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Chloride Ingress Resistance, Microstructure and Mechanical Properties of Lightweight Mortars with Natural Cork and Expanded Clay Prepared Using Sustainable Blended Cements

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Abstract: The use of lightweight aggregates in construction materials is a good solution for increasing the contribution to sustainability of civil engineering works, such as maritime ones. In this regard, the possibility of using cork granulates and expanded clay is a current research topic. The combination of eco-friendly cements with lightweight aggregates could provide solutions for developing new building materials. In this work, it has been studied mortars prepared with sustainable cements and the lightweight aggregates of natural cork and expanded clay. These cements incorporated slag, limestone and fly ash. Reference mortars with only sand as aggregate were also made. The total porosity and pore size distributions were obtained. The non-steady-state chloride migration coefficient and compressive and flexural strengths were also determined. The tests were performed at 28 days and 1 year. The differences in the total porosity between the natural cork and expanded clay series were not high, depending on the binder. Natural cork mortars showed similar or slightly higher migration coefficients than the reference and expanded clay mortars at 1 year. This adequate chloride resistance and the low mechanical strengths observed for the natural cork mortars recommend the possible use of this new aggregate in non-structural cement-based materials for civil engineering works exposed to maritime environments.

Keywords: lightweight aggregates; chloride ingress resistance; natural cork; expanded clay; slag; fly ash; limestone; sustainable cements; mechanical properties; microstructure

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1. Introduction

The reduction in the environmental impacts produced by the construction industry is currently a major subject worldwide [1–3]. Therefore, the development of strategies for improving the sustainability of buildings and other civil engineering works, such as those related to maritime and coastal engineering, nowadays constitutes a relevant topic of research [4–6]. Among these strategies, two of the most popular are the use of alternative aggregates [7,8], such as lightweight or recycled ones, as well as widening the possible applications of sustainable cements [9,10].

In relation to cements with low carbon footprints, they generally incorporate supplementary cementitious materials (SCMs) as clinker replacement [11–14]. Most of these materials improve the performance of mortars and concretes, such as their durability [15,16] and mechanical properties [17,18]. Therefore, the assessment of their performance for specific civil engineering applications [19,20], or combined with other ecofriendly building materials, still constitutes a relevant field of investigation [21–23]. Regarding the standardized SCMs, fly ash [9,12,24] and ground granulated blast-furnace slag [25,26] are two of the most popular. They are active additions, and so their components can develop hydraulic and/or pozzolanic activity [9,12,27], given as products additional CSH phases, which can improve the microstructure and performance of cementitious materials [25,28].

Furthermore, there are also inert additions, such as limestone, whose main effect in these materials is as filler [29–31].

The incorporation of lightweight aggregates in building materials is also a good solution for increasing the contribution to sustainability of the construction sector. These aggregates reduce the density of materials, resulting in a lessening of the self-weight of construction elements, and they may be suitable, for example, for use in the rehabilitation and retrofitting of civil engineering structures or buildings [32,33]. Moreover, they also improve the thermal resistance of these elements [32,33]. In order to provide alternatives to the most commonly used lightweight aggregates, such as expanded clay, at present, the possibility of using cork granulates has been assessed by several works [32–34]. In this regard, to combine eco-friendly cements, which incorporate supplementary cementitious materials with cork lightweight aggregates, could be a relevant topic of study for providing new solutions for sustainable building materials.

The study of the microstructure of cementitious materials is of major importance, and especially when novel components are used, because the microstructure of these materials has a direct influence on their durability and mechanical behavior [35,36]. Regarding the durability, chloride ions are aggressive agents that can cause embedded steel corrosion in reinforced concrete elements [37,38], reducing their service life. Then, the characterization of the chloride ingress behavior of cement-based materials would be interesting in order to evaluate their ability for specific real applications, and in particular for those related to marine environments [39,40]. Their mechanical properties are also relevant because most of the fundamental parameters established by the standards and codes for specific civil engineering works are related to these properties [41].

Thus, the purpose of this research is to analyze the effects in the long term of combining sustainable cements and new cork lightweight aggregates in the pore structure, chloride ingress resistance and the mechanical strengths of mortars. Fly ash, limestone and ground granulated blast-furnace slag additions were incorporated as partial clinker substitutions in the cements used. The new lightweight aggregate studied was natural cork, whose effects, combined with eco-friendly cements, have hardly been studied, being this the main novelty of the research included here. It has been also tested mortars prepared with only sand as aggregate and with expanded clay in order to compare their performance to that noted for mortars containing natural cork.

