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Recycled Plastic and Cork Waste for Structural Lightweight Concrete Production

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Abstract: The use of waste materials as lightweight aggregates in concrete is highly recommended in seismic risk areas and environmentally recommended. However, reaching the strength needed for the concrete to be used structurally may be challenging. In this study four dosages were assayed: the first two-specimen had high cement content (550 and 700 kg/m³ respectively), Nanosilica, fly ash and superplasticizer. These samples were high performance, reaching a strength of 100 MPa at 90 days. The other two mixtures were identical but replaced 48% of the aggregates with recycled lightweight aggregates (30% polypropylene, 18.5% cork). To estimate its strength and durability the mixtures were subjected to several tests. Compression strength, elasticity modulus, mercury intrusion porosimetry, carbonation, attack by chlorides, and penetration of water under pressure were analyzed. The compression strength and density of the lightweight mixtures were reduced 68% and 19% respectively; nonetheless, both retained valid levels for structural use (over 30 MPa at 90 days). Results, such as the total porosity between 9.83% and 17.75% or the chloride ion penetration between 8.6 and 5.9 mm, suggest that the durability of these concretes, including the lightweight ones, is bound to be very high thanks to a very low porosity and high resistance to chemical attacks.

Keywords: recycled plastic aggregate; recycled cork aggregate; nanosilica

1. Introduction

The low density of lightweight concrete (LWC) makes it suitable for the rehabilitation of buildings, especially those located in seismic areas.

Most of the codes define LWC as those having a density between 1200 and 2000 kg/m³. Such density is achieved by replacing natural aggregates by lightweight aggregates (LWA). However, LWCs tend to show lower compressive strength and less elastic modulus than traditional normal concrete (NC) made with natural aggregate. Besides, LWC tend to have lower durability than NC [1–3]. One of the reasons is a weaker and more porous interfacial transition zone (ITZ) [4–6].

Higher cement content can be used in order to improve the mechanical properties and durability of LWC; however, this carries high environmental costs. In fact, over 80% of the CO₂ emissions to the atmosphere in the production of concrete are caused by the production of clinker. The cement industry emits about two billion tons of CO₂, which represents between 6 and 7% of the total world emissions [7].

Eco-concrete can be defined as a concrete that reduces the amount of cement and aggregates by using waste materials, such as fly ash (FA), ash from sludge incineration, admixtures such as nanosilica (Ns), or recycled aggregates.

Ns is a nanocomposite technology. The nano-scale size of the particles can act as nucleation points in some phases of cement hydration due to their high reactivity, as well as nano-reinforcement or nano-filler to densify the microstructure, thus improving the ITZ between paste and aggregates [8]. It is used in concentrations between 0.5 and 10% by weight of cement (bwoc), as at higher concentrations the heat of hydration rises, causing increased autogenous shrinkage. It is usually commercialized as colloidal Ns, that is, a nucleus of amorphous silicon dioxide with hydroxylated surface. It has a high surface area in relation to its volume, which makes it highly chemically reactive. It reacts with the calcium hydroxide crystals ($\text{Ca}(\text{OH})_2$) found in the ITZ, producing hydrated calcium silicate (CSH gel). Due to this reaction, the amount of calcium hydroxide crystals decreases and the strength of the cement paste increases [9]. Ns does not work only at a chemical level, but also at a physical one: it is 100 times smaller than cement, so it can fill the minuscule holes left by the partially hydrated cement particles [10]. This leads to an increase of the density and a reduction of the porosity.

In this work natural aggregate is partially replaced by recycled polypropylene and cork. Plastic has become an indispensable material in our life due to its innumerable uses and possibilities; the counterpart, however, is the amount of waste that it causes. The generation of waste products involves storage or disposal issues, hence the need to reuse them as a recycled material. Polypropylene (PP) is one of the most common plastics. In Spain, it accounts for 22% of the total amount of plastic used according to the Spanish Center of Plastics [11], and yet, despite the amount of literature on recycled plastic aggregates, [1,12–17], very little has been published on PP, since most studies refer to PET or polyurethane foam [18–21]. Although plastic is always a valid aggregate, it is not always suitable for structural concrete. The most usual percentage of substitution of conventional aggregate with plastic ranges between 15% and 50% in volume, while the strength reached ranges between 10 and 50 MPa. The most commonly used plastic is crushed PET bottles used as fine aggregate [13,19].

A number of studies have analyzed the ITZ of plastic aggregates [22–31] and there is a consensus that the link between cement and aggregate is weaker compared to traditional aggregates. This is because plastic being a hydrophobic material, it does not absorb free water in the ITZ. This free water leaves pores and holes on its way out. It also causes a lack of hydration in the areas around the aggregate whereas with traditional aggregates the correct hydration of such zones is improved by the slow release of the water that had been absorbed by the aggregate. The shape of the plastic used as aggregate also affects the ITZ; a smooth and rounded shape has a negative impact on the interface, producing what Ben Fraj, Kismi, and Mounanga [20] call “wall effect”. To avoid such effects, some researchers suggest pre-treatment with sodium hydroxide [25], use of plastic in the form of foam [20,28], coating with blast furnace slag [32], or sand [21].

