Carbonation Rates of Dry Ca(OH)$_2$ Mortars for CO$_2$ Capture Applications at Ambient Temperatures

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ABSTRACT: The carbonation rates of porous mortars, pellets, and extruded forms of Ca(OH)$_2$ were determined to investigate their suitability as functional materials for direct air capture. Samples of 4–15 mm thickness and porosities between 0.2 and 0.8 were tested by monitoring the progress of the carbonation fronts on time scales from 1 to 500 h. The evolution of such carbonation fronts was found to obey Fick’s diffusion law under all tested conditions. To reach CaCO$_3$ conversions higher than 0.6, a relative humidity above 50%, preferably between 80 and 100%, was required when using dry, low-grade slaked lime with a surface area of $18 \text{ m}^2/\text{g}$ as CO$_2$ sorbent. For modest relative humidities of 50%, higher grades of Ca(OH)$_2$ (i.e., with a surface area approaching 40 $\text{m}^2/\text{g}$) still allowed carbonation conversions above 0.8. The results confirm the applicability of these commercial solids for the direct air capture of CO$_2$.

INTRODUCTION

The carbonation of Ca(OH)$_2$-containing solids under ambient conditions has been studied by a number of researchers investigating the fundamentals of this gas–solid reaction. This type of carbonation has also been investigated because of its importance with regard to mortars and as the basis of a method for direct CO$_2$ capture from the atmosphere. Regarding the latter, we have recently investigated the possibility of deploying vast volumes of purpose-built passive carbonation infrastructures of porous Ca(OH)$_2$ stacked in such a way as to maximize the exposure of active Ca(OH)$_2$ surfaces to ambient air. The development of these applications could take advantage of the low specific cost of Ca(OH)$_2$ precursor (i.e., limestone) in many locations as well as the near-term availability of technologies for the oxy-calcination of CaCO$_3$ for cement production, and other direct air capture systems using CaO. Direct air capture processes of CO$_2$ powders.

Most of the works studying the carbonation reaction of Ca(OH)$_2$ in powder form with the ambient air suggest that the reaction mechanism under such operational conditions differs from the gas–solid reaction mechanism observed for the carbonation of Ca-based materials under temperatures above 100 °C. Beruto and Botter investigated the effect of relative humidity in the air to promote faster carbonation of Ca(OH)$_2$ powders, reporting extremely slow carbonation rates for the carbonation of Ca(OH)$_2$ under dry conditions. Following these observations, a reaction mechanism was proposed based on the adsorption of liquid water from the internal surface of the particles to promote the carbonation reaction at low temperatures. This reaction mechanism was also proposed by Dheilly et al. and Shih et al. when studying the reaction at temperatures below 90 °C and CO$_2$ concentrations from 0.03 %v up to 12 %v and has been observed as well for the carbonation of other Ca-based materials in the cement chemistry. In addition to this, several authors observed linear relations between the CO$_2$ uptake and the Ca(OH)$_2$ material surface area as well as improved reaction rates as the surface area increases. More recently, Erans et al. have investigated this reaction for direct air capture applications, requiring times up to 500–1000 h to carbonate 5 mm deep layers of CaO and Ca(OH)$_2$ powders. These reaction times are in agreement with those reported in the state-of-the-art carbonation of large concrete structures, the carbonation rates of such structures being controlled by pore diffusion.

In this work, we experimentally investigate the carbonation process of Ca(OH)$_2$ porous solids with potential applicability in passive carbonation schemes. To this end, pelleted samples of Ca(OH)$_2$ powder, dry mortars of Ca(OH)$_2$ and water, and extruded shapes of the same mortars (i.e., brick-shape) are
tested under different conditions to monitor the progress of the carbonation fronts over time and develop models that can be used for the design of large-scale systems for direct CO$_2$ capture with Ca(OH)$_2$.

**EXPERIMENTAL SECTION**

**Materials.** Ca(OH)$_2$ commercial powders were used to prepare all samples: dry slaked lime, used for construction applications, and a high-grade Ca(OH)$_2$, used for environmental applications. These powders have an average particle size of approximately 5–6 µm (measured by a Beckman–Coulter LS 13320 laser diffraction analyzer using ethanol as a dispersant agent), a true density of approximately 2.2 g/cm$^3$ (measured by helium pycnometry with an AccuPyc II-1340, Micromeritics Corp.), and a main impurity of CaCO$_3$ (between 3 and 8% wt, as measured by LECO CS130). Their surface areas and pore diameters are, however, very different: 18.2 and 39.3 m$^2$/g and 223 and 68 nm, respectively, for dry slaked lime and high-grade Ca(OH)$_2$, measured by Ar adsorption at −196 °C with an ASAP 2020 instrument, Micromeritics Corp., and applying the BET equation and mercury pycnometry with Autopore IV 9500 instruments, Micromeritics Corp.

