Investigation of Carbonation of Concrete Based on Crushed Sand and Admixtures

Abstract

Carbonation is a natural aging process that occurs in all types of concrete. One of its primary implications is the acceleration of steel corrosion caused by the phenomena of depassivation. The goal of this research is to investigate the carbonation of quarry sand-based concrete. The concrete is made of 100% crushed sand 0/6.3, gravel 8/15, and 15/25 from the Arab Contractor quarry in Nomayos, Cameroon, with CEM II B-P 42.5 R from CI-MENCAM (Cimenteries du Cameroun). The study employed two admixtures: one with a dual superplasticizing and reducing action (Sikamen) and another with a water-repellent effect (Sika liquid). Carbonation was performed on concrete samples at the following dates: 0, 7, 14, 28, 56, 90, 180 days, one year, and six months. Carbonated concrete (CC) and non-carbonated concrete (NCC) samples are compared in terms of their physical attributes and mineralogical characteristics. The results of this investigation reveal that after more than a year and six months of carbonation, porosity decreases and permeability increases. Despite the high fineness modulus of quarry sand, the compressive strength of quarry sand-based concrete is satisfactory. Carbonation depth is relatively high on some dates, exceeding the minimal cover value for concrete reinforcement. Sikament additive increases concrete compactness and durability while decreasing permeability. Sika water repellant mixes with the lime in cement to generate complimentary crystallizations that block the mortar's capillaries, making it watertight.

Keywords

Carbonation, Concrete, Crushed Sand, Sikamant, Water-Repellent

1. Introduction

Since the mid-nineteenth century, the economy and industry have grown exponentially. This has led to increased pollution and greenhouse gas (GHG) emissions, and the scarcity of raw materials and natural resources will soon become more evident [1]. This is why one of the main global challenges today is to preserve the planet, which includes the sustainable use of natural resources and raw materials, the reuse of waste for new production processes, and the mitigation of climate change. Carbonation is a natural physical and chemical aging process in concrete that evolves over time and depends on internal and external factors [2]-[4]. Carbonation impacts the durability of concrete, involving a reaction of carbon dioxide (CO₂) with cement hydration products to reduce the pH of the concrete pore solution from around 12 to less than 9 [5]. This is why the use of crushed sand from quarries in the production of cementitious materials is an alternative to reduce overexploitation or to replace river sand [6], a material widely used in the construction of public works infrastructures. However, the use of crushed sand can have consequences for the mechanical behavior and durability of concrete [7]. Indeed, these sands have various characteristics compared with alluvial sands, generally containing finer particle sizes and having a morphology suitable for reducing concrete porosity. These properties ensure granular continuity between cement and gravel for better concrete cohesion [8]. In Cameroon, the use of aggregates in quarries has increased considerably in recent years, with the production of crushed sands. These sands are used in the manufacture of mortars and concretes, yet scientific recommendations on their use have remained scarce. This explains the renewed interest in the influence of crushed sand on mortar properties. Several authors have worked with crushed sand: B. Menadi et al. proved that up to 15% fines could be used [9], O. A. Cabreara et al. found a correlation between voids content and paste volume [10], and A. Al-Ameeri et al. highlighted the aspect of the mineralogical source of sand, which influences concrete behavior [11]. B. Benabed et al. showed that crushed sand from granite produced the best properties in terms of compressive strength and workability. This behavior was attributed to the significant morphology of granite sand particles [12]. As a result, various works have emerged Benachour et al. [13], M.L.K Khouadia et al. [6], Lawrence, [14], Michel, [15] etc. concerning the use of quarry sands. The analysis of published works has led to the conclusion that several parameters may come into consideration, such as the effect of the mineralogical source of the crushing sand, the texture, as well as the shape of the particles. These studies showed that crushed sand is a practical solution for limiting the exploitation of river sand, as long as the mechanical behavior and durability of concretes are not adversely

affected [16]. Other researchers have focused on the effects of adjuvants such as SIKA liquid water repellent and/or SIKAMENT superplasticizer on quarry sandbased concrete. Khalil Bouchouk et al.'s study on quarry sand-based concrete found that SIKA's liquid water-repellent admixture makes the concrete highly plastic, improving its workability and increasing its mechanical strength [17]. Nordmeyer measured water absorption by various mortars containing a waterrepellent agent and observed a reduction in water absorption [18]. Edwige Nicolas shows the effects of superplasticizers on the rheological properties of cement pastes [19]. As carbonation is a natural phenomenon that takes place in the presence of CO₂ (the main greenhouse gas and one of the main causes of global warming [20] [21]), in this study we will resort to accelerated carbonation. This requires the use of special hardening conditions with a high concentration of CO₂ [22]-[24]. In cement-based materials, CO₂ hardening makes the microstructure denser, reduces the necessary curing time, decreases permeability and improves air quality and mechanical properties of water absorption and porosity. [25] [26]. Many researchers have investigated the influence of parameters such as the chemical composition and physical characteristics of concrete, climatic conditions, curing time and calcium carbonate formation on the carbonation rate of concrete. We have set a different objective, which is to quantify the influence of carbonation on the microstructural characteristics of quarry sand-based concrete by carrying out a comparative study between carbonated concrete (CC) and non-carbonated concrete (NCC). The final objective is to provide quantitative elements for better control of the service life of concrete structures using quarry sand as an alternative to river sand.