Cork was also assayed in this research work due to its low density. Cork is a plant tissue that is obtained by extracting cork oak bark (*Quercus suber* L.). Its peculiar structure gives it a low density, between 140 and 200 kg/m^3 , and a compressive strength over 90 MPa [33]. The cork industry occupies a forest area of 22,000 km^2 and produces 340,000 tons per year [34], 68,000–85,000 tons of which are estimated to be discarded [35]. Studies on cork as an LWA obtained very low densities, between 1000 and 2000 kg/m^3 , with percentages of substitution between 2 and 18%; however, the strength was also low, below 20 MPa [36–38]. Its capacity for absorption of energy, impact strength and behavior as thermal and acoustic insulation are also to be pointed out.

Nevertheless, the effects of replacing cement by supplementary cementing materials (SCM) and replacing natural aggregates by recycled ones are difficult to predict, thus there are conflicting views on this issue. This is the reason why in the current study two different dosages of LWC were assayed, replacing a certain percentage in volume of the traditional aggregate. The aim of this work was to assay and study LWC with recycled LWA capable of replacing NC in building structures, thus no special means were used for mixing or curing.

The durability behavior was studied together with density and compressive strength to determine whether or not the mix meets the requirements for a structural LWC. To analyze the durability,

mercury intrusion porosimetry, sorptivity, penetration of water under pressure, depth of carbonation, and accelerated chloride ion penetration were studied.

2. Materials and Methods

2.1. Materials

Two mixtures of LWC were made, both replacing 48% in volume of the traditional aggregate. Of these, 30% was replaced by recycled PP plastic and 18.5% cork. To improve strength and durability, 2% of the weight cement was Ns, while the amount of cement was varied (550 and 700 kg/m³ respectively), to increase the strength. Another set of two equivalent dosages without LWA, but equal content of cement, FA, water and Ns were made in order to analyze the behaviors that are consequence of recycled LWA use. The water/binder ratio was 0.25. Both cement and FA were supplied by Cementos La Cruz S.L.

Figure 1 shows their particle size—very small and even—ranging in both cases between 30 and 20 µm, which increases their reactivity. The traditional aggregates used were unwashed crushed limestone fine aggregate, d/D 0/4 particle size group (Designation of an aggregate by lower (d) and upper (D) sieve size, expressed as d/D), with 12% of mass percent passing the sieve 0.0063 mm. and density of the particles of ≥ 2.57 Mg/m³. The gravel was, unwashed crushed limestone coarse aggregate, 6/12 particle size group, 0.7% of mass percent passing the sieve 0.0063 mm and density of the particles of ≥ 2.60 Mg/m³, both proceeding from the region of Murcia (Spain). The recycled LWA were a compound based on recycled PP, reinforced with 40% Calcium Carbonate, particle size of 1/4; density: 1.20 g/cm³, and, due to its manufacturing method, (injection molding) its shape is regular and round-cylindrical. Reused natural cork (byproduct of the agricultural sector) with granulometry 0.25/8. (Figure 1).

The superplasticizer (SP) was based on polycarboxylates. The Ns was a liquid suspension of amorphous Ns.

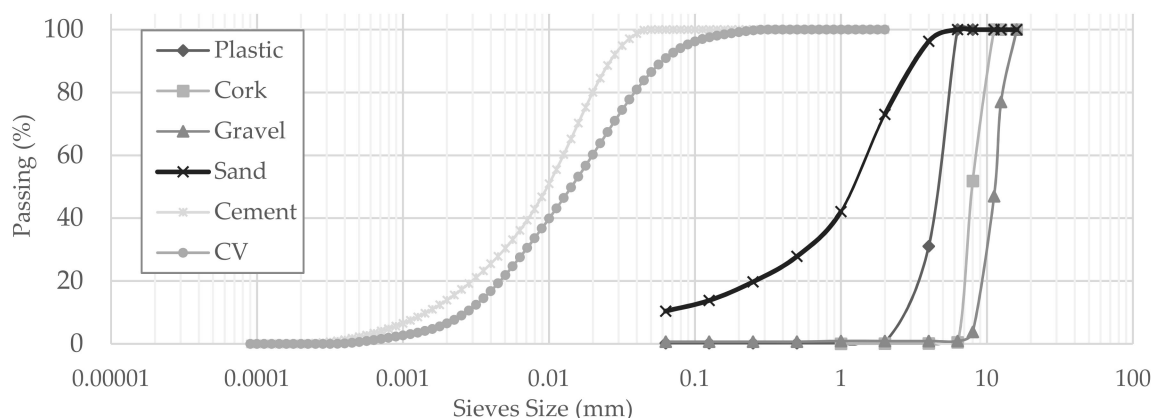


Figure 1. Grading of cement, fly ash (FA), and all types of aggregates.

2.2. Concrete Mixture, Mixing, and Curing Procedure

The dosages used are detailed in Table 1. The concrete was produced in an electric, 50-l, forced action pan mixer machine. To avoid segregation of LWC, the aggregates were poured and mixed first, then cement and FA were added and re-mixed; then half the amount of water was added to the mixture and mixed for 5 minutes. Finally, the remainder of the water was added together with the Ns and SP and mixed for another 5 minutes. Four batches were made of each mixture and samples were taken from different batches for each test. The making and curing were carried out according to standard EN 12390-2: 2009. The concrete slump test was performed to check workability. Medium workability was achieved with slump 45 mm. Compaction was carried out by means of an internal (poker) vibrator. The vibrator was inserted twice in each test tube—at mid filling and at the end, for ten

seconds each time. The test pieces did not show any irregularities, voids or segregation. They were kept in the molds for 24 hours and then cured in water until the different tests were carried out.

Table 1. Concrete mix design in kg/m³.

