Carbon Dioxide: A Raw Material for Cementitious Mortar †

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Abstract: Buildings and infrastructures can absorb CO₂ from the atmosphere because of the carbonation process that affects the calcium hydroxide of concrete elements. The aim of this research project is to initiate the absorption at casting by adding dry ice pellets to cement-based mortars. Test results demonstrate that the flexural and compressive strength of the mortars are not modified by this addition. Conversely, due to the presence of CO₂, the standard deviation of strength reduces with respect to that measured in plain mortars. Thus, carbon dioxide can be considered a valuable resource that improves the mechanical behavior of construction materials.

Keywords: cementitious mortar; dry ice pellets; compressive test; three-point bending test

1. Introduction

Among all the materials, cement and steel are two of the most important sources of carbon dioxide. It is sufficient to think that cement manufacture releases about 7% of global CO₂, half of which can be attributed to the construction industry [1]. On the other hand, cement-based composites are prone to carbonate and, therefore, tend to absorb the carbon dioxide from the atmosphere. Hence, to compensate for the emission of cement production, some procedures have been proposed with the aim of absorbing carbon dioxide during the production of cementitious composites. More precisely, three different carbonation approaches can be found in the current literature.

The first, called Solidia Cement [2], a specific binder composed of calcium silicate with a low lime content, is more capable of absorbing CO₂ than ordinary Portland cement. When using this approach, concrete must be exposed to carbon dioxide for at least 48 h. In the second approach, called pre-carbonation [3], the goal is to obtain limestone by carbonating the calcium hydroxide. Then, the mixture of water and limestone is added to the concrete components (i.e., cement, water, aggregate, and additives). Finally, in the so-called “CarbonCure” method, carbon dioxide is added when concrete components are mixed [4].

Following the last approach, Fantilli et al. [5] have recently introduced a new cement-based mortar in which carbon dioxide, in the form of dry ice pellets, is added to the mixture like a raw material. By means of this addition, the maximum content of CO₂ that is absorbed by the mortar system is equal to 1.6% of the mass of the CEM I binder. Nevertheless, to consider carbon dioxide an effective raw material, like those currently used to cast cement-based products, it is necessary not only to simplify the procedure for absorbing CO₂ but also to demonstrate that CO₂ can improve the performance of mortars.

For these reasons, a new experimental campaign, performed on 60 mortar prisms, tested in bending moment and compression (in accordance with EN 196-1 [6]), is described in the following sections. The aim was to measure the effect induced by the addition of carbon dioxide both on the strengths, in bending and compression, and on the statistical distribution of these mechanical performances.
2. Material and Methods

Cement, tap water, normalized sand, and CO\textsubscript{2} were the components of the mortars herein investigated. The type of cement was CEM I 42.5R, whereas the carbon dioxide was provided by SIAD s.p.a in the form of dry ice pellets (with a diameter of 3 mm). Dry ice was obtained through the expansion of liquid carbon dioxide. During this physical conversion, each kilogram of liquid CO\textsubscript{2} formed about 0.6–0.52 kg of gas and 0.4–0.48 kg of solids, in the form of snow carbon dioxide. Such snow was successively compressed to form the pellets, which were added to cement-based mortars at a temperature of about −78 °C. The mixture also included CEN Standard sand consisting of siliceous rounded particles, whose size distribution lies within the limits given by EN 196-1 [6].

With all the above-mentioned materials, two series of mortars were cast. As shown in Table 1, in the mortar called B- Carbon, 72 g of CO\textsubscript{2} (i.e., 1.6% of the mass of the cement) were added with respect to the standard mortar (called A-plain). With each mortar, 30 prims (40 × 40 × 160 mm\textsuperscript{3}) were cast (see Figure 1) by means of polystyrene molds. The specimens were cured in the molds for 28 days at a constant temperature of 20 °C (relative humidity = 50%) and, after demolding, were tested in three-point bending and compression following the EN 196-1 [6] rules.

