review was carried out on the Carbonation Rate (CR) of lime used in three different construction materials: air-lime mortars, mixed air-lime mortars and hemplime. Out of 205 scientific publications reviewed, only 57 provide information about CR, specifically 21 for air-lime mortars, 27 for mixed air-lime mortars and 9 for hemplime. CR is 80-92% for pure air-lime mortars, 20-23% for mixed ones and 55% for hemplime. For all the materials, the CR trend over time was also assessed, according to the Fick's law.

Introduction

Lime is a product which finds applications in a wide variety of industries: metals industry, construction materials sector, civil engineering, environmental protection, agriculture, and chemical industry. Lime can be used as quicklime (calcium oxide, CaO) or as hydrated lime (calcium hydroxide, $\text{Ca}(\text{OH})_2$) [1]. In construction materials, hydrated lime has been used as a binder in mortars, renders and plasters for millennia. It hardens slowly by reacting with carbon dioxide (CO_2) in the atmosphere to convert back into calcium carbonate (CaCO_3). Thus, the mortars made of lime are called air-lime mortar, while in mixed air-lime mortar the binder is a mixture of air-lime and a hydraulic binder. Hydrated lime is also used in the construction material hemplime, whose components are hemp shiv, which is the chopped woody core of the stalks of the hemp plant (*Cannabis sativa*), air-lime binder with pozzolanic, cementitious or hydraulic lime additives and in some cases surfactants [2].

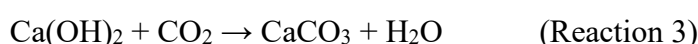
The quicklime is produced from the thermal decomposition of a rock rich in calcium carbonate, such as limestone, that decomposes in CaO and CO_2 according to the calcination reaction (Reaction 1).



Then, the hydrated lime, also called slaked lime, is produced by adding water to the quicklime forming calcium hydroxide ($\text{Ca}(\text{OH})_2$) as shown by Reaction 2.



During the lifetime of the hydrated lime application, the so-called calcium loop is closed because the hydrated lime reacts with the atmospheric CO_2 and forms calcium carbonate back according to the carbonation reaction (Reaction 3).



More precisely, during the carbonation in building materials, at first, the atmospheric CO₂ diffuses inside the material pores and dissolves in water in the pores, forming carbonic acid H₂CO₃. Then, the carbonic acid dissociates in bicarbonate (HCO₃⁻) and carbonate (CO₃²⁻) ions that form CaCO₃ reacting with the Ca²⁺ ions released by the dissolution in the pore water of Ca(OH)₂, that is contained in the binder. All processes involved in the carbonation can be synthesized in Reaction 3.

While the hydration and the carbonation are exothermal reactions, i.e., thermodynamically favourable, the calcination is endothermal, consequently a significant amount of energy is required. Thus, during lime production, CO₂ is emitted from the fossil fuel combustion for energy production and from the calcium carbonate decomposition. In this work, to distinguish the two CO₂ contributions, the CO₂ derived from limestone calcination will be referring to process CO₂ emission.

Since CO₂ is one of the greenhouse gas emission whose atmospheric concentration is increasing due to the human activities causing global warming and ocean acidification [3], CO₂ emission reduction and CO₂ removal from the atmosphere are essential to limit the average global temperature increase to 1.5°C above pre-industrial levels [4]. By the carbonation of lime a portion of the CO₂ process emission is reabsorbed from the atmosphere and stored in a permanent form as calcium carbonate. The ratio of the reabsorbed CO₂ through the carbonation over the process CO₂ emission was defined as Carbonation Rate (CR).

A review of the literature available on the carbonation of three construction materials was carried out with the aim to assess the CR of lime used in air-lime mortars, mixed air-lime mortars and hemplime. Further information about carbonation and CR in other lime applications found in literature are available in [5, 6].

Air-Lime Mortars

The literature reviews carried out by EESAC analysed the CR of pure air-lime based mortars [7]. After a pre-selection and check of 100 studies, only 21 publications were considered reliable and adequate for air-lime mortars.

