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Long-Term Behavior Related to Water Ingress in Mortars Which Combine Expanded and Natural Cork Lightweight Aggregates and Eco-Friendly Cements

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Abstract: The water ingress plays an important role in building materials' degradation. The use of lightweight aggregates is interesting in terms of sustainability, because they reduce the density of cement-based materials, among other advantages. The development and use of new lightweight aggregates, such as cork granulates, is a current research topic. In the present work, water ingress performance of sustainable mortars which combined expanded and natural cork aggregates and cements with slag, fly ash and limestone has been studied. Mortars produced with sand and expanded clay were also prepared. Bulk density, water absorption, drying capacity and gel and capillary pores were studied. Tests were carried out at 28 days and 1 year. A good behavior has been generally observed when an addition was incorporated to the binder, especially slag or fly ash. Regarding the new non-standardized lightweight cork aggregates, mortars with natural cork showed lower water absorption and lower volume of permeable pore space in the long term than mortars with expanded cork. At one year, natural cork mortars had an adequate water absorption performance compared to those with expanded clay, which may be due to the high volume of small capillary pores (100 nm–1 μm) in natural cork mortars.

Keywords: lightweight aggregates; expanded clay; expanded cork; natural cork; blended cements; water ingress

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1. Introduction

The improvement of sustainability in the construction sector is nowadays an important issue around the world [1–5]. For the construction of more eco-friendly structures and buildings and improving their performance, several strategies have been developed, such as using lightweight or recycled aggregates [6,7], supplementary cementitious materials [8,9] and fibers [10,11].

With regard to sustainable cements, they usually incorporate additions, which can partially or totally replace clinker [12,13]. Apart from the environmental advantages resulting from the reduced amount of clinker needed in these cements with additives, which leads to a decrease in CO_2 emissions during their production, most of the additives used to replace clinker are actually waste materials generated in other industrial sectors. Therefore, their reuse also brings about additional ecological benefits, such as reducing the amount of these residues being stored in landfills. Moreover, it has been reported that many additions enhance the properties of cementitious materials [13–16], then the study of their behavior for specific applications or in combination with other sustainable construction materials still constitutes at present a relevant investigation field [17–20]. Regarding the standardized additions, on the one hand there are active additions, which have pozzolanic and/or hydraulic activity, i.e., fly ash [13,18] and blast furnace slag [21–23]. On the other hand, there are inert additions, which mainly have a filler effect in the materials, limestone being one of the most common [24–26].

The incorporation of lightweight aggregates is also interesting in terms of sustainability improvement. One of their most important advantages is that this type of aggregate produces a reduction in mortar and concrete density, leading to a reduction in construction elements' self-weight. Moreover, it also contributes to improving their thermal resistance. The study of new types of lightweight aggregates is a current research field, seeking to improve or complement the currently used lightweight aggregates, i.e., expanded clay. In this topic, several works [27–30] have explored the possible use of cork granulates and other industrial wastes as lightweight aggregates. The combination of new cork lightweight aggregates with cements containing additions could constitute a potential field of investigation to obtain more environmentally friendly cementitious materials for specific building applications.

One of the factors that has a high influence on building materials' durability is the presence of water. Problems associated with moisture in buildings lead to discomfort for their users, also leading to degradation over time [31]. Furthermore, water constitutes one of the main ways for the ingress of aggressive ions, such as chlorides, into the microstructure of materials [32], favoring the development of pathologies and other damages in structures and other building elements over time, which could affect their long-term service properties. According to that, it is noteworthy to analyze the performance of cementitious materials under the action of water, and in particular to characterize parameters related to water ingress of these materials [33,34]. Due to the major importance of this issue, it is currently a relevant topic of investigation [35,36].

This work aims at studying the long-term performance related to water ingress of sustainable mortars, which combined cements with additions and new cork lightweight aggregates. Cements used included fly ash, limestone and blast furnace slag as clinker partial replacements. The new lightweight aggregates consisted of expanded cork and natural cork. In addition, mortars with the abovementioned cements produced with expanded clay and sand as the only aggregate were also prepared, for comparing their performance with that observed for mortars incorporating cork aggregates. The main novelty of the carried-out research consists in quantifying the performance of the abovementioned combination of expanded and natural cork aggregates with eco-friendly cements with additions in mortars, which has hardly been studied.

2. Experimental Setup

2.1. Sample Preparation and Materials

Mortars manufactured with different aggregates and binders have been tested. Four different binders were used. For reference purposes, mortars with ordinary Portland cement, CEM I 42.5 R (standard EN 197-1 [37]), were made. This cement was referred as "1" in the mortar designation. In addition, three binders with different additions were also used for preparing the mortars. The first one consisted of commercial cement with limestone powder, CEM II/B-L 32.5 N [37], with a content of this addition (in weight) between 21% and 35% as clinker substitution, being named as "2" in the mortar designation. A commercial cement with blast furnace slag (70% in weight as clinker replacement), CEM III/B 32.5 N/SR [37], was also used, which was referred as "3" in the designation of the mortars. Finally, a fly ash binder has been studied. This binder was prepared with 20% in weight of fly ash and 80% in weight of cement CEM I 42.5 R [37], accomplishing the requirements of a standardized cement type CEM II/A-V. This last binder was designated as "4". The reason for selecting the binders with the abovementioned additions (limestone, slag and fly ash) is because they are now the most used additions in Portugal and Spain for manufacturing blended cements, so this could make a possible real application of the research presented in the work easier. For easier comprehension of the composition and meaning of the binders used, they are compiled in Table 1.

Table 1. Description of the binders used.

Designation	Binder (EN 197-1 [37])	Description (Percentages in Weight)
1	CEM I 42.5 R	Commercial cement consisting of ≥95% clinker and ≤5% minority components
2	CEM II/B-L 32.5 N	Commercial cement with 65–79% clinker, 21–35% limestone and ≤5% minority components
3	CEM III/B 32.5 N/SR	Commercial cement consisting of 70% blast furnace slag, 25% clinker and 5% minority components
4	Equivalent to CEM II/A-V	Binder prepared with 20% fly ash and 80% cement CEM I 42.5 R

The mortar series studied were made with a water/binder ratio of 0.5 (in weight). The aggregate/cement ratio was 3/1 (in weight) for all the mortar series. The mixing of the components of the mortars was performed following the steps established in the standard EN 196-1 [38], incorporating first the sand and then the lightweight aggregates. For each one of the binders, a reference series was prepared with only sand as the aggregate. In this reference series, the designation REF was added to the name of the mortar. After that, different mortars were produced, in which 50% of the sand volume was replaced by an alternative lightweight aggregate. This percentage was chosen according to the results of the previous works of the authors [27–30] with the studied lightweight aggregates. These aggregates were expanded clay, expanded cork and natural cork. They were named as ECL, ECK and NCK, respectively. The reason for studying the expanded and natural cork aggregates in this work is because several previous studies [27–30] have pointed out the possibility of using them in cement-based materials with preliminary good results. In addition, expanded clay has also been studied here because it is one of the most used standardized lightweight aggregates, so it may be relevant to compare the behavior of the new lightweight cork granulates with a currently well-known lightweight aggregate, such as expanded clay. The complete designation of the mortar series, in which the studied binders and aggregates were combined, is shown in Table 2.