**Preparation of Ca(OH)$_2$ Porous Samples.** Three different preparation methods were used to obtain porous solid samples with different initial porosities and characteristic lengths: the pelletization of dry powder (under a range of different pressures), the preparation of mortars with water that were then dried in molds to obtain the desired shapes, and the extrusion of mortars to obtain brick-like shapes.

Because the bulk open porosity of solids is known to be a major variable during the carbonation of Ca materials in ambient air, a wide range of porosities, between 0.2 and 0.8, were tested. To obtain highly porous samples (i.e., with porosities $\varepsilon_{\text{Ca(OH)}_2}$ between 0.6 and 0.8), dry Ca(OH)$_2$ powder was placed in cylindrical plastic vessels 1.5 cm in diameter and height, with the top open as the only contacting surface. Then, the powder was compressed using a dynamometer (SHIMPO FGE-100X) by applying forces of up to 200 N. The porosities of the resulting compacted beds were then dried in molds to obtain the desired shapes, and the extrusion of mortars to obtain brick-like shapes.

Figure 1. Photograph of a brick-shaped Ca(OH)$_2$ sample obtained from wet mortar extruded with the DEX LAB equipment and then dried.

The equipment (model DEX LAB) is based on a helix 80 mm in diameter capable of providing up to 7.5 kW and 80 bar for extrusion. The extrusion velocity can be adjusted to between 5 and 15 rpm, allowing for a maximum production of 500 kg/h. From wet mortar (with approximately 32% wt humidity), brick-like shapes were produced and then dried in an oven at 110 °C overnight. Figure 1 shows an image of one of the obtained extruded forms. The motivation for investigating these brick-shaped Ca(OH)$_2$ materials is the possible use of carbonated bricks as construction materials. However, for the scope of the investigation described in this work, these extruded brick shapes are not too different from the handmade mortars of similar dimensions and porosities when tested under ambient conditions, as will be shown below.

**Carbonation Tests.** Carbonation experiments were carried out on the samples in three setups: by simply allowing their carbonation to proceed in a test room, by arranging the samples in a sealed glass container surrounded by a thermostatic bath to ensure a homogeneous temperature and fluxed by air containing CO$_2$ with a high relative humidity, and by using a thermogravimetric analyzer (TGA) fully described elsewhere to test the carbonation of the powdered samples of parent solids and pellet samples of less than 100 mg. The tests under ambient room conditions typically lasted 500 h. Such a long carbonation time facilitates the frequent weighing of the samples and the use of destructive analysis by arranging the carbonation of several samples in parallel. The CO$_2$ concentration in the test room was 500 ± 25 ppm, CO$_2$ the relative humidity (RH) was 52 ± 3%, and the temperature was 19 ± 2 °C, as shown in Figure 2, which were measured by analyzing the ambient conditions using a PCE-AQD 10 Air Quality Data Logger device (for measuring over the ranges of 0 to 4000 ppm, CO$_2$, 10–90% RH and 0–50 °C). A fan was located in front of the samples to ensure a velocity of approximately 0.5 m/s in their proximity, thus allowing differential conditions with respect to the ambient air.

In the setup designed for carbonation under conditions of high RH, the samples were placed in a sealed glass container situated in a thermostatic bath (JP Selecta Unicronic 27 L for temperatures of 5–99.9 °C). The system was heated at controlled temperatures between 20 and 80 °C and flushed by a flow of about 200 L/h of compressed air. As shown in Figure 2, the concentration of CO$_2$ in the compressed air was 450 ±
35 ppm, CO₂, and the RH was below 2%. This air flow was then saturated with humidity by bubbling it in a distilled water glass placed in the thermostatic bath. To avoid condensation and heat losses, the top of the glass container was heated using a heating cord, and the whole system was covered with insulated material. Additionally, in this setup, some accelerated carbonation tests (typically less than 24 h to reach maximum conversions) were carried out with CO₂ concentrations 2 orders of magnitude larger than those in ambient air (8 and 12 %v CO₂), as will be discussed below.