2. Materials and Methods

2.1. Materials and Sample Preparation

Mortars that were made using different binders and aggregates were tested in this work. For a better view of the meanings and compositions of the mortar series prepared, they are compiled in Table 1.

In relation to the binders, four different binders were studied. Firstly, an ordinary Portland cement, CEM I 42.5 R (European standard EN 197-1 [42]), was used, indicated as “1” in the designation of mortars. Furthermore, mortars produced with three binders containing additions were prepared. One of them was a commercialized cement containing limestone: CEM II/B-L 32.5 N [42]. The content of the limestone of this cement was in the range from 21% to 35% in weight as clinker replacement, and it was named “2” in the designation of mortars. Another binder was a CEM III/B 32.5 N/SR [42] commercial cement, which incorporated 70% in weight of blast-furnace slag as clinker substitution. It was indicated as “3” in the designation of mortars. Lastly, a binder with fly ash was also used, which consisted of 80% in weight of CEM I 42.5 R [42] cement, and 20% in weight of this addition. This binder would verify the requirements of a standardized cement type, CEM II/A-V, following the prescriptions of the standard EN 197-1 [42]. This last binder was named as “4” for the mortar designation.

Table 1. Components (binder and aggregates) and designation of the mortar series tested.

Designation	Binder		Aggregate (Percentage in Volume)		
	Cement Type [42]	Addition (Percentage in Weight)	Sand	Natural Cork	Expanded Clay
1REF	CEM I 42.5 R	-	100%	-	-
2REF	CEM II/B-L 32.5 N	Limestone (21–35%)	100%	-	-
3REF	CEM III/B 32.5 N/SR	Blast-furnace slag (70%)	100%	-	-
4REF	Equiv. to CEM II/A-V	Fly ash (20%)	100%	-	-
1NCK	CEM I 42.5 R	-	50%	50%	-
2NCK	CEM II/B-L 32.5 N	Limestone (21–35%)	50%	50%	-
3NCK	CEM III/B 32.5 N/SR	Blast-furnace slag (70%)	50%	50%	-
4NCK	Equiv. to CEM II/A-V	Fly ash (20%)	50%	50%	-
1ECL	CEM I 42.5 R	-	50%	-	50%
2ECL	CEM II/B-L 32.5 N	Limestone (21–35%)	50%	-	50%
3ECL	CEM III/B 32.5 N/SR	Blast-furnace slag (70%)	50%	-	50%
4ECL	Equiv. to CEM II/A-V	Fly ash (20%)	50%	-	50%

The studied mortars were made using a water to binder ratio of 0.5 (in weight), and with an aggregate to cement ratio of 3:1 (in weight). For each binder, a reference series was made, using only sand as aggregate. In these reference series, the abbreviation REF was added to the designation of the mortar. In addition, several mortars were prepared replacing 50% of the sand volume by alternative lightweight aggregates, which were natural cork and expanded clay. In the designation of the series, the acronyms NCK and ECL were used, respectively, for naming the abovementioned lightweight aggregates.

The lightweight aggregates used were characterized in order to determine their bulk density and granular dimensions [32,33,43]. Natural cork granulates had a diameter of 4–5 mm and a bulk density of 70 kg/m^3, and they were manufactured by Corticeira Amorim, Portugal. The expanded clay aggregate showed dimensions of 0/2 mm and a bulk density of 490 kg/m^3, and it was produced by Argex. The particle size distributions of the aggregates used in this work are represented in Figure 1.