2. Materials and methods

2.1. Materials

2.1.1. Cement

The cement used in this study is Portland NC 234: 2017 CEM II 42.5 R. The results of the physical characteristics are presented in **Table 1. Tables 2-4** show the chemical, and mineralogical composition of cement and paste

Table 1. Physical characteristics.

| Status | | On Powe | ler | | On paste | | | | |
|--------|-----------------|------------|-----------|-----------|-----------|--------|---------|------|--|
| Natura | Refusal to 0.08 | SSB | DA | PS | Expansion | ı (mm) | Setting | (mn) | |
| Nature | mm (%) | (cm^2/g) | (T/m^3) | (T/m^3) | Cold | Hot | Start | End | |
| CEM II | 1.01 | 3396 | 0.86 | 3.08 | 0 | 0 | 2h58 | 3h46 | |

⁻ SSB: Bladine Specific Surface; - DA: Apparent Density, - PS: Specific Weight.

Table 2. Chimical Composition of cement (manufacturer's data).

| Elements | SiO ₂ | Al ₂ O ₃ | Fe ₂ O ₃ | MnO | MgO | CaO | Na ₂ O | K ₂ O | TiO₂ | P ₂ O ₅ | LOI |
|----------|------------------|--------------------------------|--------------------------------|------|-----|-----|-------------------|------------------|------|-------------------------------|------|
| (%) | 25.5 | 7 | 6 | 0.13 | 5 | 52 | 5 | 0.91 | 0.97 | 0.26 | 2.67 |

LOI: Loss On Ignition at 1000°C.

Table 3. Mineralogical composition of cement (manufacturer's data).

| Constituents | C ₃ S | C_2S | C ₃ A | C ₄ AF | CaSO ₄ | Natural pozzolan |
|-----------------|------------------|---------|------------------|-------------------|-------------------|------------------|
| Proportions (%) | 65 - 79 | 65 - 79 | 65 - 79 | 65 - 79 | 2.5 - 3.5 | 21 - 35 |

⁻ C₃S: Tricalcique Silicate; - C2S: Dicalcium Silicate; - C3A: Tricalcique Aluminate; - CaSO₄: Calcium Sulfate.

Table 4. Paste composition.

| Paste composition | Cement | Normal sand | Water | Adjuvent | Start-up |
|-------------------|--------|-------------|-------|----------|----------|
| Proportions (%) | 500 g | 0 g | 162 g | 0.0 % | 8: 12 |

2.1.2. Sand

The sand used comes from the Arab Contractor quarry in Nomayos, Cameroon, and will be referred to as SA. The sieve analysis results done at LABOGENIE (Laboratoire National de Génie Civil) of sand are summarized in **Tables 5-7**.

Table 5. Sand particle size analysis.

| Nature | Nature Particle size analysis (% passing sieve) | | | | | | | | | | |
|------------------|---|------|-------|------|------|------|------|-----|-----|--|--|
| Screen | 0.08 | 0.16 | 0.315 | 0.63 | 1.25 | 2.5 | 5 | 6.3 | 8 | | |
| Sand (% passing) | 8.0 | 12.9 | 20.2 | 27.1 | 36.5 | 52.9 | 85.5 | 99 | 100 | | |

Table 6. Physical properties of sand.

| Sand | TF | MF | $DA(T/m^3)$ | PS(T/m³) | ES (%) Visual | ES (%) Piston |
|------|-----|------|-------------|----------|---------------|---------------|
| SA | 6.3 | 3.65 | 1.68 | 2.98 | 76 | 74 |

⁻ TF: Fine Content; - MF: Fineness Module, - ES: Sand Equivalent NF EN 933-8 (1999) [27].

Table 7. Chemical composition of crushed sand (%) from X Ray Diffraction (XRD) at Argile, Géologique et Environnement Sédimentaires Laboratory, University of Liège, Belgium (AGES).

| Elements | SiO ₂ | Al ₂ O ₃ | Fe ₂ O ₃ | MnO | MgO | CaO | Na ₂ O | K₂O | TiO ₂ | P ₂ O ₅ | LOI |
|----------|------------------|--------------------------------|--------------------------------|------|------|------|-------------------|------|------------------|-------------------------------|------|
| SA | 65.5 | 15.69 | 7.76 | 0.16 | 2.74 | 1.37 | 1.76 | 2.68 | 0.95 | 0.14 | 0.57 |

2.1.3. Aggregates

Two gravel fractions were used, 8/16 and 15/25 mm, all from the Arab Contractor quarry in Nomayos. They are of gneissic origin and the results of identification tests done at LABOGENIE are summarized in Table 8 and Table 9:

Table 8. Gravel particle size analysis.

| Nature | Particle size analysis (% passing sieve) | | | | | | | | | |
|--------------|--|-----|-----|------|------|------|------|----|------|--|
| Screen (mm) | 5 | 6.3 | 8 | 10 | 12.5 | 16 | 20 | 25 | 31.5 | |
| Gravel 8/16 | 2.6 | 3.2 | 6.5 | 16.7 | 42.7 | 86.3 | 100 | | | |
| Gravel 15/25 | | | 3.5 | 4.8 | 6.6 | 15.9 | 50.8 | 92 | 100 | |

Table 9. Physical properties of gravel.

| Nature | LA (%) | MDE (%) | DA (T/m³) | PS (T/m³) | AP (%) | PR (%) |
|--------------|--------|---------|-----------|-----------|-----------|------------|
| Gravel 8/16 | 1 | 1 | 1.46 | 2.94 | 18 < 30% | 0.99 < 3% |
| Gravel 15/25 | 10/14 | 10/14 | 1.45 | 2.02 | 14 + 200/ | 0.02 + 20/ |
| | 47.7 | 20.04 | 1.45 | 2.92 | 14 < 30% | 0.82 < 3% |

⁻ LA: Los Angeles; - MDE: Micro Deval; - AP: Flattening Coefficient.

