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Accelerated carbonation of recycled concrete aggregates and model materials

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Abstract. The accelerated carbonation of recycled concrete aggregates (RCA) has been suggested to improve their properties. Several physical and mechanical phenomena occur during the carbonation of adhered cement mortar, but their effects are scarcely distinguished and correlated. In this study RCA and model materials (spheres) have been exposed to carbonation, and then characterized at different times to monitor the evolution of their macroscopic properties (porosity and absorption kinetics) as a function of their carbonation degree. The decrease in porosity and saturation rate is actually observed for model materials whereas for RCA, the initial natural carbonation limits the storage of CO₂ and consequently the evolution of their properties.

Keywords: Carbonation, Recycled concrete aggregates, Model materials, Porosity, absorption kinetics.

1 Introduction

While a critical shortage of natural aggregates exists for the production of new concrete, the huge amount of construction and demolition waste, including cement-based materials, generates serious ecological issues. A possible solution is to use this concrete waste as aggregates to formulate new concrete.

Compared to natural aggregates (NA) used in concrete, recycled concrete aggregates (RCA) generally have lower quality [1] [2]. The concrete strength thus decreases when the NA is totally or partially replaced by the RCA [2]. Therefore, the use of recycled aggregates in the construction industry is limited [3]. In recent years, adopting accelerated carbonation to improve the quality of RCAs has been proposed [4] [5]. This technique aims to densify the old cement mortar attached to the RCA and to enhance their physical and mechanical properties [2]. Carbonation of RCA improves not only their own properties and those of new concrete but it also contributes to mitigate the greenhouse effect caused by the emission of carbon dioxide [3].

It is worth noting that RCAs show significant variability coming from the nature of the crushed concrete, the heterogeneity of the distribution of the adherent cement

paste in the different granular classes and from the inevitable presence of bricks and other elements such as glass or wood incorporated into the RCAs [6]. These significant heterogeneity and variability may question the representativeness of the results. Considering those aspects, model materials representative of RCAs are tested in the present study. Almost all previous studies have focused on the evolution of the saturation water rate of RCA during their carbonation. The evolution of the absorption kinetics is rarely studied although it represents an essential parameter to be taken into account during the manufacture of concrete and can have consequences in terms of recommendations for the pre-wetting of carbonated RCA. This study allows to follow the evolution of the absorption kinetics on model materials as they carbonate.

2 Materials and methods

2.1 Materials

Mortar spheres

Spherical mortar samples (Fig. 1) are manufactured to represent recycled aggregates and more precisely the mortar around natural aggregates in RCA. They make it possible to monitor the evolution of the composition and the microstructure of RCA mortar as the carbonation progresses.



Fig. 1 4 cm diameter mortar spheres (modeling material).

Portland cement CEM I 52.5 from Villiers-au-Bouin, France was used for the casting of the spheres manufactured in this study (Table 1). Its density is 3.13 g/cm^3 and its specific surface area was $3900 \text{ cm}^2/\text{g}$. Natural siliceous sand (0-2 mm) with a specific gravity of 2.61 was used as aggregates. Cement mortar was prepared with mixture proportions of cement: sand: water of 1: 2.6: 0.6.

Table 1 : Chemical composition of the used cement.

| Oxide constituents (%) | | | | | | | |
|------------------------|------------------|--------------------------------|-----|--------------------------------|-----------------|------------------|------------------|
| CaO | SiO ₂ | Al ₂ O ₃ | MgO | Fe ₂ O ₃ | SO ₃ | TiO ₂ | K ₂ O |

| | | | | | | | |
|-------|-------|------|------|------|------|------|------|
| 63.70 | 19.60 | 4.50 | 3.90 | 2.30 | 2.60 | 0.20 | 0.70 |
|-------|-------|------|------|------|------|------|------|

The mortar spheres were made using spherical silicone molds ($\text{Ø} = 40 \text{ mm}$) placed in parallelepiped PVC molds to facilitate pouring. The manufacture of PVC molds was carried out using a 3D printer (Fig. 2).

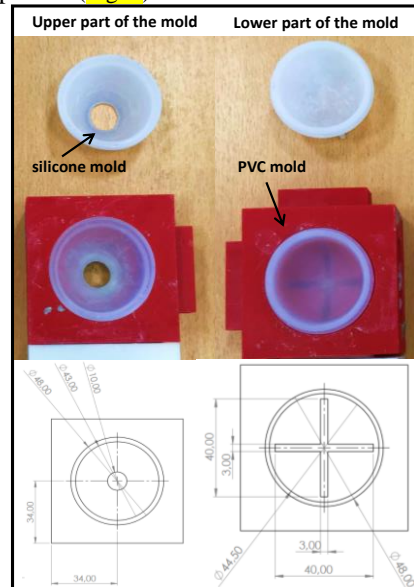


Fig. 2 upper and lower part of the mold of 4 cm

The samples were demolded after 24 h and then soaked in a tap water tank at 23 ± 1 °C for 90 days to become more representative of RCA. Apart from the cure underwater for 90 days, the spheres do not undergo any real aging.