When testing the solid samples either in the glass container or in the testing room, their lateral and bottom surfaces were covered, thus allowing the carbonation process to be monitored in just the axial direction (i.e., on one of the flat sides). For these samples, their weights were periodically monitored with a precision balance to calculate their Ca molar conversion to CaCO₃. Additionally, the carbonate contents of certain samples extracted at different experimental times were checked by LECO analysis in order to estimate their maximum Ca molar conversion to CaCO₃ ($X_{\text{max}}$). To visually observe the carbonation mechanism, some porous samples were axially cut, and a phenolphthalein solution (1% wt in 96 %v ethanol) was sprayed on their interior surfaces. Phenolphthalein is a sensitive pH indicator that turns colorless in acid solutions and pink in basic solutions, thus allowing us to distinguish between Ca(OH)₂ (colored) and CaCO₃ (colorless).

Small samples (i.e., pellets 6 mm in diameter) and Ca(OH)₂ parent solids (approximately 10–15 mg in powder form) were also tested in a TGA described elsewhere. The variations in sample weight over time under different temperatures (20 and 65 °C) and gas atmospheres (from compressed air with ppmv CO₂ up to 5 %v CO₂ in air and different RHs up to 95%) and a
total gas velocity around the sample of about 0.02 m/s) were continuously monitored during these tests.

Finally, crushing strength (CS) tests of the dried samples were conducted and the evolution of CS with the carbonation degree was measured using a SHIMPO FGE-100X dynamometer. The peak force (in Newtons) required to break the samples (in Newtons) required to break the samples was used as an indicator of their mechanical strength.

RESULTS AND DISCUSSION

The first set of carbonation experimental results obtained under the testing room ambient conditions (Figure 2 above) are presented in Figure 3, which was constructed by plotting the depth of the carbonated product layer, $z_{\text{carb}}$, as a function of time up to 500 h. Such depth has been determined from the weight measurements of the samples, considering the established fact that the carbonation of Ca materials reaches a maximum carbonation conversion $X_{\text{max}} < 1$ due to the formation of passivating CaCO$_3$ product layer on the Ca(OH)$_2$ surface

$$z_{\text{carb}} = \frac{(W_i - W_f)}{(M_{\text{CaCO}_3} - M_{\text{H}_2\text{O}})} \frac{Z_{\text{sample}}}{W_{\text{Ca(OH)2}}/W_{\text{CaCO3}}} X_{\text{max}}$$  \hspace{1cm} (1)

In Eq 1, $W_i$ and $W_f$ are the sample weights (in kg) measured at the beginning of the experiment and at different testing times, respectively; $M_i$ is the molar mass of each compound (in kg/kmol); $W_{\text{Ca(OH)2}}/W_{\text{CaCO3}}$ is the initial mass fraction of active Ca(OH)$_2$ (considering CaCO$_3$ as the main impurity in the powders); and $Z_{\text{sample}}$ is the axial height of the sample. The value of $X_{\text{max}}$ is determined experimentally by analyzing the carbon contents of the different layers of carbonated material with a LECO analyzer. As shown in Figure 3b, all values of $X_{\text{max}}$ reached a maximum stable value of 0.6 under this particular set of experimental conditions (a relative humidity of approximately 52% and a Ca(OH)$_2$ surface area of 18.2 m$^2$/g).

Figure 3a also includes lines that represent the predictions of a carbonation model considering the rate at which the carbonation front advances away from the exposed interface of the solid sample, with the ambient air being proportional to the flux of CO$_2$ given by Fick’s law

$$\frac{dz_{\text{carb}}}{dt} = \frac{(D_{\text{CO}_2}/\tau)\epsilon_{\text{carb}}C_{\text{CO}_2}}{z_{\text{carb}}\rho_{\text{Ca(OH)2}}(1 - \epsilon_{\text{Ca(OH)2}})X_{\text{max}}}$$  \hspace{1cm} (2)

where $D_{\text{CO}_2}$ is the diffusion coefficient of CO$_2$ in air (1.6 x $10^{-5}$ m$^2$/s at 20 °C), $\rho_{\text{Ca(OH)2}}$ is the molar density of Ca(OH)$_2$ (29.9 kmol/m$^3$), $\tau$ is the tortuosity factor, $C_{\text{CO}_2}$ is the CO$_2$ concentration (in kmol/m$^3$), and $X_{\text{max}}$ is the maximum carbonation conversion of the Ca(OH)$_2$ material. Because the reaction is assumed to be controlled by the diffusion of CO$_2$ in the stagnant volume of air contained in the porous carbonated layer resulting from the carbonation of Ca(OH)$_2$, the porosity of this carbonated layer ($\epsilon_{\text{carb}}$) is the one considered in Eq 2. This has been calculated assuming that there is no expansion of the porous solid during carbonation [i.e., $\rho_{\text{Ca(OH)2}}(1 - \epsilon_{\text{Ca(OH)2}}) = \rho_{\text{CaCO3}}(1 - \epsilon_{\text{CaCO3}})$], where $\epsilon_{\text{CaCO3}}$ is the porosity of a CaCO$_3$ layer and $\rho_{\text{CaCO3}}$ is the molar density of CaCO$_3$ of 27.1 kmol/m$^3$] and considering the maximum carbonation conversion, as follows