The results of the granulometric analysis of the aggregates are shown on the curve in **Figure 1**.

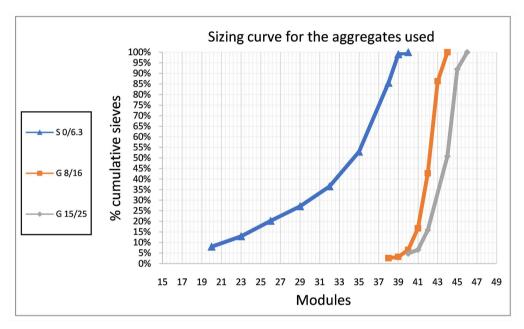


Figure 1. Sizing curve for the aggregates used.

2.1.4. Admixtures

We used two admixtures: Sikament and water repellent. They are poured one after the other into a container, stirred and then poured back into the mixer in the following proportions: Sikament 1.4 liters and Hydrofuge 2.8 liters. The Standard Specification for Chemical Admixtures for Concrete used is ASTM C 494 type G [28] and NF EN 934-2 [29].

2.2. Methodology

The 16×32 cm test specimens were made at LABOGENIE and prepared for the various tests at the Laboratoire de mécanique et matériaux de Génie civil (L2MGC), CY Cergy Paris University.

The design method is Dreux-Gorisse method.

The concrete composition proposed for 1 m^3 Water/Concrete (W/C) ratio = 0.471 was mixed experimentally. The proportions of the constituents are shown in **Table 10**.

Table 10. Concrete formulation with quarry crushed sand.

| Concrete constituents C30/37 | Dosage for 1 m ³ | Dosage for a 50 kg bag of cement |
|------------------------------|-----------------------------|----------------------------------|
| Crushed sand 0/6.3 | 889.0 kg | 111.3 |
| Gravel 8/16 | 398.6 kg | 49.82 |
| Gravel 15/25 | 712.7 kg | 89.09 |
| Cement CEM II B-P 42.5R | 400 kg | 50 |
| Water | 188.4 liters | 23.55 |
| Adjuvent Sikament | 1.4 liter | 0.35 |
| Adjuvent water repellent | 2.8 liters | 0.98 |
| Slump test (cm) | 7.5 cm | |
| Theorical density (kg/m³) | 2589 | |

2.2.1. Carbonation

The samples were subjected to an accelerated carbonation test over 1 year and 6 months under the following conditions: Carbon dioxide (CO₂) content of 3%; relative humidity (RH) of 65% and controlled temperature of 20°C. The whole setup is placed in a laboratory room where the temperature is maintained at T = 20°C \pm 1°C. The samples were preheated for 14 days in an oven at 45°C and 50% RH (to reduce the presence of free water, which could influence carbonation).

Measurements were taken at 3 d, 7 d, 14 d, 28 d, 56 d, 90 d and 180 d, 1 year and 1 year 6 months. The samples were removed from the chamber, and after splitting a specimen for each formulation, the carbonation depth was measured on the fresh fractures using a phenolphthalein color indicator according to the AFPC-AFREM (French Association For Construction - French Association For Research and Testing on Materials and Construction) procedure [30] [31]. Carbonate thickness is characterized by the change in color of the indicator, which turns dark pink in the non-carbonate zone and colorless in the carbonate zone. A control specimen had previously been broken and soaked in phenolphthalein, then placed in an air-conditioned room at 20°C and 23% RH to compare the evolution of carbonation depth with the specimens exposed in the chamber.

The test was carried out on $150 \times 100 \text{ mm}^2$ mortar specimens in accordance with the standard [NF XP 18 458, 2022] [32]. The images in **Figure 2** show the specimens and the device used.

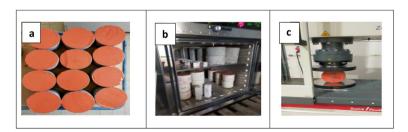


Figure 2. Shows: a) preparation of specimens filmed on both cross-sectional faces to impose CO₂ diffusion in one direction only: b) placing of specimens in the carbonation chamber: c) breaking of specimens by splitting to spray phenolphthalein.

2.2.2. Water-Accessible Porosity and Density

The water-accessible porosity test was carried out on specimen pieces before and after carbonation. The test was carried out only after 28 days of hardening, in accordance with standard [NF P18-459, 2022] [33]. Specimens are evacuated to a pressure of less than 25 mbar using a vane pump and vacuum bell jar. This pressure is maintained for 3 hours. Water impregnation is then carried out under vacuum, via a water line connected to the vacuum chamber. The water is degassed to ensure that the vacuum is maintained when the vacuum chamber containing the samples is connected to a vial containing water. Degassing also evacuates air bubbles suspended in the water. Pressure is maintained for over 44 hours. The porosity and density of the materials are then calculated as follows in Equation (1):

$$\theta = 100 \times \frac{M_{Sat \ air} - M_{Sec}}{M_{Sat \ air} - M_{eau}}, \quad \rho = \rho_{eau} \times \frac{M_{Sec}}{M_{Sat \ air} - M_{Imm}}. \tag{1}$$

where $M_{Sat \ aii}$. The mass (g) of the sample saturated in air, M_{Sec} : The mass (g) of the sample dried at the desired temperature, M_{Imm} : The mass (g) of the sample saturated under water.