Table 2. Designation of the mortars studied according to the binder and aggregate combined in each one.

Designation	Binder	Aggregate (Percentage in Volume)			
		Sand	Expanded Cork	Natural Cork	Expanded Clay
1REF	CEM I 42.5 R	100%	-	-	-
2REF	CEM II/B-L 32.5 N	100%	-	-	-
3REF	CEM III/B 32.5 N/SR	100%	-	-	-
4REF	Equiv. to CEM II/A-V	100%	-	-	-
1ECK	CEM I 42.5 R	50%	50%	-	-
2ECK	CEM II/B-L 32.5 N	50%	50%	-	-
3ECK	CEM III/B 32.5 N/SR	50%	50%	-	-
4ECK	Equiv. to CEM II/A-V	50%	50%	-	-
1NCK	CEM I 42.5 R	50%	-	50%	-
2NCK	CEM II/B-L 32.5 N	50%	-	50%	-
3NCK	CEM III/B 32.5 N/SR	50%	-	50%	-
4NCK	Equiv. to CEM II/A-V	50%	-	50%	-
1ECL	CEM I 42.5 R	50%	-	-	50%
2ECL	CEM II/B-L 32.5 N	50%	-	-	50%

Table 2. *Cont.*

Designation	Binder	Aggregate (Percentage in Volume)			
		Sand	Expanded Cork	Natural Cork	Expanded Clay
3ECL	CEM III/B 32.5 N/SR	50%	-	-	50%
4ECL	Equiv. to CEM II/A-V	50%	-	-	50%

The abovementioned lightweight aggregates were characterized to assess their granular dimensions and bulk density. Cork granulates were produced by Corticeira Amorim, Portugal. ECK possessed dimensions ranging from 1 to 2 mm and a bulk density of 249 kg/m^3; NCK had a diameter of 4–5 mm and a bulk density of 70 kg/m^3. ECL was produced by Argex, with dimensions of 0/2 mm and a bulk density of 490 kg/m^3 [27,28,30]. The particle size distributions of the lightweight aggregates used are depicted in Figure 1 and they can be observed in the pictures included in Figure 2.

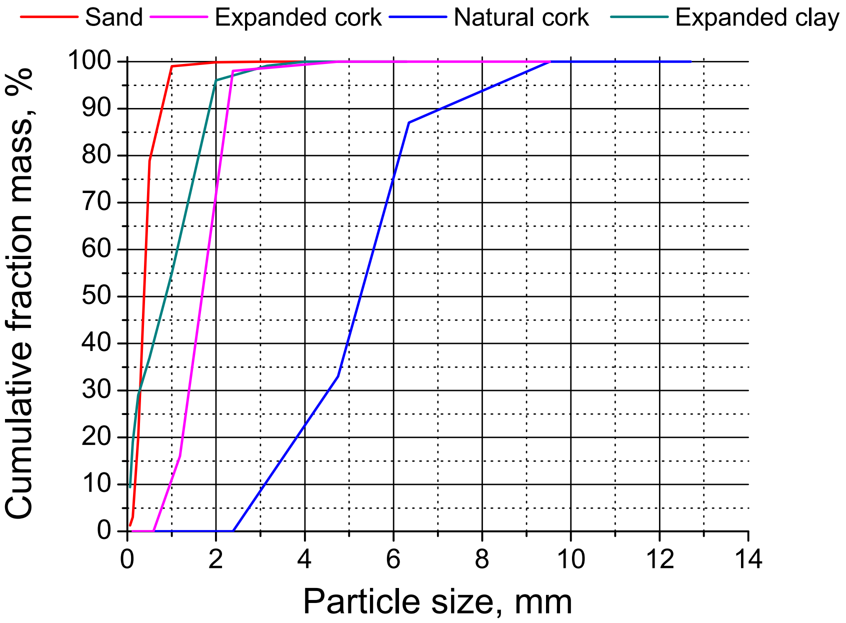

Figure 1. Particle size distribution of the sand, natural cork, expanded cork and expanded clay used in this work.


(**a**) (**b**) (**c**)

Figure 2. (**a**) Expanded cork aggregate (ECK); (**b**) natural cork aggregate (NCK); (**c**) expanded clay aggregate (ECL).

Prismatic specimens with dimensions of 4 cm × 4 cm × 16 cm were manufactured. Several images of the cross-sections of specimens prepared with the binders and aggregates studied can be observed in Figure 3. After the initial 24 h, they were removed from the casts and they were stored submerged in tap water at a controlled temperature (20 ± 2 °C) until the testing ages. Lastly, the testing ages were 28 days and 1 year, with the exception of the drying kinetics assessment test, which was only performed for one-year-old specimens.

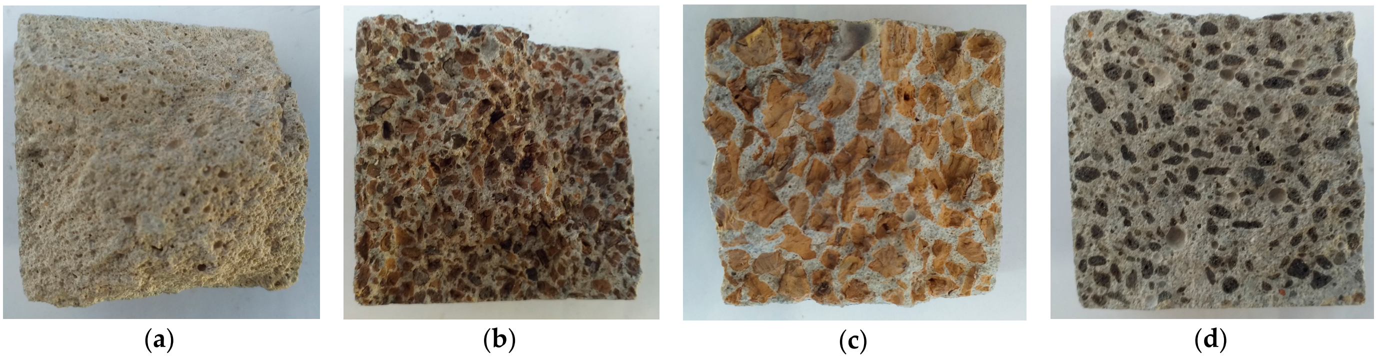
(**a**) (**b**) (**c**) (**d**)

Figure 3. (**a**) Cross-section of a prismatic sample prepared with CEM II/B-L 32.5 N cement and sand as the only aggregate (2REF series); (**b**) cross-section of a prismatic sample prepared with a binder equivalent to CEM II/A-V and expanded cork aggregate (4ECK series); (**c**) cross-section of a prismatic sample prepared with CEM III/B 32.5 N/SR cement and natural cork aggregate (3NCK series); (**d**) cross-section of a prismatic sample prepared with CEM I 42.5 R cement and expanded clay aggregate (1ECL series).