Table 1. The mortars used to cast the specimens.

| Series     | Cement (g) | Water (g) | Sand (g) | CO\textsubscript{2} (g) | CO\textsubscript{2}/Cement (%) |
|------------|------------|-----------|----------|-------------------------|-------------------------------|
| A-Plain    | 4500       | 2250      | 13,500   | 0                       | 0                             |
| B-Carbon   | 4500       | 2250      | 13,500   | 72                      | 1.6                           |

![Figure 1. The prisms tested in three-point bending and compression following the EN 196-1 [6] rules: (a) the prisms of the series A-plain; (b) the prisms of the series B-carbon.](image)

3. Results and Discussion

From the three-point bending tests, the flexural strength $\sigma_{\text{flex}}$ of all the 30 specimens of each mortar was measured, whereas the compressive strength $\sigma_{c}$ was obtained with the uniaxial compression test. The latter was performed on 60 halves of the prisms previously tested in bending.
Figure 2 illustrates the statistical distributions of both $\sigma_{\text{flex}}$ and $\sigma_{c}$, related to the mortar of series A-Plain (Figure 2a,b) and to that of series B-Carbon (Figure 2c,d), respectively.

![Figure 2](image_url)

**Figure 2.** The statistical distributions of strength: (a) flexural strength in the mortar A-Plain; (b) compressive strength in the mortar A-Plain; (c) flexural strength in the mortar B-Carbon; (d) compressive strength in the mortar B-Carbon.

With respect to the series A-Plain, the use of CO$_2$ (in the series B-Carbon) does not modify the modal values of both compressive and flexural strength, but the probability density increases. Moreover, the dispersion of strength around the modal values tends to reduce in the mortars containing CO$_2$. To better quantify these aspects, the density probability function $f(x)$ of the normal Gaussian distribution $f(x)$ can be computed (when the variable $x$ is equal to $\sigma_{\text{flex}}$ and $\sigma_{c}$, respectively) and is depicted in Figure 2:

$$f(x) = \frac{1}{\delta \sqrt{2\pi}} e^{-\frac{(x-\mu)^2}{2\delta^2}}, \quad (1)$$

The average value $\mu$ and the corresponding standard deviation $\delta$ are:

$$\mu = \frac{\sum_{i=1}^{n} x_i}{n} \quad \delta = \sqrt{\frac{1}{n-1} \sum_{i=1}^{n} (x_i - \mu)^2} \quad (2)$$

where $N = $ number of specimens used to measure the strength (i.e., $N = 30$ in bending and $N = 60$ in compression). For both the series, the values of $\mu$ and $\delta$ are reported in Table 2.
Table 2. The statistical parameters of the mortars investigated herein.

| Mechanical Property | $\sigma_{\text{flex}}$ (MPa) | $\sigma_c$ (MPa) | Degree of freedom | $F$-test | Equal variances |
|---------------------|-----------------------------|-----------------|-------------------|----------|----------------|
| Type of Mortar       | A-Plain | B-Carbon | A-Plain | B-Carbon |
| $\mu$ (MPa)          | 4.59    | 4.48    | 30.0    | 29.4    | 29             | $F = 1.63 < f_{0.05} = 1.86$ | Yes |
| $\delta$ (MPa)       | 0.319   | 0.250   | 2.40    | 1.53    | 59             | $F = 2.44 < f_{0.05} = 1.54$ | No |

The average values of compressive and flexural strength are nearly the same in both the mortars, whereas the lowest values of the standard deviation are those of the mortar B-Carbon, in which CO$_2$ is added. These results are in accordance with those obtained by Monkman & Cail [7], regarding the compressive strength of concrete samples in which CO$_2$ was added using the CarbonCure procedure. Moreover, the results of the $F$-test [8], summarized in Table 2, show that $F$ is smaller than the critical value in the 95% confidence interval ($f_{0.05}$) only in the case of flexural strength. On the contrary, the compressive strength of A-plain and B-carbon do not have equal variance with a 95% confidence interval. As $\sigma_c$ is the most important parameter that characterizes cement-based mortars, it is clear that statistically significant differences exist when dry ice pellets are added to a mortar system.

4. Conclusions

According to the experimental results illustrated in the previous sections, the following conclusions can be drawn:

- Carbon dioxide can be absorbed by cement-based mortars through a very simple approach, which can be easily implemented at the construction site. Specifically, CO$_2$, in the form of dry ice pellets, was added to concrete mixture like a common additive.
- If the mass of the added CO$_2$ is 1.6% of the cement, the average values of both flexural and compressive strength of the cementitious mortars are not modified.
- However, such a content of carbon dioxide leads to a remarkable reduction in strength distribution (i.e., a reduction in the standard deviation) with respect to that measured in plain mortars.
- The analysis of variance shows significant improvements in the mortars containing CO$_2$. Accordingly, the latter can be considered as a raw material for cement-based composites.

The effect produced by the addition of carbon dioxide on mortars containing other types of cement as well as other waste materials will be investigated in future experimental and theoretical analyses.

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