2.2.3. Gas Permeability

Permeability was measured on three 150×50 mm slices from 1500×300 mm specimens cut using a water saw. The test was carried out with a constant-load permeameter (type CEMBUREAU from L2MGC of CY Cergy Paris Université). The gas used for measurement is oxygen. Specimens for permeability measurements were preconditioned as follows: dried in a ventilated oven at $T = 80^{\circ}$ C for 7 days, followed by drying in a ventilated oven at $T = 105^{\circ}$ C until the mass (weighed to the nearest 0.01 g) had stabilized to within 0.05%. The test is carried out on carbonated and non-carbonated concrete samples. Apparent permeability is then calculated using Equation (2) combining Darcy's law and the Hagen-Poiseuille equation, assuming laminar flow, compressible fluid and steady state.

$$K_{gaz} = \frac{2 \cdot P_0 \cdot Q \cdot L\mu}{A \left(P_{adm}^2 - P_0^2 \right)}.$$
 (2)

With Q: volume flow rate of fluid (CO₂) in (m^3/s), L: sample thickness (m), A: cross-sectional area of the sample (m^2), μ : dynamic viscosity of the fluid at 20°C (Pa·S), P_{adm} : permissible pressure (Pa), P: inlet pressure (Pa), $P_{0:}$ atmospheric pressure (Pa)

Having different gas permeabilities for different inlet pressures, the intrinsic permeability of the concrete is then calculated using the Klinkenberg method [34] in Equation (3);

$$K = K_{\text{int}} \left(1 + \frac{\beta}{P_m} \right), \quad P_m = \frac{P_0 + P_{adm}}{2}. \tag{3}$$

where P_0 : Atmospheric pressure (Pa), β : Klinkenberg constant, K_{int} : Slope of the Klinkenberg line, P_m : Mean pressure (Pa).

2.2.4. Scanning Electron Microscope (SEM) Observation

The SEM study was carried out on non-carbonated and carbonated samples using SEM of L2MGC equipped with a JEOL JXA-8500F electron probe microanalyzer. Samples ranging in size from 1.5 to 2.0 cm were taken from the core of the mortar specimen. The dried samples were impregnated with epoxy resin and dried for 24 hours. They were then polished using a semi-automatic polishing machine following the method indicated by K. Scrivener, 2004 [35].

3. Results and discussion

3.1. Carbonation

The images in **Figure 3** show the evolution of carbonation depths at the different dates of the study. In these different images of fresh concrete fractures, the colorless areas are carbonated and those colored pink are non-carbonated. The measurement of the discolored zone where the hydrogen potential (pH) drops from 13 to around 9 (with phenolphthalein) or around 10 (with thymolphthalein). More precisely, phenolphthalein is colorless for a pH below 8.2 and deep pink for a pH above 9.9, and thymolphthalein is colorless for a pH below 9.3 and blue for a pH above 10.5.

Figure 4 shows the evolution of carbonation depth as a function of the square root of time.

- It can be seen that the longer the test period, the greater the depth of carbonation.
- There is a mixed zone, i.e. where samples show both colorations together, implying that there is a pH turning zone, which is in line with some of the authors' research [36]-[38]. The mixed zone suggests the gradual disappearance of portlandite.
- The images in **Figure 3** show: The carbonate zone, in which portlandite has completely disappeared; The mixed zone, in which the amount of calcium carbonate decreases and the amount of portlandite increases; the healthy zone, in which no mineralogical changes are observed.

Carbonation of concrete is a natural aging process. In the presence of water, it is responsible for the depassivation and corrosion of steel reinforcement in reinforced concrete, and the bursting of the concrete coating protecting the steel reinforcement. The reinforcement cover is the distance between the concrete surface and the nearest reinforcement. It must be sufficient to guarantee: good protection of the steel against corrosion; good transmission of bond forces; adequate fire resistance. NBN EN 1992-1-1 [39] prescribes minimum cover values for reinforced concrete structures according to structural class, exposure class and environmental class. It accepts that the structural class to be used for common buildings and civil engineering structures is S4, corresponding to a service life of 50 years. With a carbonation depth of 3.6 cm; for a respective duration of 1 year and 6 months of carbonation in our case. We have on reinforced concrete an excess of 1.1 cm, 0.6 cm respectively for exposure classes XC2/XC3 and XC4. This is due to the

accelerated carbonation and the test conditions, the cumulative role of the aggregates and above all the two admixtures used. These results reflect the influence of several factors on carbonation rate.

| Exposure time | Image of the carbonation front | Carbonation depth |
|----------------------|---|-------------------|
| 0 day (indicator) | 33 34 35 36 37 38 30 40 41 42 43 44 45 46 47 4 | 0 mm |
| 7 days | | 3 mm |
| 14 days | | 8 mm |
| 28 days | | 11 mm |
| 56 days | | 20 mm |
| 90 days | | 22 mm |
| 180 days | | 25 mm |
| 1 year | 2003/00-444 500 (Serri7 et 6 9 10 11 12 13 14 15 16 | 31 mm |
| 1 year 6 months | 200 300 A 1 500 Quary (m. 6) 0 10 11 12 13 14 16 1 | 36 mm |

Figure 3. Changes in carbonation depths at different study dates.

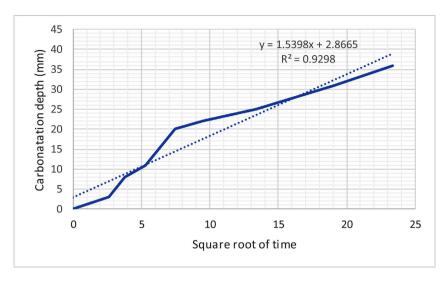


Figure 4. Carbonation depth as a function of the square root of time.