$$\epsilon_{\text{carb}} = 1 - (1 - X_{\text{max}})(1 - \epsilon_{\text{Ca(OH)2}}) - \frac{X_{\text{max}}\rho_{\text{Ca(OH)2}}}{\rho_{\text{CaCO3}}} (1 - \epsilon_{\text{CaCO3}})$$  \hspace{1cm} (3)

In an integrated form, Eq 2 gives the $t$ dependency similar to the one used in other studies of the slow carbonation of cementitious materials

$$z_{\text{carb}} = \sqrt{2(D_{\text{CO}_2}/\tau)\epsilon_{\text{carb}}C_{\text{CO}_2}t} - \sqrt{\rho_{\text{Ca(OH)2}}(1 - \epsilon_{\text{Ca(OH)2}})X_{\text{max}}}$$  \hspace{1cm} (4)
In Eqs 2−4, $\varepsilon_{\text{Ca(OH)}_2}$ refers to the open porosity of the Ca(OH)$_2$ samples. This value can be obtained by comparing the Ca(OH)$_2$ true density with the one calculated by measuring the weight ($W_0$) and total volume of the sample ($V_{\text{sample}}$); thus, $\varepsilon_{\text{Ca(OH)}_2}$ can be calculated as in Eq 5.

$$\varepsilon_{\text{Ca(OH)}_2} = 1 - \frac{W_0}{V_{\text{sample}} \rho_{\text{Ca(OH)}_2} M_{\text{Ca(OH)}_2}}$$

The best fits of Eq 4 to the experimental results noted in Figure 3a have been obtained when using tortuosity factor values of 1.20, 1.29, 1.30, and 1.85 for samples with $\varepsilon_{\text{Ca(OH)}_2}$ of 0.83, 0.71, 0.52, and 0.25, respectively. These fitted $\tau$ values are consistent with those reported in the literature for other porous media.

Figure 4 shows similar experiments under testing room conditions but using samples of high-grade Ca(OH)$_2$ (i.e., with a higher surface area of 39.3 m$^2$/g vs 18.2 m$^2$/g of those used in Figure 3) and $\varepsilon_{\text{Ca(OH)}_2}$ values of 0.80 and 0.66 and fitted $\tau$ values of 1.15 and 1.42, respectively. The most remarkable difference is the much higher carbonation conversion of this material due to the increased specific surface, with $X_{\text{max}}$ values reaching 0.85−0.90, as noted in Figure 4b. These results are consistent with those of previous studies on the carbonation of Ca(OH)$_2$ and can be explained by taking into account the impact on the maximum conversion of the passivating CaCO$_3$ product layer. From a practical point of view, the use of these commercial high-surface-area Ca(OH)$_2$ materials in direct air capture applications seems justified in view of both their larger CO$_2$-capture capacity and maximum carbonation conversion and their lower requirements of relative humidity to achieve said maximum conversions.

The capability of Fick’s law (i.e., Eq 4) to properly fit the observations in Figures 3 and 4, once the values of $X_{\text{max}}$, $\varepsilon_{\text{Ca(OH)}_2}$, and $\tau$ are considered, suggests that the carbonation reaction rates at the transition between the unreacted Ca(OH)$_2$ and the layer of carbonated solids at $X_{\text{max}}$ must be very fast compared to the rate of carbonation defined by Eq 4. In other words, it implies a rapid conversion, from 0 to $X_{\text{max}}$ of the Ca(OH)$_2$ grains near this reaction front. This is consistent with the results from studies of Ca(OH)$_2$ carbonation in which the high conversion of Ca(OH)$_2$ to CO$_2$ under suitable conditions of RH has been reported. As stated in the Introduction section, the importance of an intermediate H$_2$O adsorption step in the mechanism of carbonation, enhanced by high values of RH, has been discussed in other works by proposing that the carbonation of Ca(OH)$_2$ takes place in solution, with the CO$_2$ first dissolving in adsorbed layers of liquid water. Under low RH, the CaCO$_3$ formed would cover the Ca(OH)$_2$ surface more uniformly and passivate the material. In contrast, when RH increases (to values above 70%), the increased number of adsorbed water layers would provide a wider volume for the species to move all along the interfacial reaction regions and thus promote faster carbonation. Based on this, the proposed carbonation model of Eqs 2 and 4 has been applied only when the relative humidity, RH $\geq$ 50%.