2.2. Gel and Capillary Pores

The volume of gel and capillary pores was determined with mercury intrusion porosimetry [39–41]. The porosimeter used was a Poremaster-60 GT, manufactured by Quantachrome Instruments. The specimens were oven-dried at 50 °C for 48 h before the test. In this research, the volume of the pores of the size intervals < 100 nm, 100 nm to 1 µm and 1–10 µm was analyzed. The distribution of pore volume according to size has a great influence on the durability and mechanical properties of concretes, mortars and cement pastes [42–45]. In relation to the durability properties of those cement-based materials, especially regarding their water ingress performance, gel pores and capillary pores particularly play an important role. As has been reported in several studies [42–45], gel pores are those with diameters lower than 100 nm, while the pores with sizes in the ranges 100 nm to 1 µm and 1–10 µm are associated with small and large capillary pores, respectively. Two measurements were performed on each series of mortar at 28 days and 1 year. Pieces obtained from the manufactured prismatic samples were tested.

2.3. Absorption, Density and Volume of Permeable Pore Space

The absorption after immersion, bulk density and volume of permeable pore space were determined according to the ASTM Standard C642-06 [46]. Six pieces taken from manufactured prismatic samples were tested for each mortar series at 28 days and 1 year.

2.4. Water Absorption by Immersion after 48 H

This test was performed according to the specification E394 [47] of the National Laboratory of Civil Engineering (LNEC) of Portugal. It allows obtaining the water absorption after total immersion for 48 h. For performing this test, the manufactured prismatic samples were used. The preconditioning procedure of the specimens consisted of drying them using an oven at 60 ± 5 °C until they reached a constant mass. Once that process finished, the samples were cooled in a desiccator and they were weighed in order to obtain their dried mass. The test consisted of totally immersing the specimens in a hermetic box filled with water for 48 h. The immersion procedure [48] is described in the specification E394 [47]. After finishing the 48h period, the specimens were taken out of the box and cleaned using a damp cloth for finally obtaining the saturated mass of each sample.

The parameter determined with this test is the percentage of water absorption by immersion after 48 h, calculated using the expression:

$$W_{48h} = \frac{m_2 - m_1}{m_1} \cdot 100 \tag{1}$$

where W_{48h} is the percentage of water absorption by immersion after 48 h (%), m_2 is saturated mass of the specimen after immersion (g), m_1 is the mass of the dried sample prior to immersion (g).

For each of the mortar series, three specimens were tested at 28 days and 1 year.

2.5. Drying Kinetics Assessment

This test is useful for obtaining data about the mortar performance regarding the process of internal water evaporation and their drying capacity. The analysis of the drying capacity could be interesting because the internal water content in mortars has an influence on their thermal performance, which is a relevant property for specific applications, such as plasters or coatings. The drying kinetics evaluation of the analyzed mortars was performed according to the procedure developed by Pavão [49] and Gomes [50]. For this test, the manufactured prismatic samples were used.

Prior to the test, the four lateral faces of the specimen were sealed with two coats of epoxy resin [48] and 24 h after applying the last coat, the specimens were oven-dried at 60 ± 5 °C up to reaching constant mass. After that, the samples were completely submerged in water for 48 h [48]. Once that period ended, they were taken from water and their surface was cleaned using a damp cloth. Right after, one of the unsealed base faces of the specimen was waterproofed with a plastic sheet fixed with elastic bands [48]. This ensures that the process of drying occurs only through one of basal faces of the specimen, through a unidirectional drying flow.

The initial mass of the specimens was registered, and during the test, they were exposed to a non-optimum laboratory condition (20 ± 2 °C and 50 ± 5% RH). It consisted of monitoring the drying process and weighing the specimens at different times from the start of the test: 30 min, 1 h, 2 h, 4 h, 6 h, 24 h and then each 24 h up to the registration of constant mass. The parameter resulting is the percentage of evaporated water, determined using the expression:

$$W_i = \left| \frac{m_i - m_0}{m_0} \right| \cdot 100 \tag{2}$$

where W_i is the percentage of evaporated water at the corresponding measurement time (%), m_i is the mass of the sample at the corresponding measurement time (g), m_0 is the initial mass of the sample at the beginning of test (g).

For each of the mortar series analyzed, three specimens were tested at 28 days and 1 year.

3. Results and Discussion

3.1. Gel and Capillary Pores

The evolution of the gel and capillary pores volume for the analyzed series is represented in Figure 4. This volume was lower for reference mortars without lightweight aggregates, followed by those that incorporated expanded clay, while the highest presence of gel and capillary pores was noted for specimens with natural and expanded cork. In relation to reference series, the percentage of gel pores was higher when active additions (fly ash and slag) were incorporated into the binder, especially in the long term, as indicated by the results of the 3REF and 4REF series, while it was smaller for those containing limestone (2REF series). In addition, the total intrusion volume of gel and capillary pores was lower for reference mortars prepared with ordinary Portland cement (1REF series) at both testing ages.