Interpretation of Factors Influencing Carbonation Rate

With a low W/C ratio = 0.471 like ours, the cement grains in a unit volume are numerous and close together, and the spaces between the cement grains are smaller. This reduction not only decreases the total volume of capillary pores, but also reduces their diameter. So, for a low W/C ratio, capillary porosity is made up of a finer, more discontinuous pore network. Under these conditions, it is not easy for CO_2 to penetrate the cement paste. This is in line with the results of [40]. As a result, the W/C ratio has a major influence on the rate of carbonation.

From Figure 3, we obtain the following Table 11 on carbonation rate.

Table 11. Carbonation rate.

| Thickness (mm) | 0 | 3 | 8 | 11 | 20 | 22 | 25 | 31 | 36 |
|------------------------------|---|-------|-------|-------|-------|-------|-------|-------|-------|
| Time (days) | 0 | 7 | 14 | 28 | 56 | 90 | 180 | 365 | 547.5 |
| Speed (m/s) 10 ⁻⁹ | 0 | 4.960 | 6.613 | 4.546 | 4.133 | 2.829 | 1.607 | 9.830 | 7.610 |

From **Table 11**, we obtain **Figure 5**, which shows the carbonation rate as a function of carbonation exposure time. The results are varied: from the 7th to the 14th day, the rate increases, but from the 14th to the 180th day of carbonation, the rate decreases. It decreases from day 180 to day 365 of carbonation, and then decreases again from day 365 to day 5475.5. As our dosage is 400 kg/m³, our results show that the carbonation rate is either slowed or accelerated, as shown in **Table 11** and **Figure 3** above.

However, according to [41], this parameter is less influential than the W/C ratio. The rate of progression of the carbonation front depends on the characteristics of the material (porosity, nature of the cement, etc.) and the relative humidity (RH) of the surrounding environment, which determines the water content of the concrete [42]. In our case, the RH is 65%, and the results interpreted below on the speed of the carbonation front are varied. In our study, we used two admixtures,

Sikament and Sika liquid water repellent. Sikament with its double facet acts as a superplasticizer or high-water reducer. As a superplasticizer, it gives concrete very high plasticity and prolonged rheology. As a water reducer, it reduces the water content of concrete by 25%, leading to a significant gain in final strength, increased compactness and durability, and reduced permeability. Sika Liquid water-repellent combines with the lime in the cement to form complementary crystallizations that obstruct the mortar's capillaries, making it watertight. The combined effect of the two admixtures has a major influence on reducing the rate of carbonation in concrete, and improves other concrete properties.

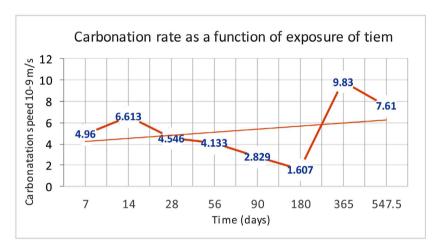


Figure 5. Carbonation rate as a function of exposure time.

3.2. Porosity

a) Effect of carbonation on porosity

Figure 6 shows the results of the water-accessible porosity test on carbonated and non-carbonated concrete samples.

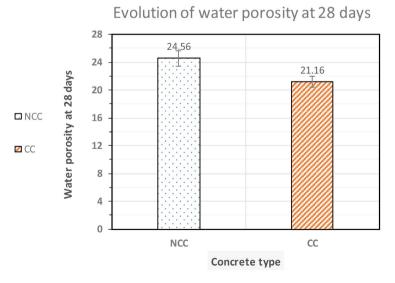


Figure 6. Evolution of porosity before and after carbonation.

Concrete and mortar are porous materials. In their hardened state, they contain voids, usually filled with air or water. The volume of these pores is highly variable, and can reach 10% to 20% or even more, *i.e.* 100 to 200 l per m³ of concrete. Porosity is therefore an important factor in material quality. In particular, it determines strength, water content, compactness and durability. A distinction is made between compaction pores due to concrete placement, aggregate pores and cement paste pores.

It can be seen that:

- Pore volume decreases with carbonation
- The decrease is estimated at 3.6% between non-carbonated and carbonated concrete samples.
- This decrease can be explained by the difference between the molar volumes of portlandite (33 cm³·mol⁻¹) and calcite (35 cm³·mol⁻¹). In this case, the reaction involving the transformation of one mole of Ca(OH)₂ into one mole of CaCO₃ by the action of CO₂, induces an increase in the volume of the solid phase, and therefore a reduction in the pore space, of 2 cm³·mol⁻¹.

It is generally accepted that the total reduction in porosity is mainly due to the carbonation of portlandite, leading to an increase in solid phase volume. However, the variation in molar volume linked to C-S-H carbonation must also be taken into account. According to Thiéry [41], this can be assessed by considering the variation in porosity linked solely to the carbonation of portlandite and C-S-H. In other words, the carbonation of minority hydrated phases (AFt and AFm in particular) is neglected. In this context, it should be borne in mind that the increase in solid phase volume depends on the calcium carbonate polymorph formed. Thus, the fall in porosity is directly related to the proportions of carbonate polymorphs formed. Considering that all the portlandite consumed contributes to the formation of calcium carbonate (calcite in our case).

b) Effect of W/C ratio on porosity

It can be seen that the W/C ratio has a major influence on the porosity of hydrated cement paste, as it directly governs the initial spacing between cement grains suspended in the mixing water (Table 12).