From the literature review, it results that during the lifetime of the air-lime mortars the hydrated lime reacts with atmospheric CO₂ forming calcium carbonate carbonates, i.e. the contained calcium hydroxide carbonates. In the reviewed studies, the carbonation depth is measured with the phenolphthalein method, while the carbonation level, i.e. the percentage of initial slaked lime that carbonates, is measured by means of thermogravimetric analysis. The first method consists in spraying on mortar samples a phenolphthalein solution, that is a pH indicator, i.e. it indicates the acidity of a solution. The color of the phenolphthalein solution changes according to the pH of the solution. In this way, two areas are distinguishable: the uncarbonated (pink) and the carbonated (colorless) area, characterized by pH higher than 8.2 and lower than 8.2, respectively, since carbonation causes a pH reduction. The boundary which separates the two areas is called carbonation front, which moves over the time from the surface to the inner part of the construction product. The distance between the surface and the carbonation front is called carbonation depth, whose evolution in time can be described by Fick's law (Equation 1):

$$d = K \cdot t^{1/2} \quad (\text{Equation 1})$$

where d is the carbonation depth (mm); K is the carbonation coefficient (mm/year^{1/2}) and t is the time of exposure to CO₂ (years). From EESAC literature review, it results that K value is about 1 mm/(day)^{1/2} in the case of air-lime mortars natural carbonation. Assuming a time period of 100 years, the carbonation depth is equal to 190 mm according to (Equation 1) with $K=1$ mm/(day)^{1/2}. Furthermore, through thermogravimetric analysis, it results that in the carbonated area, i.e. from the surface to the carbonation front, 80-92% of the initial slaked lime is carbonated, while in the uncarbonated area, i.e. ahead of the carbonation front, the carbonation potential is in the range of 10-18%. As a result, pure air-lime mortars application with thickness lower than 190 mm can be considered 80-92% carbonated after 100 years [7].

Mixed Air-Lime Mortars

EESAC [8] analysed the CR of mixed air-lime mortars. 90 publications were found by searching in different bibliographical datasets for both types of mortars, but only 27 publications were considered reliable and adequate to assess the carbonation rate in mixed air-lime mortars. The mixed air-lime mortars consist of air-lime and Ordinary Portland Cement (OPC), that is the most commonly used hydraulic binder. The substitution of 20% of the binder amount with cement is sufficient to decrease the porosity and consequently the diffusivity of CO_2 in the mortars' pores. Thus, the CR decreases. Since K of OPC is about $0.25 \text{ mm}/(\text{day})^{1/2}$, i.e. 4 times lower than in air-lime mortars partially due to the decrease in pore size, it was estimated that partial substitution of air-lime with hydraulic binder causes a reduction of CRs by a factor of 2 to 5 compared to pure air-lime mortars [8]. Despotou et al. [9] have also investigated which factors affected lime-based mortars carbonation in addition to the thickness and the composition of the walls, i.e. the temperature and the water content. The temperature rise lowers the carbonation process, while the amount of water in the pores is pivotal because pore water is needed for dissolution of calcium hydroxide and CO_2 , but at the same time high water content hinders the CO_2 diffusion.

In conclusion, CR of mixed air-lime mortars is one fourth of the pure air-lime mortars one for an application with the same thickness, i.e. 20-23% after 100 years. The values of 20% and 23% as natural carbonation, are well aligned with a recent study conducted by Anderson et al. [10] on annual CO_2 uptake from concrete and mortar.

Hemplime

Different studies available in the literature have analysed the carbonation of lime binder in hemp-based construction material. Nine studies out of fifteen were found to provide adequate and reliable information. Six of them [11-16] evaluated the carbon footprint of hemplime material through the Life Cycle Assessment (LCA) methodology. One of those six studies [14], together with the study by Jami and Kumar [17] estimated the CR of lime binder through the X-Ray Diffractometric (XRD) analysis, while only the study by Chabannes et al. [18] carried out a Thermal Gravimetric Analysis (TGA) under natural and accelerated carbonation conditions. Only one study [19] measured the carbonation depth at different ages and assumed an equation which links carbonation depth to time. Thus, four out of nine are experimental studies.