Concrete ID	HPC-550	HPC-700	ECOHP-700	ECOHP-550
Cement	550	700	550	700
Water	160	180	160	180
FA	130	70	150	70
Ns	14	14	14	14
SP	11	11.75	11	11.75
Sand	1015.16 (66.7%) ¹	927.19 (66.7%) ¹	646.94 (42.7%) ¹	593.39 (42.7%) ¹
Gravel	507.58 (33.3%) ¹	463.6 (33.3%) ¹	142.27 (9.3%) ¹	129.82 (9.3%) ¹
Plastic	-	-	206.16 (29.5%) ¹	189.49 (29.5%) ¹
Cork	-	-	21.5 (18.5%) ¹	19.77 (18.5%) ¹

¹ The figures in parentheses are the percentages of 100% of the aggregates used in volume.

2.3. Test Methods

- Before the destructive tests, **density** was measured by weighing and measuring all the test pieces, by means of a standard scale and a tape measure and working out the average.
- The **compressive strength tests** were performed according to EN 12390-3 2009. Cylindrical specimens of 150 mm diameter and 300 mm height were used. After the curing period (7, 28, and 90 days) the specimens were removed from the immersion container and polished, then weighed and introduced into an hydraulic press.
- The **elasticity modulus** was obtained following the ASTM C 469 standard "Standard Test Method for Static Modulus of Elasticity and Poisson's Ratio of Concrete in Compression". A cylindrical concrete specimen is arranged vertically between uniaxial compression load plates, using two circular steel punches arranged concentrically on top of and underneath the sample to be tested. When performing the test, the applied load produces an increase of the traction on the planes that contain the cylinder axis, thus the sample fractures along these planes. Usually three radial fissures may be observed in the specimen tested, even though 2 or 4 fissures may occur. Under the breaking load, two conical wedges form under the steel punches. The deformation is measured by a wire strain gauge.
- The **mercury intrusion porosimetry (MIP) test** was performed according to the procedure described by ISO 15901-1 using Quantachrome Poremaster 60 GT, with an approximate pressure of 60.000 psia (414 MPa). Surface tension of mercury; $\gamma = 0.48 \text{ Nm}^{-1}$ and mercury contact angle; $\theta = 140^\circ$. At this pressure, the smallest pore size at which the mercury can be introduced is 3 nm. This test was carried out 60 days after the production of the concrete. After drying 12 hours at 60 °C, the samples were weighed on a precision scale, placed in the porosimetry equipment and mercury was introduced at low pressure, then the samples were weighed again. Finally, the process was repeated, but at high pressure.
- **Sorptivity or absorptivity** measures the ability of a medium to absorb water by capillarity. The Faguerlund method (UNE 83982:2008) was used. The samples were dried in an oven at 60 °C for 48 hours and placed in hermetic bags until the temperature stabilized. Then, the bag was removed and the sample placed in a rectangular plastic container filled with water, sitting on bars. After 6 hours, it was retrieved from the container, excess water was wiped out with absorbent paper and the sample was weighed on standard scales (initial absorptivity). It was then replaced in the container until the next weighing, 24 hours after the first immersion (secondary absorption).

The standard requires that both initial and secondary sorptivity are measured because absorption rates differ in these two periods. Sorptivity is measured according to the Equations (1) and (2):

$$S = I/t^{0.5}, \quad (1)$$

$$I = (W_1 - W_2)/(A \cdot d), \quad (2)$$

where: S = Sorptivity (mm/s^2), t = Time (s), W_2 = Total weight absorbed (gr), W_1 = Weight when leaving the oven (gr), A = Area where water penetrates (mm^2), and d = Water density ($0.001 \text{ gr}/\text{mm}^3$).

- **Penetration of water under pressure** was assayed according to standard EN 12390-8 2009. Cylindrical specimens with the upper face polished were subjected to a constant pressure of $500 \pm 50 \text{ kPa}$ for 72 ± 2 hours. To ensure the tightness of the junction between the tube and the device, a rubber O-ring 100 mm in diameter was placed and centered on the polished face of the specimen, so that during the test water pressure acted locally. At the end of the test period, the specimens were cut longitudinally and the boundary between the two zones were marked to enable appropriate measurements of maximum and average depth later on.
- To determine the **depth of carbonation** (EN 83993-1. 2013), prismatic specimens of $100 \text{ mm} \times 100 \text{ mm} \times 400 \text{ mm}$ were introduced in a climatic chamber with $1\% \text{ CO}_2$ at a temperature of $21 \pm 2 \text{ }^\circ\text{C}$ and relative humidity of $60 \pm 10\%$. After 56 days, the phenolphthalein indicator was atomized by spraying and the depth of attack was measured.
- **Accelerated Chloride ion penetration.** The immersion method was used to simulate the attack (AASHTO T259). The specimens were subjected to continuous ponding with a sodium chloride solution during 56 days. The attack was measured with the colorimetric method using silver nitrate.

3. Results and Discussion

The results obtained in the tests are shown in Table 2.

Table 2. Test results.