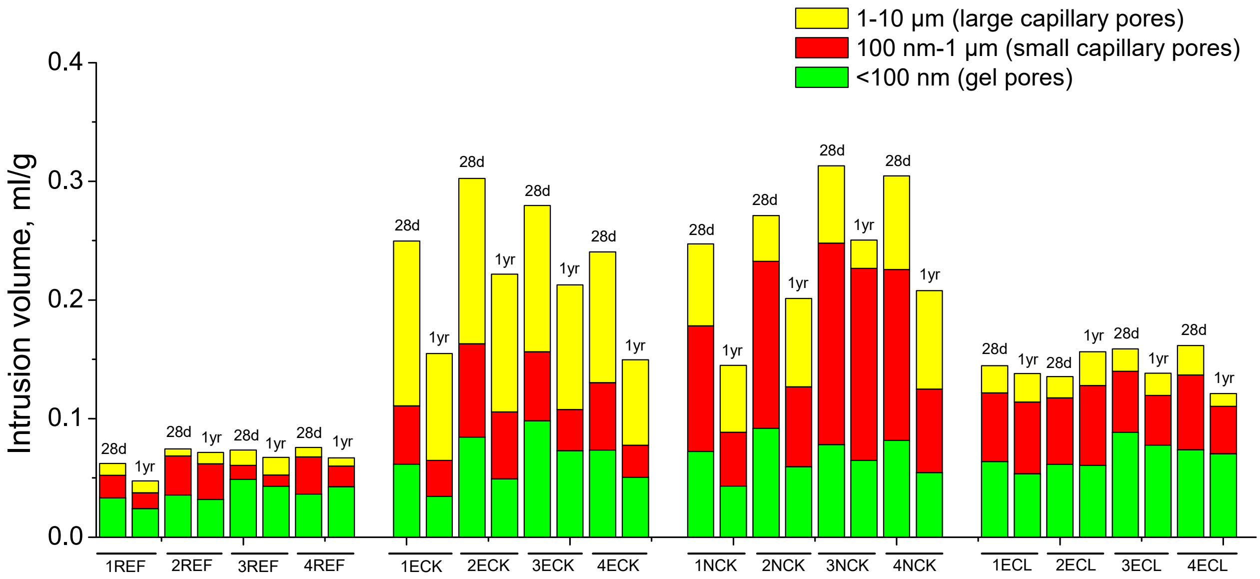

Figure 4. Volume of gel and capillary pores obtained for the analyzed mortars.

With regard to the series containing expanded cork, at 28 days, the total volume of gel and capillary pores was higher for 2ECK mortars, followed by 3ECK ones, whereas this volume was smaller for 1ECK and 4ECK mortars, being very similar for both series. At that age, minor differences in the volume of large capillary pores were observed between mortars with expanded cork, although this volume was hardly lower for series with fly ash and slag (4ECK and 3ECK series). The volume of gel pores at 28 days was higher for the abovementioned 3ECK and 4ECK mortars, when compared to 1ECK and 2ECK ones. In relation to the small capillary pores, 2ECK mortars presented a higher volume of these pores in the short term than other series with expanded cork. Between 28 days and 1 year, a lessening of the total volume of gel and capillary pores was noted for all the mortars which incorporated this lightweight aggregate. At one year, the lowest total volume corresponded to the 1ECK and 4ECK series, while it was greater for the 2ECK and 3ECK ones. However, there were differences between the distribution of gel and capillary pores in the long term depending on the cement type used. After one hardening year, the proportion of large capillary pores was relatively high for all the mortars with expanded cork, whereas the volume of small capillary pores was generally low, being higher for the 2ECK series compared to other series with this aggregate. Furthermore, the gel pore volumes after a long maturing period were higher for the 3ECK and 4ECK mortars than for 1ECK and 2ECK.

In relation to the mortars with natural cork, their total volume of gel and capillary pores was overall similar to that noted for series with expanded cork. The main difference between these series is that the inclusion of natural cork led to a lessening of the volume of large capillary pores and increased the small capillary pores' volume in comparison with expanded cork, regardless of the type of cement used. At 28 days, the total volume of capillary and gel pores was greater for the 3NCK and 4NCK series, compared to the 1NCK and 2NCK ones. Nevertheless, scarce differences in the volume of gel pores and large capillary pores were observed at that age between the different series with natural cork, whereas when an addition was used in the binder (2NCK, 3NCK and 4NCK series), a greater proportion of small capillary pores was noted, when compared to the 1NCK series prepared with ordinary Portland cement. The total volume of gel and capillary pores showed a reduction with time for all natural cork samples. For the 1NCK, 4NCK and 2NCK series, this reduction was mainly produced in the small capillary pores and to a lesser extent in the gel pores. On the contrary, the 3NCK samples mainly developed a decrease in large capillary pores between 28 days and 1 year. At this last age, the distribution of gel and capillary pores hardly differed between the 2NCK and 4NCK mortars, the total volume of these pores being scarce lower than that noted for the 3NCK series, although the proportion of pores with sizes under 1 μm was considerably greater for the 3NCK specimens compared to the 2NCK and 4NCK ones. The lowest volume of gel and capillary pores at one year was noted for 1NCK mortars. Despite that, they showed a smaller proportion of gel pores and smaller capillary pores than that obtained for natural cork mortars with additions.

The samples with expanded clay presented a similar total volume of gel and capillary pores, independently of the cement type used. In these mortars, the volume of capillary pores was lower compared to those with expanded and natural cork. The decrease with time in the capillary and gel pores was more noticeable in samples with slag and fly ash (3ECL and 4ECL series), which generally showed a higher volume of gel pores than 1ECL and 2ECL mortars at both testing ages.

Regarding the discussion of the characterization of gel and capillary pores, first of all, the lower total volume of these pores obtained for reference mortars would be consistent, and it may be due to the higher porosity of lightweight aggregates [51], which could affect the global solid fraction of the composite mortar. Moreover, the reduction with maturing age in the volume of gel and capillary pores for most of the series analyzed would be due to the progress of slag and clinker hydration [22,52] and the pozzolanic reactions of fly ash [17,18], which would produce a progressive formation of solid phases, partially closing the microstructure in the long term. The higher volume of finer pores generally observed for series with fly ash and slag, regardless of the aggregate used, could be related to the reactivity of these additions, and this result would be in keeping with other authors [17,18,53–55]. In the case of slag, it has hydraulic activity [22,52,56], so it is capable to react directly with the setting water, forming new solids, which would fill the existing pores, thus reducing the volume of larger voids in the microstructure. For fly ash addition, it develops pozzolanic activity [18,57,58], so its components are able to react with the portlandite produced in the hydration of clinker, forming additional CSH phases which also reduced the pore size. Therefore, with the results obtained in this work, the good performance of these active additions when they are combined with new lightweight aggregates, such as natural and expanded cork, has been shown. Furthermore, the result that the pore network was overall coarser when a cement with partial replacement of clinker by limestone was used may be due to the inert character of this addition [24], which has no pozzolanic or hydraulic activity, so its benefits in the microstructure are more limited, being mainly reduced to a filler effect [25,26,59].

3.2. Absorption, Density and Volume of Permeable Pore Space

The results of the absorption after immersion [46] are depicted in Figure 5. For all the studied series, an absorption reduction from 28 days to 1 year has been observed. This reduction was more noticeable for mortars that incorporated lightweight aggregates. For mortars made with slag cement, a smaller lessening of the absorption between the two testing ages was noted, especially for reference mortars and those with the two types of cork studied. The absorption after immersion was overall lower for reference mortars without lightweight aggregates (1REF, 2REF, 3REF and 4REF series), followed by specimens with expanded clay (1ECL, 2ECL, 3ECL and 4ECL series). The highest absorption values were overall registered for series with cork, being lower for samples with natural cork (1NCK, 2NCK, 3NCK and 4NCK mortars) than for those with expanded cork (1ECK, 2ECK, 3ECK and 4ECK series).