Recycled concrete aggregates

Class 4/22.4 recycled aggregates are used. They come from a Lafarge site in Nantes and have a water absorption of 4.3% and a density of 2.3.



Fig. 3 recycled concrete aggregates

Table 2 : Gradation of recycled aggregates.

| Diameter (mm) | Cumulative residue on sieve (%) |
|---------------|---------------------------------|
| 4 | 5.5 |
| 6.3 | 14.2 |
| 11.2 | 51.6 |
| 16 | 74.8 |
| 20 | 89.4 |
| 22.4 | 94.3 |

2.2 Methods

Accelerated carbonation procedure (treatment)

Before carbonation test, a preconditioning is carried out. Recycled aggregates and model materials were dried at 40°C for 10 days, and then stored for another 10 days in an air-conditioned room (T=20°C, relative humidity RH =55%) to homogenize the internal humidity. The purpose of such pretreatment was to reach a hydric state of samples in equilibrium with the relative humidity of the environment chosen for the carbonation. The aggregates and spheres were then subjected to a diffusion of CO₂ in a climatic chamber connected to a CO₂ tank at 20°C, 65% relative humidity and 15% of CO₂. The concentration of CO₂ in the chamber was controlled by an automatic CO₂ regulator at 15%. This percentage was chosen so that it is equal to the percentage of CO₂ in cement plant fumes, which could be used for the accelerated carbonation of RCA.

CO₂ uptake measurement

In this study, the oven is used to determine the amount of CO₂ fixed **after** carbonation of recycled aggregates and model materials. Indeed, the TGA can be used for this purpose, but due to the high variability and heterogeneity of recycled aggregates and

the low mass that can be tested in the TGA, we opt to use the oven as it allows us to test a more representative sample.

The KITTEC oven used in this study can heat up to 1300°C with an adjustable heating rate and a possible stabilization of the temperature (isothermal sequences). Before starting the oven test, the sample is placed consecutively in three ovens regulated at 40°C, 80°C and 105°C respectively. The sample remains in each oven until its mass stabilizes, then it is transferred to the higher temperature oven. When the mass at 105°C becomes stable, the oven test begins. The samples are heated to 1000°C with a heating rate of 10°C/min and stabilized at specific temperatures. The temperatures chosen for the isothermal sequences are the critical values often used to dehydrate portlandite (400°C, 475°C, 500°C, 550°C, and 600°C), and to decarbonate calcium carbonate polymorphs (675°C, 730°C, 800°C, and 875°C). At these temperatures, oven temperature is stabilized for 30 min, then the sample is weighed every 10 min. The temperature rise is resumed when the difference between two consecutive mass measurements is less than 0.5%.

It is important to note that all mass percentages used below are calculated relative to the mass of the sample dried at 950°C.

In order to determine the amount of carbon dioxide captured and stored **after** accelerated carbonation C_{AC} , two analyses were performed, before and after accelerated carbonation. The C_{AC} can be calculated using the following formula:

$$C_{AC} \text{ (g/kg RCA)} = (\%M_c - \%M_{c0}) * 10$$

Where $\%M_c$ and $\%M_{c0}$ represent the mass percentage of carbon dioxide present in the cementitious material before and after accelerated carbonation, respectively. These percentages are calculated from the amounts of calcium carbonate obtained by oven analysis before ($\%M_{CaCO_3}$) and after ($\%M_{CaCO_3}$) accelerated carbonation and relative to the dry mass at 950°C. **The interval 600-875°C was allocated to $CaCO_3$ decarbonation.**

$$C_{AC} \text{ (g / kg RCA)} = (\%M_c - \%M_{c0}) * 10 = ((\%M_{CaCO_3} - \%M_{CaCO_30}) * 44) / 100 * 10$$

Porosity

Connected open porosity is determined under vacuum according to the French standard NF P18-459.