Results of the three experimental studies on carbonation hemplime construction material are compiled in Table 1. Since carbonation is related to the content of calcium hydroxide ($\text{Ca}(\text{OH})_2$) in the binder, and it depends on its composition, all such parameters are reported in Table 1.

Experts of lime binder producers estimated that 249 kg CO_2 are absorbed for each metric tonne of lime binder, as reported in the study by Boutin et al. [11], assuming that only hydrated lime carbonates, the CR is 56%, i.e. more than half of the hydrated lime is carbonated after 100 years.

CR of $\text{Ca}(\text{OH})_2$ in hydrated lime was assumed equal to 100% by Ip and Miller [12] and Prétot et al. [13]. Thus, the hydrated lime CO_2 uptake over the building material lifetime is equal to the entire amount of CO_2 emitted during lime production. Prétot et al. [13] made a further assumption that also calcium hydroxide in hydraulic lime fully carbonates. Based on Prétot et al. study [13], Sinka et al. [16] LCA made the same assumptions. A lower CR was assumed by Lecompte et al. [15], based on thermogravimetric analysis results on carbonation of non-hydraulic lime mortars [20], i.e. 85% instead of 100%.

Arrigoni et al. [14] carried out a LCA study considering two types of binders and measured through XRD the CaCO_3 formed in hemplime samples after 8 months. It was assumed that calcium hydroxide in "dolomitic lime" fully carbonates (the $\text{Ca}(\text{OH})_2$ content of "dolomitic lime" is 50%), while cement carbonation was assumed 75% of the calcium in cement. As a result, the maximum CO_2 uptake per metric tonne of binder is 325 kg CO_2 and 297 kg CO_2 , respectively for mixed binder and pure "dolomitic lime" binder. Samples of the hemplime bricks with the two different types of binders were analysed. The content of $\text{Ca}(\text{OH})_2$ and CaCO_3 were measured at regular time intervals from hemplime bricks production up to 8 months. Samples at different depths were extracted, crushed and sieved in order to separate hemp shives from the binder. After 8 months, no carbonation was detected at the

inner depth. Thus, CR is 3.7% and 2.4% respectively for mixed binder and pure “dolomitic lime” binder. Given these low values, the assumption of a complete carbonation seems unrealistic.

Jami and Kumar [17] also evaluated through the XRD analysis the CO₂ uptake. The samples were hemplime cubes made of 118 g of binder which is composed of Ca(OH)₂ by 90%. Thus, the maximum CO₂ uptake is 70 g CO₂ per cube if only Ca(OH)₂ carbonates, calculated multiplying Ca(OH)₂ binder content by the ratio of the molecular weight of CO₂ over the one of Ca(OH)₂. After 28 days, the total CO₂ uptake is 14 g CO₂ per cube, i.e. CR is 20%. This estimation of the CR is much higher than the one found by Arrigoni et al. [14]. This difference can be partially explained by the different composition of the binders analysed by the two studies and by the fact that the faces exposed to air are five in Jami and Kumar study while it is only one in the study by Arrigoni et al. [14].

The study by Chabannes et al. [18] evaluated CR through TGA of concrete based on two different biogenic materials, i.e. rice husks and hemp. Once the cylindrical specimens were demoulded after 24 hours, they were cured under two different natural conditions for 10 months and accelerated carbonation conditions. Accelerated carbonation curing was preceded by an initial conditioning which consisted in storing the specimens during 40 days in the climate-controlled room at 20°C and 50% relative humidity. Then, the CO₂ curing was carried out in a glass enclosure fed by CO₂ for one month. In the glass enclosure, the room temperature was 20±2°C and the relative humidity was fixed at 65±5%. At first, a partial vacuum was created in the enclosure with a vacuum pump in order to reach an absolute pressure equal to 0.50±0.05 bar. CO₂ was then injected until the absolute pressure reached the atmospheric one, i.e. 1 bar. Thus, the volume concentration of CO₂ was maintained at 50%. The enclosure was refilled 42 times by CO₂. After the curing time, the samples were powdered and sieved before TGA. Between 600°C and 900°C the weight loss was assumed due to the release of CO₂ by CaCO₃. It was called loss of CO₂ (LCO₂) and measured as %. The amount of Ca(OH)₂ which is carbonated can be calculated subtracting the initial CaCO₃ content in binder (10%) to the CaCO₃ (%), calculated from LCO₂ and the molecular weights of CaCO₃ and CO₂, and multiplying it by the ratio of the molecular weight of Ca(OH)₂ over the one of CaCO₃ [18].