Comparing the reference series, the absorption after immersion at 28 days was slightly higher for samples with limestone addition (2REF series), although at 1 hardening year it was very similar for all reference mortars. With regard to the mortars with expanded cork, at 28 days, the highest absorption values were observed for the 1ECK and 2ECK series, while the lowest values corresponded to the mortars that combined the addition of fly ash with expanded cork aggregate (4ECK series). After one hardening year, the 4ECK series still continued to show the smallest absorption for mortars with expanded cork, and it was quite similar for the rest of the series with this aggregate, though it was scarce higher for the 2ECK and 3ECK mortars, in comparison with 1ECK ones.

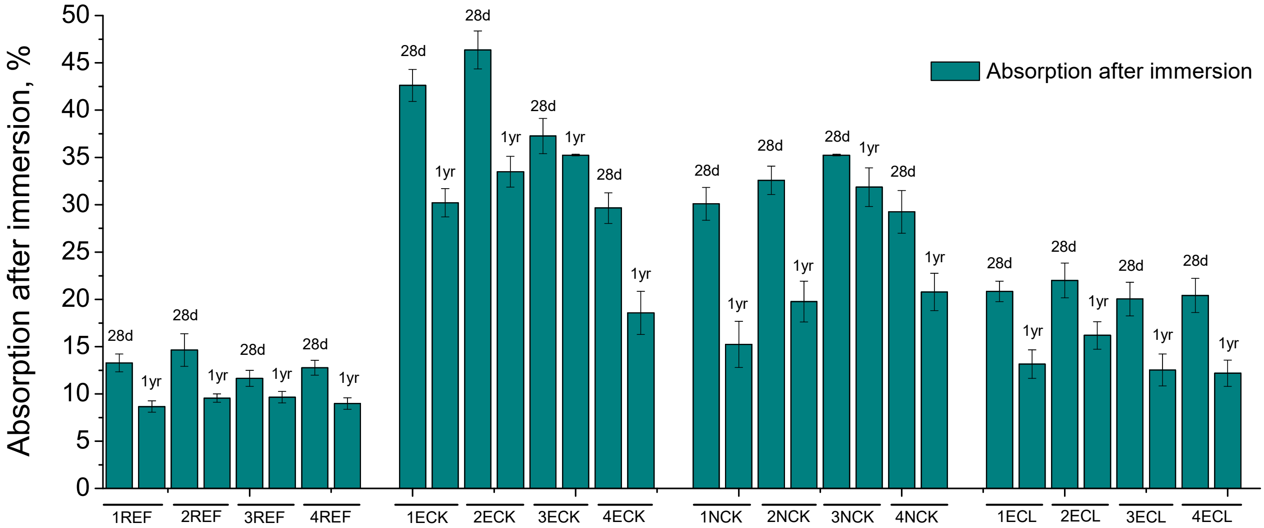

Figure 5. Absorption after immersion results.

In relation to the mortars containing natural cork, at 28 days, no high differences were noticed among them, the absorption after immersion being slightly higher for those prepared with slag cement (3NCK series). However, at one year, the lowest absorption was registered for the 1NCK samples, nearly followed by the 2NCK and 4NCK series, while it was higher for the 3NCK mortars. For specimens with expanded clay, the values of absorption noted were quite similar at both ages studies, independently of the cement type used, although it is interesting to highlight that this parameter was scarcely lower for the 1ECL, 3ECL and 4ECL mortars than for the 2ECL ones at the hardening age of one year.

The results of the absorption after immersion, previously described, showed coincidences with those obtained in the study of the gel and capillary pores in the microstructure. The decrease with maturing age would agree with the reduction in the total volume of gel and capillary pores observed with mercury intrusion porosimetry, which was explained due to the formation of solids as products of hydration reactions of clinker and slag [53–55] and pozzolanic reactions of fly ash [17,18]. Furthermore, the higher values of the absorption after immersion noted for series with expanded cork compared to those with expanded clay and natural cork may be related to the higher volume of large capillary pores observed in mortars with expanded cork. In addition, the lower presence of small and large capillary pores in expanded clay specimens in comparison with natural cork ones could explain the greater absorption values obtained in these last mortars.

The bulk densities obtained for the analyzed series are depicted in Figure 6. This parameter was mainly influenced by the aggregate used for preparing the mortars, and in general the effect of the cement kind was almost negligible. The highest bulk densities corresponded to reference mortars without lightweight aggregates. The smallest values were overall registered for expanded cork series, showing that the mortars with fly ash (4ECK series) had a slightly higher bulk density than the other mortars with this aggregate. For series with natural cork, this parameter showed scarce greater values compared to those with expanded cork. In the case of mortars which incorporated expanded clay, their density was higher than those noted for series with cork, although it was smaller compared to reference mortars.

These results would also be in line with those obtained for the pore network characterization. The higher bulk density of reference series would be compatible with the lower total volume of gel and capillary pores noted for these reference mortars compared to those that incorporated lightweight aggregates. As has been previously explained, this may be due to the higher porosity of lightweight aggregates [51]. Moreover, the greater bulk density of expanded clay mortars than that noted for those with both studied types of cork would also be in agreement with their smaller total volume of gel and capillary pores.

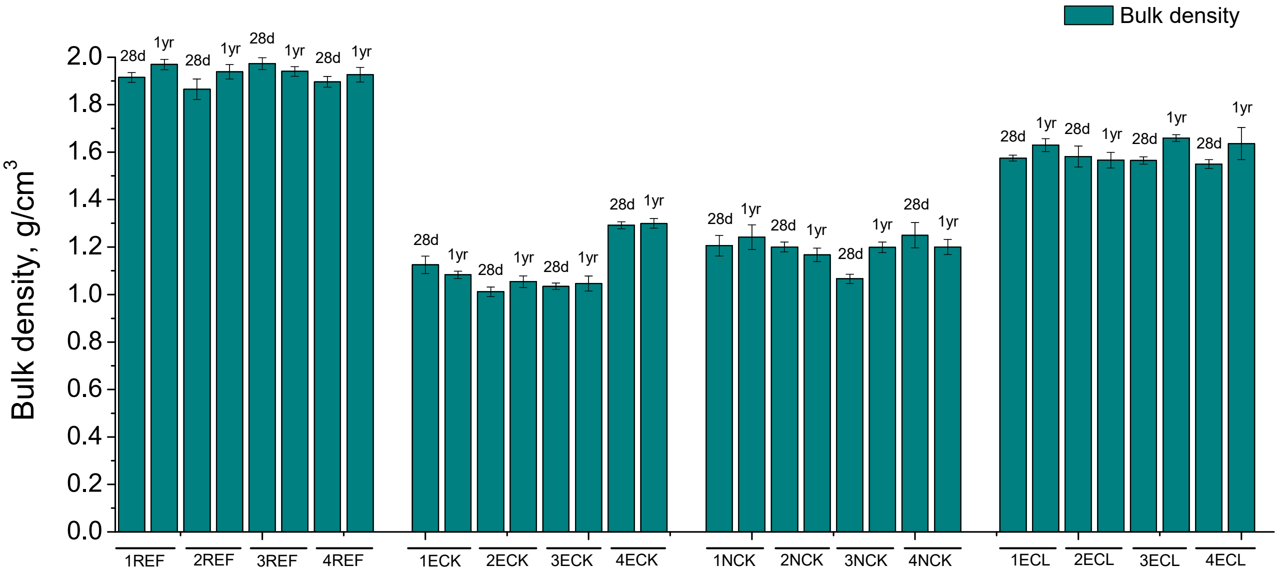

Figure 6. Bulk density results registered for the studied mortars.