		HPC550	HPC700	ECO-HPC550	ECO-HPC700
Density (kg/m^3)		2291.6	2341.4	1873	1891.2
Compressive Strength (MPa)	7 days	71.5	74.59	27.12	27.46
	28 days	83.16	88.69	27.76	28.84
	90 days	96.96	102.57	30.19	32.61
Elastic modulus (GPa)	28 days	84.8	48.4	20.9	35.2
	90 days	71.6	133.8	40.1	38
Mercury intrusion porosimetry (MIP) Test	Total porosity (%)	9.83	9.73	13.4	11.17
	Average pore radius (μm)	226.7	254	295.1	267
	Porosity in capillary range pores (%)	5.1	4.2	8.11	5.64
Sorptivity ($\text{mm}/\text{s}^{0.5}$)	S initial $\times 10^{-3}$	3.54	3.55	3.94	3.53
	S secondary $\times 10^{-3}$	2.2	2.16	2.71	2.37
Water penetration under pressure (mm)	Average penetration	7.9	5.5	7.5	9.2
	Maximum penetration	17	11	11.5	12.75
Carbonation depth (mm)	56 days	2.88	0	1.66	3.97
	90 days	3.26	0	5.89	4.88
Chloride ion penetration (mm)	28 days	3.6	5.7	6.2	7
	56 days	5.9	8.6	8.2	8.6

3.1. Compressive Strength

Test results regarding compressive strength are shown in Figure 2. The increase in the amount of cement from 550 to $700 \text{ kg}/\text{m}^3$ did not carry almost any improvement in the strength (4.5% average), probably due to the traditional curing by immersion at 7, 28, and 90 days. Numerous studies on reactive powder cement (RPC) achieved mixtures with high cement content (between 800 and

1000 kg/m³) and very high strength (between 200 and 800 MPa), but using special curing such as steam, high temperature or autoclave. E.Freyssinet [39] already, in the 1930s, applied pressure to fresh concrete getting to increase its strength, later on during the 1970s, Roy and Gouda [40] obtained concrete with a strength of 650 MPa under pressure and temperature. The aim of our study, however, was to obtain a concrete capable of replacing NC for in situ usage, which prevented us to subject it to such treatments.

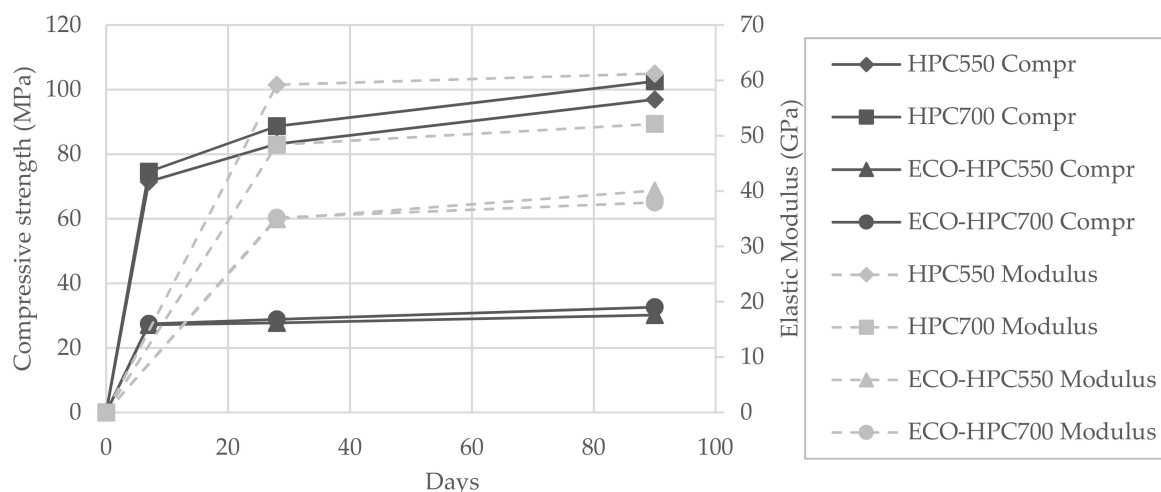


Figure 2. Compressive strength and elastic modulus evolution over time.

Generally, LWCs reach almost the total (85%) of their compressive strength by 7 days. Thorenfeldt [41] pointed out that LWC reaches around 80% of its strength by the 7th day, possibly due to the limit of the strength of LWA. As it was to be expected, the use of the LWA that was selected had a negative impact on the development of strength, reducing it by 68%, this is due to many parameters, among others, an irregular graining of the fine aggregates caused by the high regularity in the shape and size of recycled aggregates, as Katzer and Kobaka [42] asseverated, an established optimal field of graining of fine aggregate allows to achieve the most advantageous strength parameters of the concrete produced. In Table 2 a reduction in strength can be observed, which also means a substantial decrease in density. Even if the strength remains within the ranges of structural usage, a linear relationship between compressive strength and density can be observed.

3.2. Elastic Modulus

Figure 2 shows the evolution of the results of the tests over time. The concretes assayed reach high elastic modulus values. The fewer voids the concrete contains, the higher its modulus will be [43]. High compactness in the microstructure restricts the movement of the shrinkage in the concrete, which causes high internal tension. Such pressures lead to high elastic modulus values.

The elastic modulus is a characteristic of concrete that is related to strength, shrinkage and microstructure. Neville [44] stated that LWC usually bear great shrinkage due to their low elastic modulus and the substantial proportion of fine aggregate that originates many voids. The relation between shrinkage and modulus seemed to be so close that Reichard [45] proposed a correlation between both. Even if such correlation is not valid for LWC, it does suggest that at low modulus should expect large shrinkages. Li and Li [46] explored this issue and proposed a model to determine autogenous shrinkage of SCC based on the knowledge of the critical capillary diameter, the Poisson's ratio and the elastic modulus. This was also observed by Beushausen and Gillmer [43], who obtained values similar to those of our LWC in their mortars with low w/b ratio, both in elastic modulus and in shrinkage.