To confirm the reported trends, we carried out dedicated experiments using TGA and intensified carbonation conditions, as shown in Figure 5. As seen, RHs close to saturation allowed a molar Ca conversion to CaCO$_3$ over 0.9 in less than 10 min (see Figure 5a for the TGA of the carbonation at 65 °C and 5 %v CO$_2$). After a first rapid step, the carbonation of Ca(OH)$_2$ enters a slower phase. As the RH decreases, a transition in the carbonation rate appears at conversions between 0.2 and 0.3, with the rate of conversion progressively decreasing over time but still evolving slowly toward the maximum level of conversion reachable in time scales of several hours (not shown in the figure for simplicity). In contrast with the previous results, dry air with 5 %v CO$_2$ (but RH = 0%) is unable to facilitate the carbonation of samples beyond conversions of 0.05.

This strong impact of RH on carbonation rates can also be observed in Figure 5b, although at different time scales. In this case, a Ca(OH)$_2$ porous pellet is carbonated with air, and abrupt changes in the slope of the conversion curve were measured when changing from dry compressed air to fully

![Figure 5. Ca(OH)$_2$ molar conversion to CaCO$_3$ vs time for different relative humidities (RHs) for high surface area Ca(OH)$_2$ in the TGA: (a) powder at 65 °C and 5 %v CO$_2$ and (b) pellet of 6 mm diameter carbonated under compressed air (450 ± 35 ppm CO$_2$) at 20 °C.](https://doi.org/10.1021/acs.iecr.2c01675)
saturated air. From a practical point of view, it is clear that RH values > 80% guarantee fast carbonation rates of any Ca(OH)\(_2\) material and the highest values of \(X_{\text{max}}\) at the grain level, making the overall carbonation process controlled by diffusion, as shown in Figure 5. Consistent with the proposed mechanism,\(^1\)\(^-\)\(^3\)\(^,\)\(^5\) and for long reaction times, it is also beneficial to use Ca(OH)\(_2\) materials with high surface areas when there is a need to moderate the RH values to just over 50% (e.g., to minimize the large consumption of water if a direct capture system is located in desert regions), as observed in the results reported in Figure 4. However, due to the extremely slow carbonation rate of Ca(OH)\(_2\) observed in dry air conditions,\(^1\)\(^-\)\(^4\)\(^,\)\(^6\)\(^,\)\(^9\) treating dry air should be avoided in future capture systems relying on the carbonation of Ca(OH)\(_2\).

To further confirm these observations with extruded shapes at the macroscopic level, experiments were carried out in a sealed glass container fluxed with compressed air saturated with humidity (i.e., RH > 95%) on extruded materials of different porosities (i.e., with measured \(\epsilon_{\text{Ca(OH)2}}\) values of 0.51, 0.44, and 0.34). These were prepared using slaked Ca(OH)\(_2\) with a modest initial surface area. These tests revealed that RHs close to saturation enhanced the overall degree of carbonation of the sample by increasing the value of \(X_{\text{max}}\) to approximately 0.9 as measured by LECO analysis, making the use of high-surface-area Ca(OH)\(_2\) unnecessary. As shown in Figure 6, the curves of \(z_{\text{carb}}\) vs time can be well adjusted with Eq 4, as with previous samples, by fitting the \(\tau\) values to 1.42, 1.49, and 1.95, respectively.

To assess the validity of Fick’s diffusion model under a wider range of conditions and time scales, accelerated tests using air with 2 orders of magnitude larger concentrations of CO\(_2\) (12 % v CO\(_2\) and 8 %v CO\(_2\)), characteristic of combustion flue gases, were carried out in sealed glass containers under humidity saturation conditions at 25 and 80 °C. Under these conditions, less than 24 h is sufficient to ensure the complete conversion of the samples, as shown in Figures 7a and 8 (for extruded materials, as in Figure 6). Remarkably, the fitting quality of Fick’s model in Figure 7a was retained for samples prepared from Ca(OH)\(_2\) powder with porosities of about 0.4−0.5 when using tortuosity factors comparable to those used in previous figures (i.e., \(\tau\) of about 1.4−1.5). Figure 7b also shows the clear evolution of the...
depth of the carbonated layer over time by spraying a phenolphthalein solution over the axial section of the samples.