Table 12. Influence of carbonation on water-accessible porosity.

| Concrete | W/C | R _C (7 days) in MPa | Rc (28 days) in MPa | Ø Water (%) |
|----------|-------|--------------------------------|---------------------|-------------|
| NCC | 0.471 | 27 | 36 | 24.56 |
| CC | | | | 21.16 |

Indeed, the lower W/C ratio, the closer the cement grains are initially to each other. The spaces to be filled between cement grains are smaller, and there is less chance of a large void that cannot be completely filled by hydrates. Reducing the W/C ratio not only reduces the total volume of capillary pores, it also reduces their diameter. At lower W/C, capillary porosity is in fact made up of a finer, more discontinuous pore network [42].

3.3. Gas Permeability

Permeability is very sensitive to the drying conditions that precede measurement.

a) Influence of carbonation on gas permeability

If porosity is the main strength parameter of a structure, there's another physical quantity among many others that's important: the material's permeability. It is considered a key indicator of durability. It is highly dependent on the porous network, its connectivity and the water content of the material [43]. Because of its abundant use, concrete is increasingly raising the issue of its durability in anticipation of damage that could occur in the medium or long term.

Figure 7 shows the values for intrinsic concrete permeability, highlighting the correlation with water-accessible porosity. The curve of K as a function of the inverse of the mean pressure is given by the straight line fitting the point clouds obtained (Klinkenberg straight line). The intersection of this line with the ordinate axis gives the intrinsic permeability Kint. The intersection points were used to obtain the results.

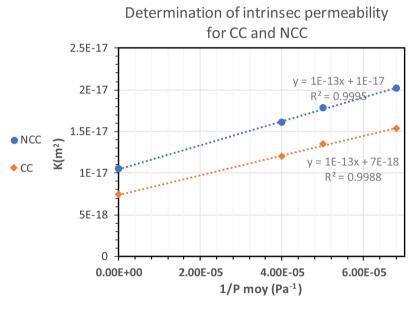


Figure 7. Determination of intrinsic permeability for CC and NCC.

It can be seen that the direction of variation of intrinsic permeability before and after carbonation is different for carbonated and non-carbonated concrete. K_{int} increases by 3.1338 from non-carbonated to carbonated concrete. Permeability increases, implying that porosity is greater, which is contradictory to the results found for porosity. However, high porosity does not necessarily mean high permeability, but it is the size of the pores making up this porosity, and in which the flow takes place, that defines this permeability [44].

b) Effect of W/C ratio on gas permeability

Some authors have noted that gas permeability is particularly sensitive to the value of W/C [45]. Carbonation can create new pores after the dissolution of

portlandite crystals, a phenomenon all the more remarkable when concrete is initially porous. According to T. Chaussadent [46], the $Ca(OH)_2$ crystals have become larger, so after their massive dissolution, new pores will be created, providing preferential paths for the passage of gas. This may explain the increase in permeability after carbonation.

4. Conclusions

The results obtained show that quarry sand concrete has very good physical and mechanical properties despite its high fineness, and in particular good compressive strength. In comparison with the work of Abdias *et al.* [47], this sand can be used as a substitute for river sand. What's more, if we consider the mix of aggregate proportions, the role played by the ratios also gives an additional satisfaction according to the work of P. Bame Che *et al.* [48].

- Carbonation of quarry sand-based concrete results in considerable changes in physical and mineralogical properties over time.
- Concrete's microstructure becomes more resistive to hostile chemicals, despite decreased porosity and increased permeability.
- Reducing water and pore space leads to mass loss and increased elastic deformation. This suggests that the concrete undergoes extensive chemical and mechanical alterations during carbonation.
- Substituting river sand for quarry sand in concrete yields nearly identical physical and microstructural qualities. Quarry sand-based concrete has high fineness modulus but good compressive strength due to inter-granular void filling.
- On some dates, carbonation depth surpasses the minimum cover value for reinforcing. This is due to the accelerated carbonation and test conditions, the
 aggregates' cumulative role, and, most importantly, the two admixtures utilized. The microstructure becomes firmer and more resistant to hostile treatments
- The combined impact of Sikament admixture and Sika water repellent is a result of carbonation, particularly at a deeper level. Sikament additive increases concrete compactness and durability while decreasing permeability. Sika water repellant reacts with the lime in cement to generate comparable crystallizations that block the mortar's capillaries, rendering it watertight.