3.3. Mercury Intrusion Porosimetry (MIP)

Table 2 shows the results of MIP. It is observed that total porosities of both HPCs are very similar and lower than those of the LWCs. The porosity of ECO HPC 700 is 24% lower than that of ECO HPC 550. Although the cement content does not influence compressive strength, it does modify the porous structure of the material, especially in concrete with a high volume of paste. As for capillary porosity ranges, HPC 700 has 18% less capillary porosity than HPC 550; and ECO HPC 700 30% lower than ECO HPC 550. Regarding the substitution of LWA by natural aggregate, MIP shows that HPC 700 has 25% less capillary porosity than ECO HPC 700 and HPC 550, 37% less than ECO HPC 550.

The dimensional distribution of the pore size is shown in Figure 3. It may be observed that HPCs have low porosity when pore diameter is higher than $0.070\ \mu\text{m}$; porosity rises sharply between $0.050\ \mu\text{m}$ and $0.030\ \mu\text{m}$ diameter then porosity decreases from 0.03 to the smaller pores where is consistently low. The lines of both HPCs overlap, which means that they do not show substantial differences in their porosity ranges. This suggests that, as far as total porosity is concerned, the amount of cement or Ns used has no effect.

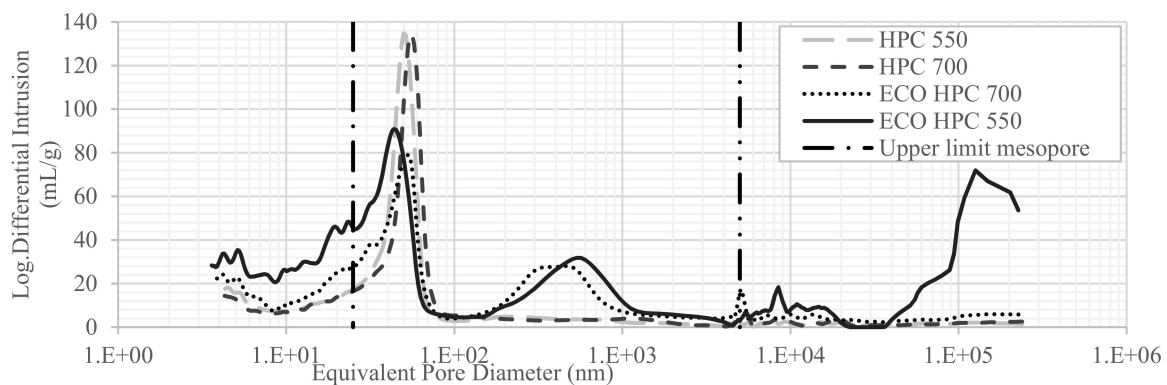


Figure 3. Dimensional distribution of sample pores.

On the other hand, the LWCs have 3 peaks, at diameter $6\ \mu\text{m}$, $0.5\ \mu\text{m}$ and $0.045\ \mu\text{m}$. The last two are within the range of capillary porosity, the latter being the most pronounced. The main difference is in the pores under $0.045\ \mu\text{m}$: at this peak ECO HPC 550 shows higher porosity than ECO HPC700; besides, it decreases more slowly. This situation remains unvaried at diameters below $0.045\ \mu\text{m}$.

Therefore, in LWC with plastic and cork aggregates replacing 50% of the natural aggregates, increasing the amount of cement and Ns reduces the porosity at pore diameters under $0.045\ \mu\text{m}$ due to the improvement in the ITZ between LWA and cement paste, a fact that does not affect the ITZ between natural aggregate and cement paste.

Figure 4 compares the concretes assayed in this study with others in the scientific literature. It can be observed that porosities are among the lowest. Bermejo Núñez, et al. [47] compare concretes obtained using different SCMs (pozzolans and slags): for the mixtures with SCMs curves are similar to the LWCs of the present work, while for the mixture with cement only, the curve obtained is similar to HPCs. The authors attribute this to lower durability of mixtures with pozzolans.