In the case of extruded materials of Figure 8, larger \( \tau \) values are required to fit the experimental results using Eq 4 when compared to the non-extruded. Values of 1.62, 1.75, and 3.05 have been used to fit the experimental results obtained for samples with porosities of 0.51, 0.44, and 0.34, with the differences of special relevance in the case of the sample with the lowest porosity. This can be attributed to the existence of a certain fraction of inaccessible voids resulting from the higher pressures needed in the extrusion process to reach these lower porosities. Other effects, such as bottlenecks, could also reduce the effective porosity, thus increasing the tortuosity factor.

In previous Figures 3, 4, and 6−8, the tortuosity factor in Eq 4 was used as an adjustable parameter to obtain the best fit of the Fick’s model to the experimental results. This parameter depends on porous media characteristics such as the porosity, pore diameter, channel shape, and so forth. In the absence of experimental data to fit such tortuosity factor, \( \tau \) can be estimated from empirical equations such as that proposed by Bruggeman (relating tortuosity and porosity as \( \tau = 1/\sqrt{\varepsilon_{Ca(OH)_2}} \)). As shown in Figure 9, there is a good agreement between the fitted values from this work and those predicted by the empirical equation, with the main deviation observed in the extruded sample with the lowest porosity. As mentioned above, such discrepancy can be attributed to the formation of inaccessible voids during the extrusion process.

To complete the characterization of the carbonating samples, crushing strength measurements as a function of carbonation conversion were performed for some pelletized samples. As shown in Figure 10, the measured C\( \text{s}s \) significantly increased with the conversion of Ca(OH)\(_2\) to CaCO\(_3\). This improvement in C\( \text{s} \) with increasing levels of carbonation is consistent with observations of other Ca-based carbonating materials. Although it is beyond the scope of this work to elaborate on large-structure manufacturing aspects, the results of Figure 10 support the viability of future direct CO\(_2\)-capture systems relying on the carbonation of large-scale Ca(OH)\(_2\) structures arranged to be exposed to air.

**CONCLUSIONS**

The carbonation of Ca(OH)\(_2\) porous solids on the millimeter-to-centimeter scale follows Fick’s diffusion law under a variety of CO\(_2\) concentrations in air (from 12 %v to ambient conditions at approximately 450 ppmv). The advance with time of the carbonation front for Ca(OH)\(_2\) samples in the form of pellets, dry mortars, and extrudes with porosities between 0.2 and 0.8 can be well fitted in most cases when considering the governing diffusion equation, the maximum carbonation
conversion of the Ca solids, the porosities of the samples, and the tortuosity factor. The relative humidity (RH) in the air played an essential role in maximizing the Ca(OH)$_2$ solid carbonation conversion, with an RH of >50% required to achieve maximum conversion values of 0.6. This maximum value of conversion can be increased by up to approximately 0.9 by using high-grade Ca(OH)$_2$ (i.e., with specific surface areas of approximately 40 m$^2$/g compared to the less than 20 m$^2$/g measured for dry slaked limes) or RHBs in the air close to saturation (i.e., RH > 95%).

The authors declare no competing financial interest.

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## NOMENCLATURE

| Symbol | Definition |
|--------|------------|
| $C_{CO_2}$ | CO$_2$ concentration, kmol/m$^3$ |
| $D_{CO_2}$ | diffusion coefficient of CO$_2$ in air, m$^2$/s |
| $M_i$ | molar mass of compound $i$, kg/kmol |
| $t$ | time, s |
| $V_{sample}$ | total volume of the sample, m$^3$ |
| $W_0$ | initial sample weight, kg |
| $w_{Ca(OH)_2}$ | mass fraction of active Ca(OH)$_2$ |
| $W_t$ | sample weight at time $t$, kg |
| $X_{CaCO_3}$ | maximum Ca molar conversion to CaCO$_3$ |
| $Z_{sample}$ | sample axial height, m |
| $\varepsilon_{Ca(OH)_2}$ | open porosity of the samples according to Eq. 5 |
| $\varepsilon_{carb}$ | porosity of the carbonated layer according to Eq. 3 |
| $\rho_i$ | molar density of compound $i$, kmol/m$^3$ |
| $\tau$ | tortuosity factor |

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