Absorption kinetics

The hydrostatic weighing procedure proposed by Bendimerad [61] was adopted during this work. The calculation of water uptake is based on the change in apparent mass of the sample in water. The procedure consists in heating the sample in an oven at 80°C until the sample mass stabilizes. The dry mass is noted M_d . According to the European standard EN 1097-6, the dry mass is obtained at a temperature of 110° C; however, the choice of the temperature of 80° C aims at limiting the loss of water linked chemically to the cement paste of the RCA and the changes in their microstructure. Once the mass is stable, the sample is placed in a small container suspended from a balance connected to a computer. At time $t = 0$, the container is immersed in a water bath at 20 °C. Data acquisition starts 5 min after the first contact with water

($M_w(t)$) and will be stopped when there is no significant variation of the apparent mass, $\Delta m < 0.05\%$ after a time t_f , this mass is noted M_f . The RCA will then be superficially dried until the visible water films disappear, then the sample is weighed to obtain the saturated dry surface mass $M_{sat\ s,s}$. The final absorption at t_f is calculated according to equation (Eq (1)) based on EN 1097-6 (NF EN 1097-6, 2001), noted $Ab(t_f)$.

$$Ab(t=t_f) = (M_{sat\ s,s} - M_d) / M_d \quad (1)$$

In this study, the absorption at $t = 5$ min is calculated according to Eq (1).

$$Ab(t=5\text{min}) = Ab(t=t_f) - (M_w(t=t_f) - M_w(t=5\text{min})) / M_d \quad (2)$$

The water uptake at each time step can be calculated from the following equation:

$$Ab(t) = Ab(t=5\text{min}) + (M_w(t) - M_w(t=5\text{min})) / M_d \quad (3)$$

3 Results

3.1 CO₂ fixed during carbonation

CO₂ storage in recycled aggregates and model materials increases **after** carbonation. This increase is very significant for model materials but is small in the case of RCA (Fig. 4). The measured amounts of stored CO₂ are consistent with those reported in the literature [7] [8]. The difference in storage capacity between RCA and model materials is due on the one hand, to the considerably higher percentage of paste in the model materials (37%) compared to RCA (17%), and on the other hand to the significant natural carbonation of RCA prior to accelerated carbonation. **In fact, the model materials were subjected to carbonation immediately after preconditioning while the RCA was stored for one year in the open air before being collected and tested.** This initial natural carbonation decreases the fixation capacity of CO₂ since a large part of the **carbonatable hydrates** has already been carbonated and, on the other hand, it could lead to the clogging of the outer layer and thus hinder the diffusion of carbon dioxide.

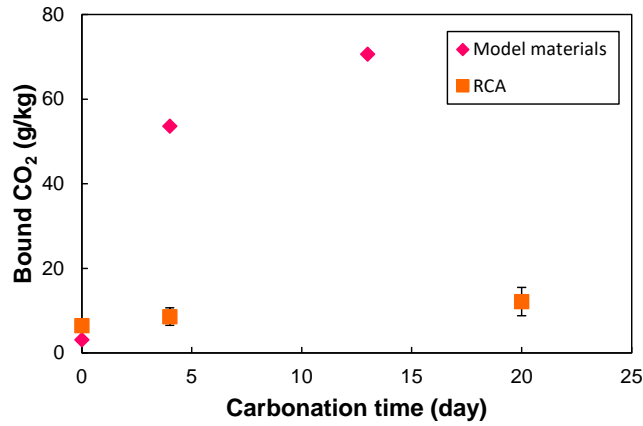


Fig. 4 Bound CO₂ stored in recycled aggregates and model materials during carbonation

3.2 Evolution of porosity and water absorption during carbonation

Fig. 5 shows the results of analyses carried out on model materials at different carbonation times for absorption kinetics. It shows a decrease in **absorption rate** as carbonation progresses. It goes from 8.5% to 5.8% after 14 days of carbonation. In addition to the decrease in the **absorption rate**, it is worth noting that the absorption kinetics become slower; the time to reach the maximum absorption becomes longer as the carbonation proceeds. These observations can be explained by the evolution of the distribution of the pores, which will take place during the carbonation. Indeed, as carbonation proceeds, mesopores with diameter $D < 50$ nm will potentially be clogged, especially on the outer zone of the grain subjected to carbonation [14], making it more difficult for water to diffuse to the center of the sample and consequently increasing the time needed for saturation.