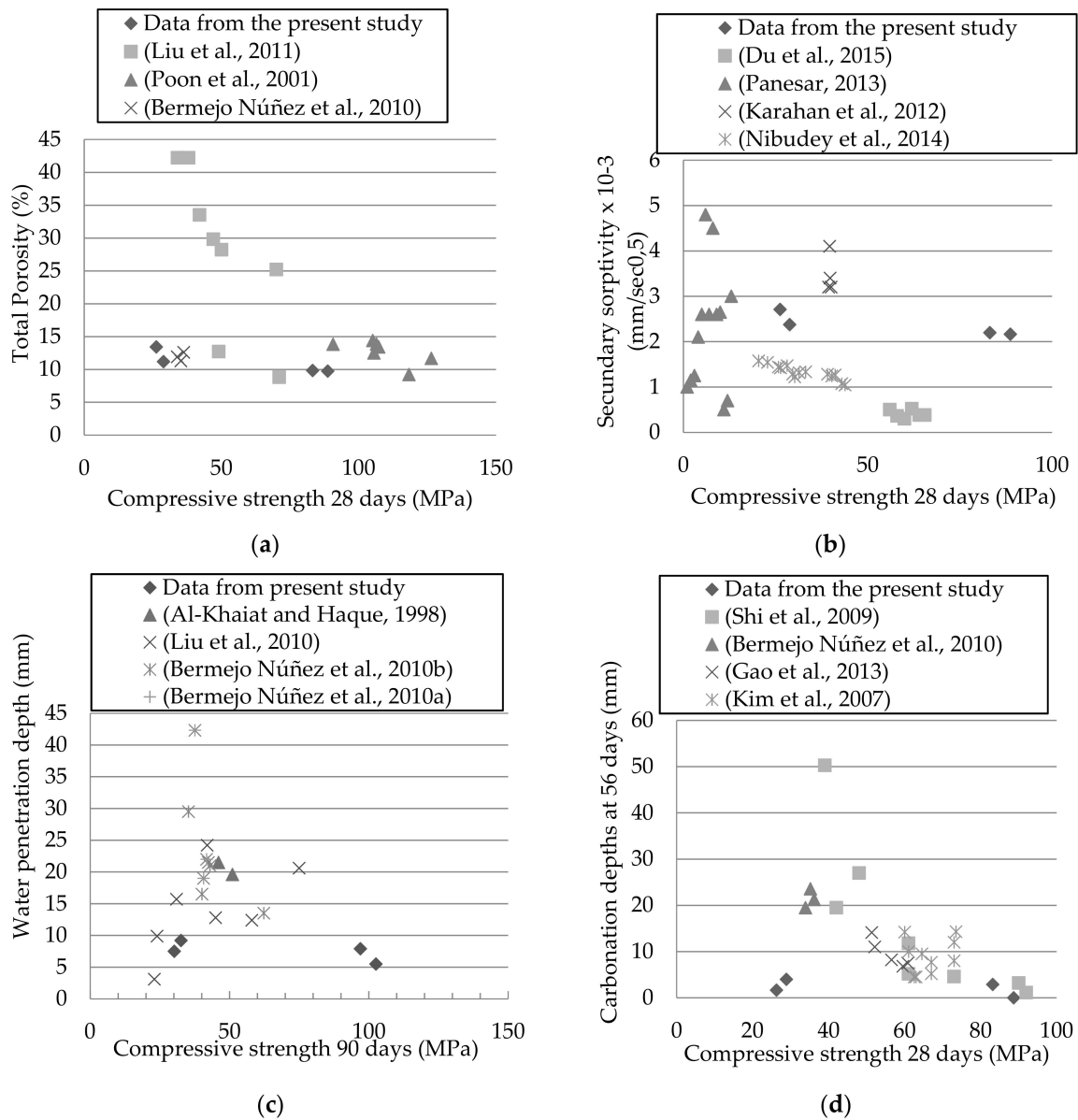


Figure 4. Results, compared with other mixtures of the scientific literature, of the test of: (a) MIP; (b) capillarity; (c) water penetration; and (d) carbonation.

3.4. Sorptivity

Figure 5 shows the water absorption data in function of time, distinguishing between initial sorption—up to 6 hours, and secondary sorption—after 24 hours. As observed in Table 2, the absorption during the first hours is very similar for all four concretes, while the differences stand out starting at 24 hours. HPC concretes have very similar levels in the early phases of secondary sorptivity with a tendency to increase over time. Differences between LWCs are more constant over time; the differences in LWC absorption are 57% higher than the differences between HPCs. This may be due to high levels of cement and Ns in HPCs, which may cause the water to be absorbed more slowly over time.

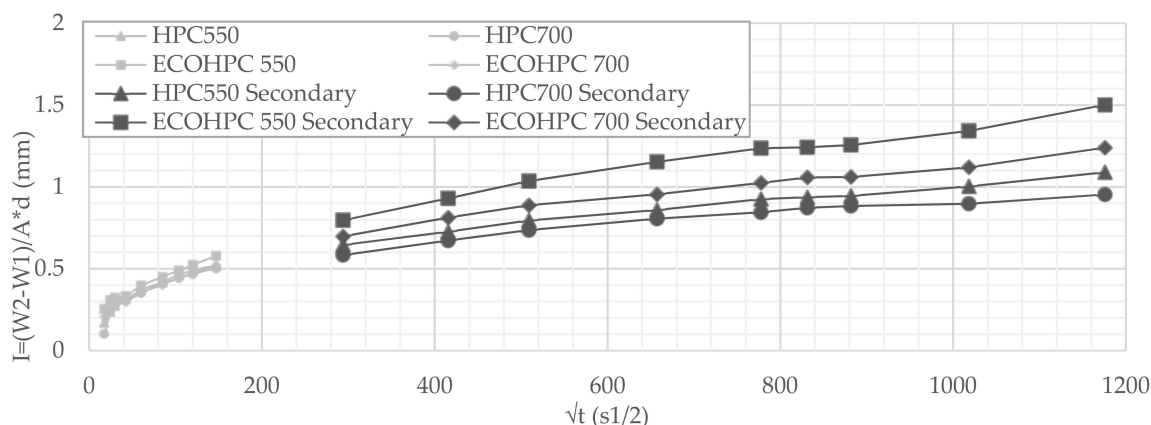


Figure 5. Water absorption versus Square root of time.

In initial sorptivity the data are very similar, ECO HPC 550 being the only one that shows absorption levels higher than the rest. In secondary sorptivity, the data show greater differences among the mixtures and results are very similar to those of porosimetry by MIP, HPC 700 being statistically equal (2% difference), to HPC 550.

LWCs absorb more water, the difference between ECO HPC 700 and 550 being greater than for HPC (12%). The impact of replacing natural aggregates with lightweight ones is low in the case of HPC 700 and ECO HPC 700—the HPC absorbs 9% less than the LWC. In 550 cement concrete the difference is greater: HPC 550's secondary sorptivity is 19% lower than that of ECO HPC 550. These data only confirm those of porosimetry and that, in fact, pore ranges within the limits of capillary porosity are directly related to the absorption of water by capillarity.