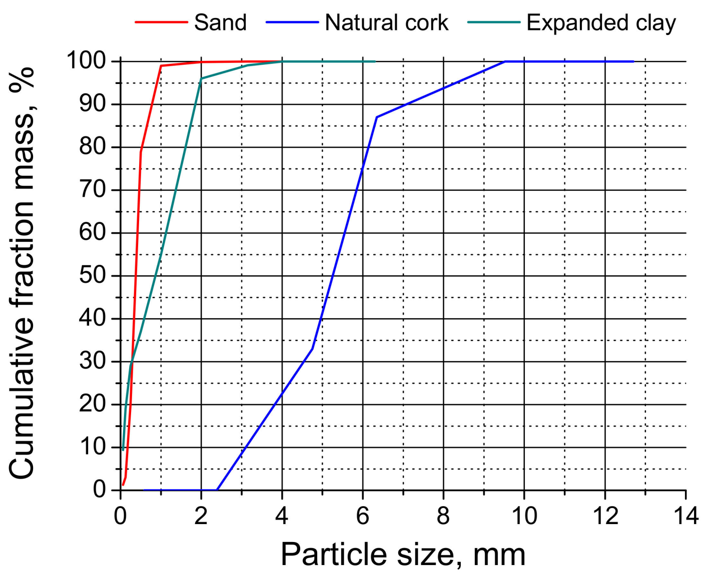

Figure 1. Particle size distribution of the aggregates used.

Prismatic and cylindrical samples were prepared. The dimensions of the first type were 4 cm × 4 cm × 16 cm, while those of the cylindrical specimens were 5 cm in height and 10 cm in diameter. All of them were demolded after the first 24 h since the setting. At that moment, the samples were kept immersed in tap water at a controlled temperature (20 ± 2 °C) up to the testing ages, which were 28 days and 1 year.

2.2. Mercury Intrusion Porosimetry

Mercury intrusion porosimetry is commonly used to characterize the pore network of different materials [44–49]. In the current research, the porosimetry test was performed with a porosimeter model Poremaster-60 GT, of the manufacturer Quantachrome Instruments. The specimens were dried at 50 °C at 48 h prior to the test. In the current research, the total porosity and pore size distributions of the mortars were analyzed. The distributions of the pores by sizes of the samples were represented using the cumulative volume of pores in percentage versus pore size curves. Two tests were carried out for each mortar series at the ages of 28 days and 1 year, on pieces taken from 4 cm × 4 cm × 16 cm prisms.

2.3. Forced Migration Test

The forced chloride migration test was carried out following the NT Build 492 standard [50]. The non-steady-state chloride migration coefficient (D_{NTB}) is obtained from the test. Three different cylinders of a 5 cm height and 10 cm diameter were tested for each mortar series at 28 days and 1 year.

2.4. Mechanical Strengths

Both the compressive and flexural strengths were obtained following the steps established by the European standard EN 1015-11 [51]. Three prismatic specimens were tested for each mortar series at 28 days and 1 year.

3. Results and Discussion

3.1. Mercury Intrusion Porosimetry

The total porosity results are depicted in Figure 2. The study of this parameter is relevant because the pores are the main way for the ingress of aggressive substances into cement-based materials. Then, the determination of the total volume of pores could be useful, especially when those materials could be used in marine works. In addition, the characterization of the total porosity is also interesting because it also provides information about the suitability of the material in relation to other engineering properties, such as thermal and acoustic properties.

The reference series presented smaller values of porosity than mortars containing natural cork and expanded clay. This could be related to the greater porosity of lightweight aggregates [52] compared with sand, which could influence the global solid fraction of the mortar, as a composite material. This parameter was very similar for the reference series at both ages, independently of the cement used, although a scarce reduction in the porosity was observed between 28 days and 1 year. This would be compatible with the results obtained by other authors [53,54], which reported that several active additions, and in particular fly ash, would not produce a noteworthy lessening of the porosity in cementitious materials, despite providing other benefits in their microstructure and properties. The small decrease with time in the total porosity could be related to the development of slag and clinker hydration [25,55] and the pozzolanic reactions of fly ash [12,24], which would lead to the formation of new solids, closing the microstructure, and thus reducing the porosity of the mortars.

With regard to the natural cork series, at 28 days, this parameter was smaller for the 1NCK and 2NCK series in comparison with the 3NCK and 4NCK series. This could be a consequence of the clinker hydration development [55,56], which would have more notable effects in the total porosity at initial ages compared with slag hydration [25,57] and the pozzolanic reactions of fly ash [12,57]. Moreover, the filler effect of the limestone addition [30,31,58] could also have an influence, and particularly in 2NCK mortars. This was more remarkable in the mortars with natural cork, due to their greater porosity, than the reference series. The reduction in the total porosity with time was more noticeable for the 1NCK and 3NCK series, showing at 1 year, lower porosities than those obtained for the 2NCK and 4NCK series. This would have a relation with the effects in the long term of the previously explained slag and clinker hydration [25,55] and fly ash pozzolanic reactions [12,24], which would produce a reduction in the pore volume in the 1NCK, 3NCK and 4NCK samples, as previously explained. In the case of 2NCK mortars, their content of clinker was lower, and limestone is an inert addition [29], which lacks reactivity, so its influence at later ages in the pore network would be less significant. According to several authors [53,54], the addition of fly ash would not produce a high porosity decrease with time, as already indicated, so the results of the 4NCK series would agree with them.