The results of the volume of permeable pore space can be observed in Figure 7. It decreased from 28 days to 1 year for all the series. At 28 days, the volume of permeable pore space was lower for reference series without lightweight aggregates. The highest values of this volume in the short term were noted for series with expanded cork. The volume of permeable pore space at 28 days was slightly higher for natural cork specimens compared to those with expanded clay. However, the differences observed were not very high. After one year, the values of this volume were very similar for mortars with natural cork and expanded clay, being scarce higher than the values obtained for the reference series. Nevertheless, in general, the volume of permeable pore space at this age was still greater for specimens with expanded cork, compared to other studied series.

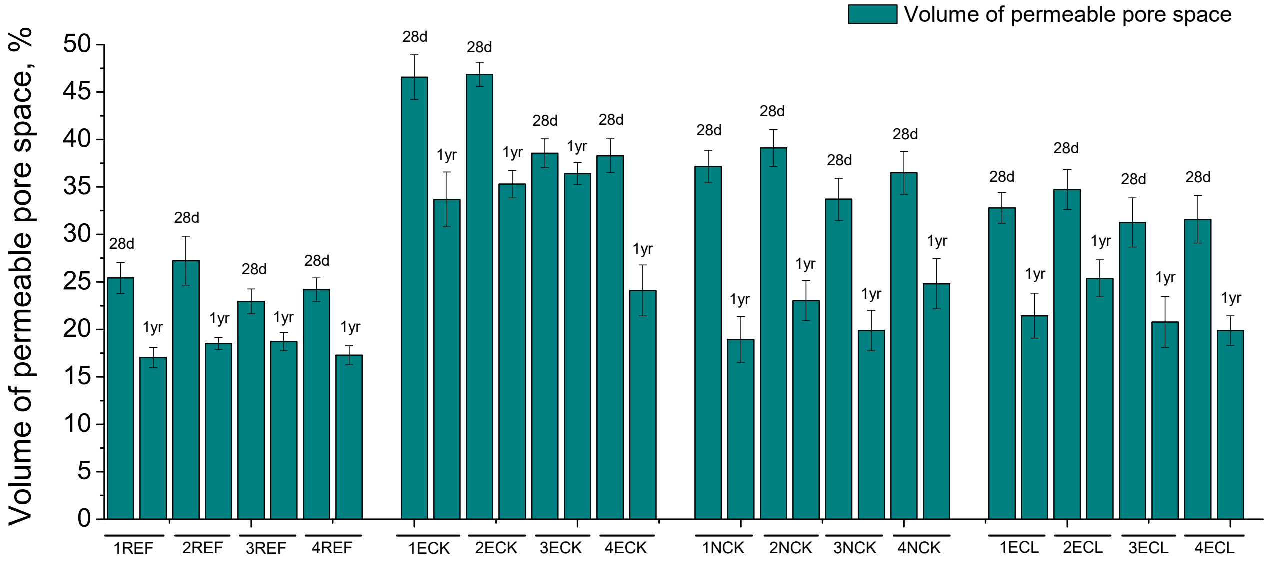

Figure 7. Evolution of the permeable pore space volume between 28 days and 1 year for the analyzed series of mortars.

Regarding the comparison between the reference mortars without lightweight aggregates, at 28 days, the volume of permeable pore space was lower for specimens with fly ash and slag (4REF and 3REF series), whereas this parameter hardly differed at 1 year for all the reference series, regardless of the cement type. For mortars with expanded cork, in the short term, the volume of permeable pore space was greater when ordinary Portland cement (1ECK series) and cement with limestone addition (2ECK series) were used, and it showed lower values when fly ash and slag additions were incorporated into the binder (3ECK and 4ECK series). At one hardening year, the 4ECK mortars presented a smaller volume of permeable pore space than the other series with expanded cork.

In relation to the samples with natural cork, series with ordinary Portland cement (1NCK series) and limestone addition (2NCK series) also presented a higher volume of permeable pore space at 28 days than those with slag (3NCK series) and fly ash (4NCK series). For all the series with natural cork, the great lessening of this parameter from 28 days to 1 year is remarkable. At this last age, the volume of permeable pore space was scarce lower for the 1NCK and 3NCK series than for the 2NCK and 4NCK ones. Furthermore, its values were quite similar at 28 days for all the mortars with expanded clay, independently of the cement used, while in the long term they were hardly greater for the 2ECL mortars in comparison with the other three series with this aggregate.

The results of the volume of permeable pore space were generally in agreement with those noted for other parameters previously discussed. Particularly, its greater values for expanded cork mortars would be in concordance with their higher total volume of gel and capillary pores, higher absorption after immersion and lower bulk density. Finally, it is noteworthy to highlight that the long-term differences of the volume of permeable pore space between specimens with expanded clay and natural cork were lower than in those observed for other parameters, showing that the majority of the natural cork mortars had similar values of this parameter than those that incorporated commercial expanded clay aggregate after one hardening year. This could be explained regarding the high presence of small capillary pores in natural cork mortars, which may rise the pore network tortuosity, reducing their permeability [60].

3.3. Water Absorption by Immersion after 48 H

The results of water absorption by immersion after 48 h [47] are shown in Figure 8. As happened for other parameters analyzed, this parameter was lower for reference mortars, independently of the cement type used, compared to those that incorporated lightweight aggregates. The highest values of the absorption after 48 h were overall noted for specimens with expanded cork. It was slightly higher for mortars with natural cork than for those with expanded clay, although the differences between them were relatively small. In general, the absorption after 48 h decreased with time, although this reduction was not as remarkable as was observed for previous parameters described, depending on the cement type used.

With regard to reference series, at both testing ages, the absorption after 48 h was slightly lower for specimens with fly ash and slag (4REF and 3REF series), showing the highest values of this parameter in those with limestone addition (2REF series). The absorption hardly reduced between 28 days and 1 year for all the reference series. In relation to the samples with expanded cork, the highest absorption after 48 h at 28 days was obtained for mortars prepared with ordinary Portland cement (1ECK series), nearly followed by those with limestone and slag additions (2ECK and 3ECK). After one year, it was slightly lower for the 1ECK mortars compared to the 2ECK and 3ECK ones. Lastly, the smaller values of the absorption after 48 h for specimens with expanded clay corresponded to those with fly ash (4ECK series) at both testing ages, which showed similar results to natural cork mortars with this addition (4NCK series) and scarce greater than those with expanded clay (4ECL series). A small decrease with age in this parameter was also observed for the expanded cork series, being more noteworthy for the 1ECK mortars.