References

- [1] Xi, C. and Cao, S. (2022) Challenges and Future Development Paths of Low Carbon Building Design: A Review. *Buildings*, **12**, Article 163. https://doi.org/10.3390/buildings12020163
- [2] Chen, C. and Ho, C. (2013) Influence of Cyclic Humidity on Carbonation of Concrete. *Journal of Materials in Civil Engineering*, 25, 1929-1935. https://doi.org/10.1061/(asce)mt.1943-5533.0000750
- [3] Turcry, P., Oksri-Nelfia, L., Younsi, A. and Aït-Mokhtar, A. (2014) Analysis of an Accelerated Carbonation Test with Severe Preconditioning. *Cement and Concrete Research*, **57**, 70-78. https://doi.org/10.1016/j.cemconres.2014.01.003
- [4] Leemann, A. and Moro, F. (2016) Carbonation of Concrete: The Role of CO2 Concentration, Relative Humidity and CO₂ Buffer Capacity. *Materials and Structures*, **50**, Article No. 30. https://doi.org/10.1617/s11527-016-0917-2
- [5] Das, B.B., Rout, S.K., Singh, D.N. and Pandey, S.P. (2012) Some Studies on the Effect of Carbonation on the Engineering Properties of Concrete. **86**, *Indian Concrete Journal*, 7-12.
- [6] Khouadjia, M.L.K., Mezghiche, B. and Drissi, M. (2015) Experimental Evaluation of Workability and Compressive Strength of Concrete with Several Local Sand and Mineral Additions. *Construction and Building Materials*, 98, 194-203. https://doi.org/10.1016/j.conbuildmat.2015.08.081
- [7] Zeghichi, L., Benghazi, Z. and Baali, L. (2014) The Effect of the Kind of Sands and Additions on the Mechanical Behaviour of S.C.C. *Physics Procedia*, 55, 485-492. https://doi.org/10.1016/j.phpro.2014.07.070
- [8] Dang, J. and Zhao, J. (2019) Influence of Waste Clay Bricks as Fine Aggregate on the Mechanical and Microstructural Properties of Concrete. *Construction and Building Materials*, 228, Article 116757. https://doi.org/10.1016/j.conbuildmat.2019.116757
- [9] Menadi, B., Kenai, S., Khatib, J. and Aït-Mokhtar, A. (2009) Strength and Durability of Concrete Incorporating Crushed Limestone Sand. *Construction and Building Materials*, **23**, 625-633. https://doi.org/10.1016/j.conbuildmat.2008.02.005
- [10] Cabrera, O.A., Traversa, L.P. and Ortega, N.F. (2010) Fluidez de morteros cementíceos con arenas machacadas. *Materiales de Construcción*, 60, 115-130. https://doi.org/10.3989/mc.2010.50909
- [11] Al-Ameeri, A. (2012) Using Different Types of Fine Aggregate to Produce High Strength Concrete. *International Journal of Arts & Sciences*, **5**, 187-196.
- [12] Benabed, B., Kadri, E., Azzouz, L. and Kenai, S. (2012) Properties of Self-Compacting Mortar Made with Various Types of Sand. *Cement and Concrete Composites*, **34**, 1167-1173. https://doi.org/10.1016/j.cemconcomp.2012.07.007
- [13] Benachour, Y., Skoczylas, F., Houari, H., et al. (2008) Étude expérimentale des

- mortiers fortement charges en fillers calcaires. Sciences & technologie b, université mentouri Constantine Algérie, **28**, 53-59.
- [14] Lawrence, P. (2000) On the Activity of Fly Ash and Chemically Inert Mineral Additions in Cementitious Materials. PhD Thesis, Paul Sabatier University.
- [15] Michel (2007) Influence of Physic-Chemical Characteristics of Limestone Fillers on Fresh and Hardened Mortar Performances. *Proceedings of the International RILEM Symposium on SCC*, Ghent, 3-5 September 2007, 205-210.
- [16] Emmanuel, E., et al. (2021) Mechanical and Mineralogical Characteristics of Mortars with Crushed and River Sand. Proceedings of the 13th Fib Internationnal PhD Symposium in Civil Engineering, Marne-la-vallée, 6-28 August 2020, 1-9.
- [17] Khalil, B., *et al.* (2016) Evaluation of Secondary Effects of Admixtures on Concrete Properties.
- [18] Nordmeyer, H. (2007) Water-Rebellent Performance in Pozzolanic and Traditional Mortars. 2007 *World of Coal Ash*, Covington, May 7-10 2007, 1-15.
- [19] Edwige, N. (2010) Cementitious Binder/Superplasticizer Compatibility and Incompatibility. PhD Thesis, University of Luxembourg.
- [20] Shi, J., Tan, J., Liu, B., Chen, J., Dai, J. and He, Z. (2021) Experimental Study on Full-Volume Slag Alkali-Activated Mortars: Air-Cooled Blast Furnace Slag versus Machine-Made Sand as Fine Aggregates. *Journal of Hazardous Materials*, 403, Article 123983. https://doi.org/10.1016/j.jhazmat.2020.123983
- [21] Liu, B., Qin, J., Shi, J., Jiang, J., Wu, X. and He, Z. (2021) New Perspectives on Utilization of CO₂ Sequestration Technologies in Cement-Based Materials. *Construction and Building Materials*, 272, Article 121660. https://doi.org/10.1016/j.conbuildmat.2020.121660
- [22] Liang, C., Pan, B., Ma, Z., He, Z. and Duan, Z. (2020) Utilization of CO₂ Curing to Enhance the Properties of Recycled Aggregate and Prepared Concrete: A Review. *Cement and Concrete Composites*, 105, Article 103446. https://doi.org/10.1016/j.cemconcomp.2019.103446
- [23] Suescum-Morales, D., Kalinowska-Wichrowska, K., Fernández, J.M. and Jiménez, J.R. (2021) Accelerated Carbonation of Fresh Cement-Based Products Containing Recycled Masonry Aggregates for CO₂ Sequestration. *Journal of CO₂ Utilization*, 46, Article 101461. https://doi.org/10.1016/j.jcou.2021.101461
- [24] Scrivener, K.L., John, V.M. and Gartner, E.M. (2018) Eco-Efficient Cements: Potential Economically Viable Solutions for a Low-CO₂ Cement-Based Materials Industry. Cement and Concrete Research, 114, 2-26.