Once the amount of carbonated Ca(OH)₂ was obtained, CR can be evaluated as the ratio between the calculated carbonated Ca(OH)₂ and the initial Ca(OH)₂ in the binder which is 65%. Thus, the CR in accelerated conditions is 65.2%, while in natural conditions it is 33.7% and 54.5% respectively for indoor and outdoor sheltered condition.

With the exclusion of the rate found by Arrigoni et al. [14], that is very different from the other ones probably due to the different binder composition, the measured CR in natural conditions ranges from 20% to 54.5% after 28 days and 10 months, respectively. Since construction material lifespan is on the order of the years, the higher value should be considered. Furthermore, the upper value measured under natural conditions is similar to the one provided by the lime binder producers experts, i.e. 56%. The experimental works on carbonation of hemplime construction materials showed that the assumption of achieving full carbonation seems overestimated, and a CR lower than 100% should then be considered. On this matter, a higher CR (65.2%) can be reached under accelerated carbonation conditions.

Differently from the previous studies, Hirst et al. [19] measured at different ages up to 91 days the carbonation depth, i.e. the depth behind which lime is carbonated. Three different binders (THB-A, THB-B and BC) were used for producing the specimens. THB-B was the commercial binder composed mainly of CL90 air-lime with the addition of hydraulic components and other additives to improve porosity and consistence. THB-A is the same binder of THB-B blended with components for reducing setting time and making it more suitable for use in colder temperatures. BC was a binder based on natural hydraulic lime. The binder:hemp:water ratio and the mix density of the specimen are reported in Table 2. Specimen were cast in cylindrical cardboard moulds with diameter of 100 mm and height of 200 mm at temperature of 20±1°C and relative humidity of 60±5% for one week. Then, the specimens were demoulded and cured in a conditioning room at 20°C and 60% relative humidity for up to 91 days. At selected ages, samples were removed from the conditioning room and split in half and the exposed face sprayed with an ethyl alcohol phenolphthalein solution. Since hemplime material shows a more irregular carbonation boundary than one of lime mortars due to its porosity and the relatively large size of hemp shivs, the carbonation depth was measured from

eight equally spaced points around the cross-section of each specimen and then the mean of the measurements was calculated. Based on the measurements, the authors [19] assumed that the carbonation depth (d) is proportional to the square root of time as for mortars and cement materials (Equation 1). According to this assumption, the estimated carbonation constants are reported in Table 2. For all specimens except THB-A at a density of 220 kg/m^3 , the correlation was good ($R^2 > 0.9$), i.e. Equation 1 explains well the relationship among carbonation depth and time. Furthermore, the specimens are fully carbonated after 91 days, i.e. the carbonation depth is higher than half of the specimens' diameter (50 mm). It results that carbonation of hemplime begins at early age and it is slower at higher mix density.

Table 1. Binder CO_2 uptake and CR of hemplime constructions according to binder composition.