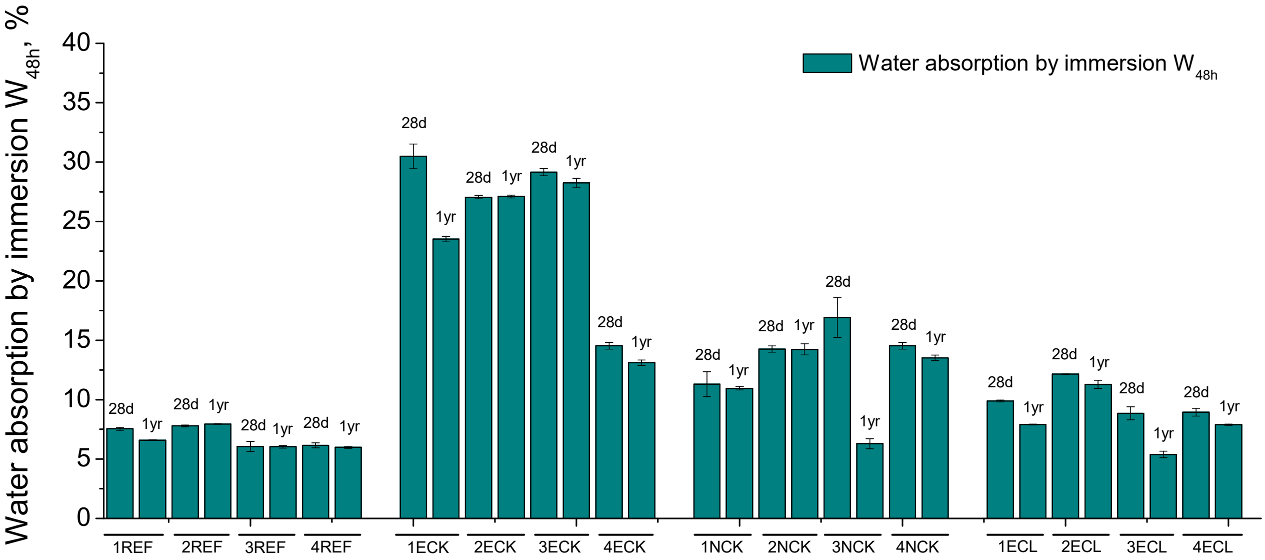

Figure 8. Water absorption by immersion after 48 h results.

Regarding the series with natural cork, the mortars with ordinary Portland cement (1NCK series) showed the lowest absorption after 48 h at 28 days, while the greatest values were registered for those with slag cement (3NCK series). From 28 days to 1 year, this parameter was noticeably reduced for 3NCK mortars, presenting the smallest absorption in the long term of all the natural cork series studied. In addition, these long-term absorption values of the 3NCK series were very similar to those obtained for the mortars which combined slag cement and expanded clay (3ECL series), and they also did not differ too much from reference specimens with slag addition (3REF series). The highest values of the absorption after 48 h for natural cork mortars at one year were observed for the 2NCK and 4NCK series. On the other hand, this parameter showed slight differences at 28 days between the different expanded clay mortars analyzed, with the exception of the 2ECL series, which presented higher values of the absorption after 48 h. It fell with age for all expanded clay series, showing the lowest values after one year for the 3ECL mortars, followed by the 1ECL and 4ECL series, while the greatest absorption was observed for the 2ECL mortars at that age.

The results of water absorption by immersion after 48 h were compatible with those obtained for previously described parameters. Firstly, the smaller values of absorption after 48 h noted for the reference series would coincide with their lower total volume of gel and capillary pores, higher bulk density and lower volume of permeable pore space, as already discussed. On the other hand, the higher values of absorption after 48 h observed for specimens with expanded cork would agree with their greater presence of gel and capillary pores, lower bulk density and higher volume of permeable pore space. Additionally, the lower large capillary pore volume of the series with natural cork could explain their smaller values of the absorption after 48 h compared to those with expanded cork.

The differences between mortars with expanded clay and natural cork with regard to the water absorption by immersion after 48 h were lower than that observed for other parameters, though it was still scarce greater for samples with natural cork. The same happened with the permeable pore space volume. It may be due to the high volume of small capillary pores in natural cork mortars, which would suggest a higher pore network tortuosity [23,52]. This fact would reduce the microstructure channel size through which the water absorption is produced [60], thus making the development of this phenomenon in the material more difficult [61], resulting in the lower values of the observed water absorption after 48 h.

Comparing the results of water absorption by immersion obtained after 48 h following the procedure of the specification E394 [47] and those obtained for the absorption after immersion according to ASTM Standard C642-06 [46], similar trends have been observed for both parameters. However, the differences between the analyzed series are lower in terms of absorption after 48 h [47] when compared to those noted for absorption after immersion [46]. Furthermore, for the same mortar series, the values of the absorption after 48 h [47] were smaller than the values of absorption after immersion [46]. The possible reason behind this could be the variation in the preconditioning method used to obtain both parameters. In the case of absorption after 48 h [37], the specimens were oven-dried at a constant temperature of 60 °C until their mass remained stable. On the other hand, for determining the absorption after immersion [36], the drying process involved storing the samples at 105 °C, resulting in a higher level of drying in this particular scenario. Finally, it is interesting to highlight that the positive effects of the active additions (fly ash and slag), probably due to their hydraulic [53,54] and pozzolanic activity [17,18] as has been explained, were more noticeable in water absorption by immersion after 48 h.

3.4. Drying Kinetics Assessment

The evolution of the percentage of evaporated water during the development of the drying kinetics assessment test for the series analyzed at one hardening year is represented in Figure 9. The series with expanded cork overall showed higher values of this percentage compared to mortars with other studied aggregates. For reference mortars, the greatest percentages corresponded to the 1REF and 2REF series, while it was lower for specimens with fly ash and slag (4REF and 3REF series). Regarding series with expanded cork, the mortars prepared with ordinary Portland cement (1ECK series) and those which incorporated limestone addition (2ECK series) had higher percentages of evaporated water, followed by mortars with slag (3ECK series), whereas the lowest values corresponded to the 4ECK series with fly ash.