 https://doi.org/10.1016/j.cemconres.2018.03.015
- [25] Zhang, D., Ghouleh, Z. and Shao, Y. (2017) Review on Carbonation Curing of Cement-Based Materials. *Journal of CO₂ Utilization*, 21, 119-131. https://doi.org/10.1016/j.jcou.2017.07.003
- [26] Merino-Lechuga, A.M., González-Caro, Á., Fernández-Ledesma, E., Jiménez, J.R., Fernández-Rodríguez, J.M. and Suescum-Morales, D. (2023) Accelerated Carbonation of Vibro-Compacted Porous Concrete for Eco-Friendly Precast Elements. *Materials*, 16, Article 2995. https://doi.org/10.3390/ma16082995
- [27] NF EN 933-8 (1999) Évaluation Des Fines Équivalent de Sable, 4 p.
- [28] ASTM C494 Standard Specification for Chemical Admixtures for Concrete Type G Water-Reducing, High Range, and Retarding Admixtures.
- [29] NF EN 934-2+A1, August 2012, Admixtures for Concrete, Mortar and Grout Part

- 2: Concrete Admixtures Definitions, Requirements, Conformity, Marking and Labelling.
- [30] AFPC-AFREM (1997) Essai de carbonatation accélérée, Mesure de l'épaisseur de béton carbonaté, Mode opératoire recommandé par l'AFREM. Compte rendu des journées techniques AFPC-AFREM Durabilité des bétons, Toulouse, 11-12 Décembre 1997, 153-158.
- [31] Baroghel-Bouny, V., Chaussadent, T., Croquette, G., Divet, L., Gawséwitch, J., Godin, J., Henry, D., Platret, G. and Villain, G. (2002) Caractéristiques microstructurales et propriétés relatives à la durabilité des bétons Méthodes de mesures et d'essais de laboratoire, Méthodes d'essai n58 dans : Techniques et Méthodes des Laboratoires des Ponts et Chaussées.
- [32] NF XP 18-458 (2022) Essai pour béton durci-Essai de carbonatation accélérée-Mesure de l'épaisseur de béton carbonate, 6 p.
- [33] NF P18-459 (2022) Béton-Essai pour béton durci-Essai de porosité et de masse volumique, 7 p.
- [34] Zhang, D. and Li, K. (2019) Concrete Gas Permeability from Different Methods: Correlation Analysis. *Cement and Concrete Composites*, 104, Article 103379. https://doi.org/10.1016/j.cemconcomp.2019.103379
- [35] Scrivener, K.L. (2004) Backscattered Electron Imaging of Cementitious Microstructures: Understanding and Quantification. Cement and Concrete Composites, 26, 935-945. https://doi.org/10.1016/j.cemconcomp.2004.02.029
- [36] Rafaï, N., Hornain, H., Villain, G., Baroghel Bouny, V., Platret, G. and Chaussadent, T. (2002) Comparaison et validité des méthodes de mesure de la carbonatation. *Revue française de génie civil*, **6**, 251-274. https://doi.org/10.3166/rfgc.6.251-274
- [37] Rahman, A.A. and Glasser, F.P. (1989) Comparative Studies of the Carbonation of Hydrated Cements. Advances in Cement Research, 2, 49-54. https://doi.org/10.1680/adcr.1989.2.6.49
- [38] Parrott, L.J. and Killoh, D.C. (1989) Carbonation in a 36 Year Old, In-Situ Concrete. Cement and Concrete Research, 19, 649-656. https://doi.org/10.1016/0008-8846(89)90017-3
- [39] NBN EN 1992-1-1 Eurocode 2 Calcul des structures en béton Partie 1-1: règles générales et règles pour les bâtiments (P18-711-1 :2005-10, NA :2016-03, A1 :2015-02)
- [40] Viet, D.N. (2015) Contribution à l'approche probabiliste de la durabilité des structures en béton soumises à la carbonatation. Thèse de doctorat à l'Institut National des Sciences Appliquées de Toulouse.
- [41] Thiery, M. (2005) Modélisation de la carbonatation atmosphérique des matériaux cimentaires, prise en compte des effets cinétiques et des modifications microstructurales et hydriques. Thèse de doctorat, Central Laboratory Ponts Et Chaussees.
- [42] Gagné, R. (2000) GCI 714—Durabilité et réparations du béton. Université de Sherbrooke Centre de Longueuil.
- [43] Vincent, P. (2001) Influence d'un endommagement mécanique sur la perméabilité et sur la diffusivité hydrique des bétons. Thèse de Doctorat, université de Nantes.
- [44] Abbas, A., Carcasses, M. and Ollivier, J.P. (2000) The Importance of Gas Permeability in Addition to the Compressive Strength of Concrete. *Magazine of Concrete Research*, **52**, 1-6. https://doi.org/10.1680/macr.2000.52.1.1
- [45] Jaafar, W. (2003) Influence de la carbonatation sur la porosité et la perméabilité des bétons, Rapport de stage LCPC.

- [46] Chaussadent, T. (1999) Etat des lieux et réflexions sur la carbonatation du béton armé. Etudes et recherches des LPC, série ouvrages d'Art OA29, Edité par LCPC Paris
- [47] Abdias, M.W.M., Blanche, M.M., Nana, U.J.P., Abanda, H.F., François, N. and Chrispin, P. (2023) River Sand Characterization for Its Use in Concrete: A Revue. *Open Journal of Civil Engineering*, **13**, 353-366. https://doi.org/10.4236/ojce.2023.132027
- [48] Che, P.B., Emmanuel, Y.B. and Billong, N. (2022) Equal Volumes of Sand and Gravel Concrete Mix Ratios in Cameroon and Its Effect on Concrete Compressive Strength. World Journal of Engineering and Technology, 10, 539-549. https://doi.org/10.4236/wjet.2022.103034