The results of secondary sorption with compressive strength are plotted in Figure 4, in which the sorptivity–density relationship of the mixtures assayed in this work is compared with other studies. Du, Du, and Liu [5] analyze the effect of Ns on several properties of LWC. They conclude that, Ns in a concrete with type I cement, improves compressive strength, sorptivity, water penetration, porosity and chloride attack. Nibudey, et al. [48], study the compressive strength and the sorptivity of concrete with recycled plastic fibers. Karahan, et al. [49] analyze the effect of Metakaolin in self-compacting concrete, finding it has a positive effect, and Panesar [50] tests LWC with different types of foaming agents and analyzes their effects.

3.5. Water Penetration under Pressure

The results of the test are given in Table 2. As in the MIP and sorptivity tests, the results show that HPC 700 shows the best performance given its lower porosity. On the other hand, HPC 550 allows the same penetration as its equivalent in LWC. As for LWCs, ECO HPC 700 has higher water penetration values than ECO HPC 550. There is a relation between density and water penetration.

In any case, despite the differences obtained, the penetration values are much lower than the maximum values permitted by law. In case of aggressive environmental exposure, the EHE-08 Spanish standard requires to comply with the specifications for the water penetration test, which are 50 mm maximum depth and 20 mm average depth, that is, 54% more than the concrete with highest penetration among those who have been assayed. This implies that although some values do not correspond directly to the sorption and MIP tests, overall they are so low that the differences between the four concretes are minimal and all of them have markedly low values.

Figure 4 compares the values obtained for average penetration with other studies. The results of the test match those obtained by Liu, et al. [51] that developed LWC with high strength to penetration of water and chlorides and obtained concrete with penetrations as low as those of the present study. For this they designed a concrete with 600 kg/m³ of cement, that is, a content in cement higher than the 500 they used in the rest of mixtures, adding silica fume (not used in the rest), a carefully selected

granulometry and several different LAWs, expanded glass among others. In both cases the high cement content is the same, as well as the use of SCMs with silicon and the combination of several types of aggregate.

Ji [9] points out that water penetration depth is 55% lower in Ns concretes compared to mixtures with no Ns. Besides, the analysis of their microstructure justifies this improvement insofar as Ns absorbs $\text{Ca}(\text{OH})_2$ crystals reducing its quantity, which densifies the interstitial zone. In addition, it can fill the gaps of CSH gel.

Therefore, the high cement content and the presence of Ns explain the reduced levels of penetration of the samples, while the low accessibility of the porosity due to its tortuosity explains the differences with the porosity tests.

3.6. Carbonation Depth

One of the main reasons for the deterioration of concrete constructions is carbonation. The reaction of atmospheric carbon dioxide (CO_2) with the products of cement hydration (such as $\text{Ca}(\text{OH})_2$), produce calcium carbonate (CaCO_3). This reaction results in a reduction of the PH of the concrete from 12.6 to less than 9, which can destroy the passive oxide layer that protects the reinforcement from corrosion [52,53].

Table 2 shows the carbonation depth of the concrete object of the study. The values achieved are very low, even zero carbonation after 90 days in the chamber in the case of HPC 700. This is explained by the fact that when CO_2 enters the concrete, it reacts first with the available calcium hydroxide ($\text{Ca}(\text{OH})_2$) in the pore network and, once it is used up, it reacts with CSH [54]. The depth of carbonation of concrete is conditioned by the porous structure; Valcuende and Parra [55] showed that the thinner and more tortuous the porous structure, the slower the process. For the concretes assayed in this work, MIP, sorptivity and water penetration tests show very low porosity in all the mixtures.

The incorporation of Ns produces a small reduction in PH and the content of alkali ions in the porous structure of the concrete. High pH and alkaline content accelerate the chemical reaction of carbonation [56]. In addition, Ns increases the chemical stability of the products of cement hydration, which also improves their resistance to carbonation [57].

The concrete curing method also has an impact. Concretes cured in a humid chamber usually present higher carbonation than those that are immersion cured. This is the case of the concretes of this study [58]. This is possibly due to the slight superficial drying necessary to convey the water to the interior, which increases the capillary porosity of the concrete surface.

Figure 4 shows how the carbonation depths of the concretes assayed are in the lower ranges of the studies analyzed. In the work of Shi, et al. [59], there are concretes with similar levels to those analyzed that have high amounts of cement (550 kg/m^3) and low water/cement ratio.

There is consensus on the direct relation between low w/c ratio and low carbonation [60,61], but not on the influence of the amount of cement. Wassermann, Katz, and Bentur [60] found that concrete with different amounts of cement but equal relation w/c showed the same depths of carbonation; however, maintaining the amount of cement but varying the a/c ratio, carbonation was indeed affected. On the other hand, in that study the maximum amount of cement used was 450 kg/m^3 , and in studies with amounts of cement above 500 kg/m^3 the penetration of carbonation was very low or zero. This is the aforementioned case of Shi, Xu, and Zhou [59] and others [62–64]. In scientific literature, therefore, it has been stated that an amount of cement greater than 500 kg/m^3 and w/b > 0.4 ratio lead to very low or even zero carbonation. The tests of accelerated carbonation at 56 and 90 days confirm this.