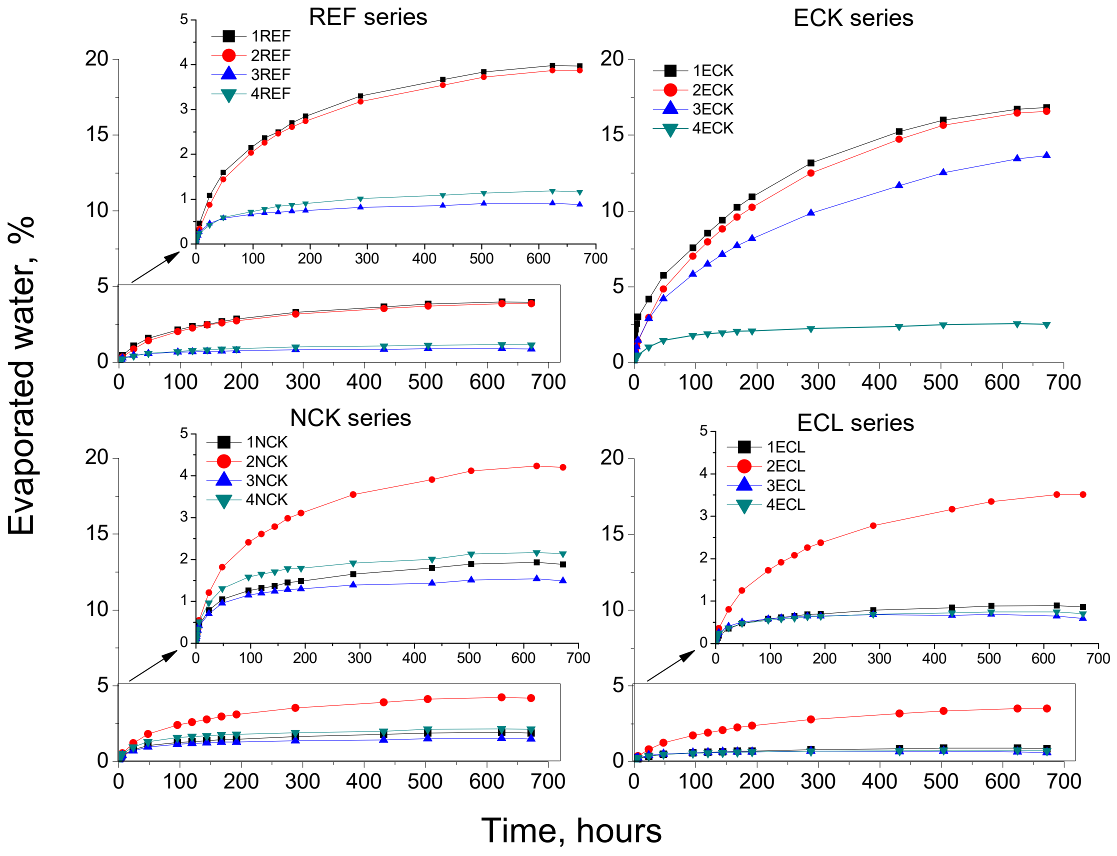

Figure 9. Variation of the percentage of evaporated water along the drying kinetics assessment test for the studied mortar series.

The percentage of evaporated water for series with natural cork was higher when cement with limestone addition was used (2NCK series). Small differences were observed in relation to this parameter between the mortars with other cements analyzed (1NCK, 3NCK and 4NCK series), although it was slightly lower when fly ash was incorporated into the binder (4NCK series). For expanded clay series, the results of the percentage of evaporated water were similar to those obtained for mortars with natural cork, as previously described, with higher values for the 2ECL series compared to the 1ECL, 3ECL and 4ECL ones, which presented scarce differences between them. Comparing mortars with expanded clay and natural cork, for the same cement type, in general, the percentage of evaporated water was hardly smaller when expanded clay was used as an aggregate. Finally, for the same binder, the values of this parameter during the drying process were similar or even lower for expanded clay and natural cork mortars compared to reference ones with only sand as the aggregate. An exception was noticed for those prepared with ordinary Portland cement without additions, which presented higher percentages of evaporated water for the 1REF series than for the 1NCK and 1ECL ones.

These results would suggest that the large capillary pores would play an important role in the drying process of internal water evaporation and the drying capacity in the mortars, because the greatest percentages of evaporated water were observed in the expanded cork series. The mortars with this aggregate had a higher volume of large capillary pores (1–10 μm), as abovementioned, through which the drying process of the material would be facilitated. In view of the results obtained, a great presence of small capillary pores (100 nm–1 μm) would not have a major influence on the development of this drying, as it would indicate the fact that the percentage of the evaporated water did not differ too much between the reference series and those made using natural cork and expanded clay. Regarding the effects of the different additions used in combination with the studied lightweight aggregates, the lower values of evaporated water in the series with fly ash and slag, for each one of the aggregates analyzed, may be due to their hydraulic [53,54] and pozzolanic activity [17,18]. These phenomena would produce a higher presence of finer pores, as has been explained for the results of mercury intrusion porosimetry. The worst performance observed in this parameter for mortars with limestone would be a consequence of the lack of reactivity of this addition [24,59], as already described, giving more limited beneficial effects in the pore structure of the material as a result, which would negatively influence the drying capacity of the material.

4. Conclusions

The main conclusions of this research can be summarized as follows:

- A decrease with time in the total volume of gel and capillary pores was observed for most of the mortar series studied, regardless of the aggregate used. This may be related to the progressive formation of solid phases as products of clinker and slag hydration and fly ash pozzolanic reactions.
- The abovementioned reduction in the proportion of gel and capillary pores had an influence on the water absorption and the volume of permeable pore space, which showed a decrease in the long term.
- For each of the lightweight aggregates studied, a good behavior has been generally observed in the mortars when an addition was incorporated to the binder, especially when slag or fly ash were used.
- Regarding the new non-standardized lightweight cork aggregates, mortars with natural cork showed lower water absorption and lower volume of permeable pore space in the long term than mortars with expanded cork. Therefore, in the context of this research, the use of lightweight natural cork aggregate would be more advisable than expanded cork in terms of protection against water ingress.
- At one year, natural cork mortars had an adequate behavior regarding the water absorption by immersion after 48 h and the volume of permeable pore space compared

to mortars with expanded clay, which may be due to the high volume of small capillary pores (100 nm–1 µm) in natural cork mortars.
- The drying capacity and the internal water evaporation were higher for mortars with expanded cork. This could be related to their high volume of large capillary pores (1–10 µm), which would make the drying process of the material easier. Scarce differences in this property were observed between reference mortars and those with natural cork and expanded clay.
- In accordance with the results obtained in this work, the use of binders with additions did not produce a noticeably worsening or even improved performance related to water ingress of the mortars with the analyzed lightweight aggregates, in comparison with those with Portland cement without additions. These results would be relevant because the combination of new cork lightweight aggregates and blended cements would provide new solutions for eco-friendly cement-based materials, which could be used in specific applications, contributing to sustainability in several ways.

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