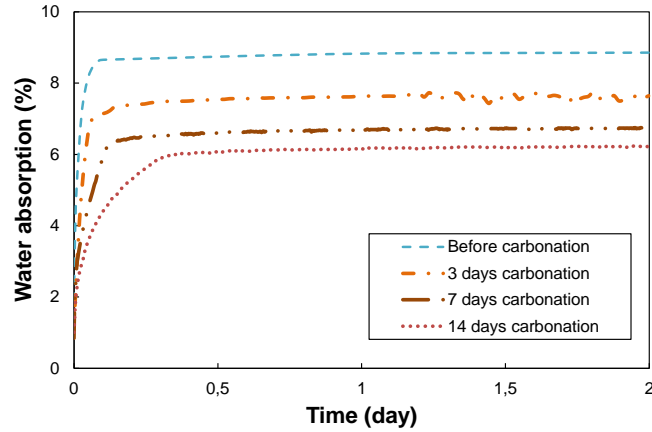


Fig. 5. Evolution of the absorption kinetics of model materials during carbonation

Fig. 6 and Fig. 7 shows a decrease in the connected open porosity and an increase in the time required to reach 80% of the final water uptake as carbonation progresses for RCA and model materials spheres. The decrease of the porosity and water content at saturation are expected and in agreement with the literature [9] [10] [11]. **During portlandite carbonation** the volume of products formed (calcium carbonate) is greater than the volumes of carbonated hydrates initially present in the cement matrix which causes the clogging of pores and the reduction in total porosity [12] [13]. **In addition, Ca transport from the core to the outer layers of the sample can also cause clogging. This clogging makes it more difficult for water to diffuse to the center of the sample, which increases the time required for saturation.** Comparing the evolution of these two parameters between model materials and RCA (Fig. 6 and Fig. 7), we observe a different behavior as carbonation progresses. The evolutions are well observed in the model materials while for RCA, small variations are recorded even over a longer carbonation period. In fact, the low rate of CO₂ stored **after** carbonation by RCA (Fig. 4) explains the small changes observed.

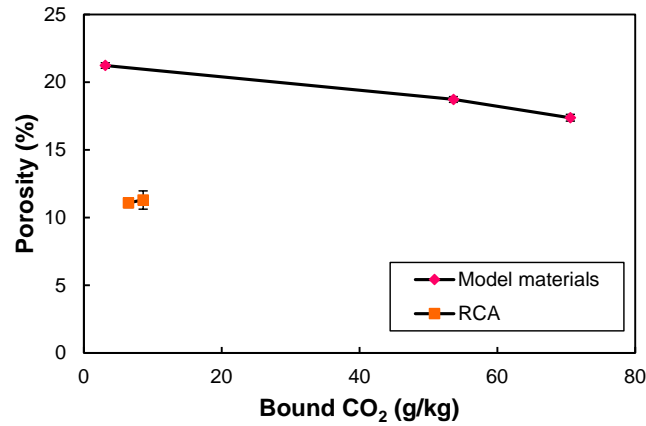


Fig. 6. Evolution of the porosity of RCA and model materials as a function of the amount of carbon dioxide fixed.

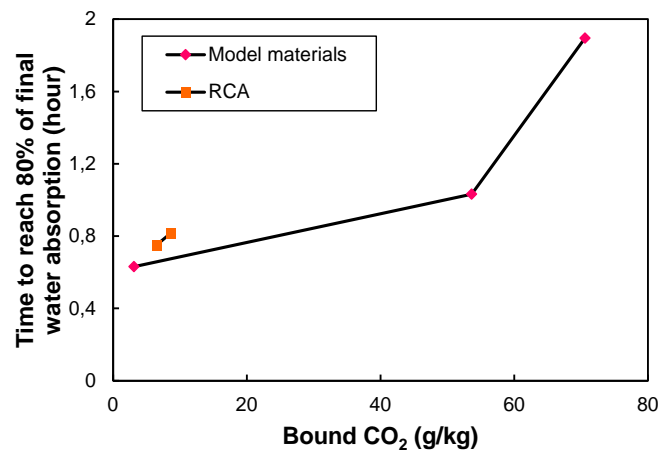


Fig. 7 Evolution of the time required to reach 80% of the final water uptake of RCA and model materials as a function of the amount of carbon dioxide fixed

4 Conclusion

RCA are heterogeneous materials; they can present very large disparities in paste content for the same granular class, and thus generate a dispersion in the test results. Their initial natural carbonation limits the storage of CO₂ during their treatment by accelerated carbonation and consequently limit their evolution. This is why model materials have been manufactured. Spherical samples make possible to monitor the evolution of the microstructural properties upon carbonation.

By storing CO₂ during accelerated carbonation, in the form of calcium carbonates, the porosity and water absorption coefficient are reduced. However, it should be noted that the kinetics of absorption are also modified, with a longer absorption time, which may have consequences in terms of recommendations for the pre-wetting of carbonated **RCA**.

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