Regarding the influence of aggregates—plastic and cork in this case, according to Tragardh [65] or Tam, et al. [66], the ITZ is the weakest area of cementitious compounds. Therefore, if the aggregates get an ITZ resistant with the paste, the carbonation will be higher. Thus, for LWC, Rodhe and X-Betong. [67] compared three types of LWA obtaining different depths of carbonation independently of the strength, which in their opinion is due to the improvement in the porous structure of the ITZ. The use of Ns,

given that its positive impact on the ITZ of the aggregate-paste has been demonstrated, is another factor of improvement in the penetration of carbonation.

3.7. Chloride Ion Penetration

Concretes exposed to marine environments are attacked by salts such as sodium chloride, magnesium sulfate, and bicarbonates. Such salts dissociate and migrate by capillarity to the concrete; the attack is particularly aggressive on the armor of reinforced concrete.

There are different methods to measure the resistance to attack by chlorides in concrete. These, in turn, lead to studies to be able to compare the results or analyze their validity [68]. In this case the immersion method has been chosen to simulate the attack and the colorimeter process to measure it.

The penetration was measured by spraying the specimens with silver nitrate solution (AgNO_3). The depth was determined by the whitish color due to silver chloride precipitation. The concentration of chlorides needed for the color change to occur is 0.07 N in concrete with Portland cement. According to Otsuki, et al. [69], the color change becomes visible when the chloride content is greater than 0.15% of the weight of the cement.

The results of the tests carried out after 28 and 56 days of exposure are shown in Table 2. The four concretes show low penetration, three being statistically similar, while HPC 550 is slightly lower than the others. As have been seen, in the present work porosity of the concretes is reduced and low porosity hinders the advance of the attack by free chlorides [51].

Other authors have studied mixtures in which LWCs were more resistant to such attack than NC. Nyame [70], for example, analyzed mortars with fine LWA, with higher porosity than its equivalent with traditional aggregate, but with lower chloride ion penetration. This was attributed to the reduced filtration in the ITZ and to the better internal hydration of the cement by the aggregate (Thomas [71] and Liu, Chia, and Zhang [4]).

Regarding the use of SCMs, Hooton and Titherington [72] and Detwiler, et al. [73] studied their effects on chloride ion penetration, and found that their use had a positive impact against this attack.

Ns also plays an important role in resistance to this attack. Said, et al. [74], for example, analyzed the properties of concrete with Ns, also using the colorimetric method for the penetration of chlorides. They found that the samples without Ns showed penetrations of 8 and 10 mm while those with Ns obtained levels close to 5 mm, similar to those of the concrete of this work. The use of FA replacing the cement was also analyzed in the previous work, resulting in similar levels in the samples regardless of the substitution ratio.

The work of Zahedi, et al. [75] analyzes mortars with high content in cement and Ns, being the mixtures containing Ns more resistant to chlorides. The variation in cement content (between 825 and 1000 kg/m^3) does not seem to have an effect.

4. Conclusions

In accordance with the results of the experimental work carried out, the following observations and conclusions can be drawn:

- The incorporation of recycled aggregates produces a decrease in the compressive strength due to their lower strength and surface characteristics, weakening the ITZ aggregate-cement paste. In this work it has been obtained concrete with light recycled aggregates within the margins of strength established by Eurocode 2, FIB Model code 2010, as well as many of the international codes and standards for structural LWCs.
- Replacing traditional aggregate with plastic does not alter the strength as much as other traditional LWAs, while cork greatly reduces density. The combination of both LWAs allows to balance density and strength to obtain structural LWC. The LWA assayed reduces strength by 68% and density by 19%.

- The elastic modulus is also related to compressive strength, but such relationship is much more diffuse. This is because the modulus is highly influenced by microstructure and shrinkage. A low porosity in the capillary range diminishes the shrinkage capacity of concrete and the stresses caused induce high values of elastic modulus. It is interesting to note how little influence aggregates have on porosity, elastic modulus and shrinkage.
- In all concretes, the volume distribution of the obtained pores follows a similar pattern. It is porosity in the capillary range that affects strength. In this range HPC 700 has the lowest percentage (4.2%), HPC 550 has lower overall porosity (9.83%) but, in a range of finer pores, a greater quantity of smaller pores causes a higher shrinkage due to the loss of humidity by capillary attraction.
- ECOHPCs have greater capillary absorption (58% for those with 550 kg of cement and 50% for the 770 s). The absorption of HPC is much lower than ECOHPC, probably because of the absence of light aggregates. HPC 550 is 8.20% more absorbent than HPC 700, perhaps due to the lower density of its cementitious matrix. ECOHPC 700 is 35% less absorbent than HP and 80% less than ECOHPC 550.
- All the concretes have a very low carbonation depth (below 6 mm at 56 days), with faces of specimens in which the carbonation front has practically no depth, especially in HPC 700. Similar results were obtained from tests of water penetration (below 10 mm) and chloride ion penetration (below 10 mm at 56 days).

The concretes designed and analyzed in this research have demonstrated that the use of recycled LWA changes substantially the density of the concrete and its compressive strength (basically in a linear way). However, on the positive side, it has been found that the 4 reference dosages adopted have behaved similarly in the durability tests (chlorides, carbonation, penetration of water, etc.); in particular the zero or almost zero carbonation of all HPC and ECOHPC stands out along with chloride penetrations of less than 1 cm in all the mixtures. This means that once the loss of mechanical strength of ECOHPC compared to HPC is controlled, it will be possible to design very high-performance concrete for different structural applications, with an extended useful life and a low carbon footprint.

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