Reference	Chabannes et al. [18]			Arrigoni et al. [14]		Jami and Kumar [17]
Type of study	Experimental study			Experimental and LCA study		Experimental study
Geographical context	France			Italy		India
Type of construction	Cylindrical samples of Lime Hemp Concrete (diameter 11 cm and height 22 cm) Binder to hemp weight ratio = 2:1			Non-load-bearing wall (density of 330 kg/m^3 and U-value of $0.27 \text{ W/m}^2/\text{K}$) Construction method: blocks		Analysis on a cube of 343 cm^3 with dry density of 567.05 kg/m^3 (after 28 days)
Composition	Binder: 285 kg/m^3 Hemp: 142.5 kg/m^3 Water: 427.5 kg/m^3			Hemcrete block composition: Binder: 44.5 kg/m^2 Hemp: 31.4 kg/m^2 Water: 58.6 kg/m^2		Binder to Hemp weight ratio = 2.15 Water to Binder weight ratio = 1.3
Binder composition	50% hydraulic lime, 50% hydrated lime			Case A 80% dolomitic lime 20% cement	Case B 100% dolomitic lime	100% Hydrated lime (with 90% of $\text{Ca}(\text{OH})_2$ content)
Binder CO_2 uptake	ISC 130 kgCO_2 /t binder	OC 210 kgCO_2 /t binder	ACC 252 kgCO_2 /t binder	12 kg CO_2 /t binder (measured) 325 kg CO_2 /t binder (full carbonation)	7 kg CO_2 /t binder (measured) 297 kg CO_2 /t binder (full carbonation)	14 kg CO_2 /t binder (measured) 70 kg CO_2 /t binder (assumed)
Carbonation rate of lime	33.7% (measured)	54.5% (measured)	65.2% (measured)	3.7% (measured) 100% (assumed)	2.4% (measured) 100% (assumed)	20% (measured)
Carbonation time	10 months	10 months	40 days (initial curing) + 1 month (ACC)	240 days	240 days	28 days
Notes	Natural carbonation – Indoor Standard Condition (ISC) Temperature: 20°C Relative humidity: 50%	Natural carbonation – Outdoor exposed Condition (OC) Temperature: 0°C - 30°C Relative humidity: 45%-75%	Accelerated Carbonation Condition Temperature: $20 \pm 2^\circ\text{C}$ Relative humidity: $65 \pm 5\%$ [CO_2]: 50%v/v	Binder CO_2 uptake: 75% of CaO in cement carbonates and 100% of $\text{Ca}(\text{OH})_2$ in dolomitic lime carbonates. Dolomitic lime $\text{Ca}(\text{OH})_2$ content: 50%. Measurement through X-Ray Diffractometric (XRD) analysis (only one blocks face exposed to air)		Samples under ambient condition Measurement through XRD analysis

Table 2. Carbonation constant for different binders and different mix density due to different binder:hemp:water ratio.

Binder	Binder:hemp:water ratio (by weight)			Mix density (kg/m^3)	Carbonation constant K ($\text{mm/day}^{1/2}$)
	Binder	Hemp	Water		
THB-A	1	1	1	220	8.4
	1.5	1	2	275	6.7
	2	1	3	330	3.7
THB-B	1.5	1	2	275	5.8
BC	1.5	1	2	275	5.5

Conclusions

From different literature databases, peer-reviewed papers about the carbonation of lime used in construction materials were assessed. Out of 205 scientific publications reviewed, only 57 provide information about the CR, specifically 21 for pure air-lime mortars, 27 for mixed air-lime mortars and 9 for hemplime.

In conclusion, the carbonation of mortars is a well-known process and a wide scientific literature on the topic is available as shown by the review carried out by EESAC. CR of pure air-lime mortars is 80-92% after 100 years in application with thickness lower than 190 mm, while CR of mixed air-lime mortars lowers to 20-23% due to the porosity decrease caused by cement addition in mortars mix. Based on the results of this review, CR of hemplime found in literature is around 55% of the lime. From the results, the highest CR is achieved by pure air-lime mortars, while hemplime has a lower CR but has the additional benefit of storing carbon contained in the biogenic material which was absorbed from the atmosphere during the plant growth through photosynthesis.

It is expected that further studies will analyse the carbonation depth over time also in real cases, since there is a growing interest in construction materials with low carbon footprint, that can be used for storing CO₂ temporary or permanently [21]. Moreover, the results of this study could be the starting point for developing methodologies aimed to quantify the CO₂ uptake from carbonation because the carbonation begins to be taken into account as carbon sink in Global Carbon Budget [22] and it is proposed to be accounted in National Greenhouse Gases Inventories [10].

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