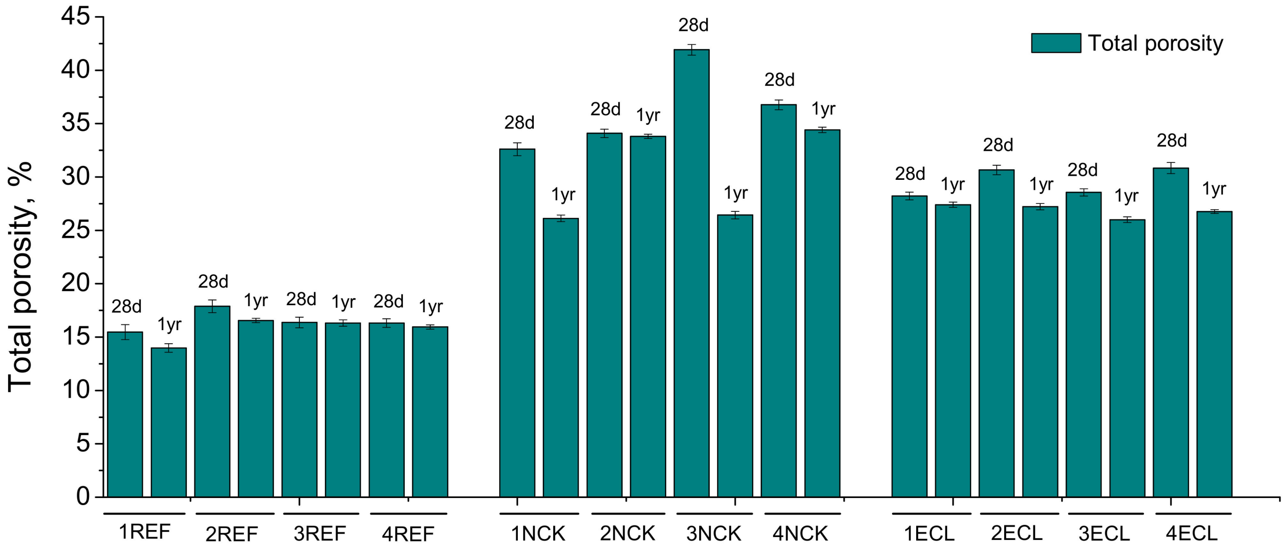

Figure 2. Total porosities obtained for the tested mortars.

Regarding the mortars with expanded clay, at 28 days, the porosity was slightly smaller for the 1ECL and 3ECL series than that observed for the 2ECL and 4ECL series, being higher for this last series with fly ash. This can be explained in relation to the hydration of clinker [25,55,56] and the hydration of slag [25,57], the effects of which were more noteworthy in the short-term total porosity of the expanded clay mortars than those due to the filler effect of the limestone addition [30,31,58] and fly ash pozzolanic activity [12,24,57]. This parameter decreased with time for all series with expanded clay, showing for the 3ECL and 4ECL mortars, the lowest values of the total porosity at one hardening year. Similar trends were observed for the reference and natural cork mortars, which were mainly associated with the influence at later maturation times of the new solid phases formation as products of the hydration reactions of the clinker and slag components [25,55], and the pozzolanic reactions of fly ash [12,24].

Comparing the results obtained for the series with natural cork and expanded clay in the long term, it is noteworthy to underline the similar total porosities noted for those made with ordinary Portland cement (1NCK and 1ECL mortars) and for those that incorporated slag (3NCK and 3ECL series). However, when cements with limestone and fly ash were used, this parameter was lower for mortars with expanded clay (2ECL and 4ECL series) compared with the specimens with natural cork (2NCK and 4NCK series).

The pore size distributions for the reference series can be observed in Figure 3. At 28 days, a higher percentage of pores with sizes under 100 nm was observed for the 3REF mortars, representing approximately 60% of the total pore volume. This result could be related to the slag hydration, already described, which could produce the formation of solids, thus reducing the pore sizes, and it is in agreement with the results of other researchers [57,59,60].

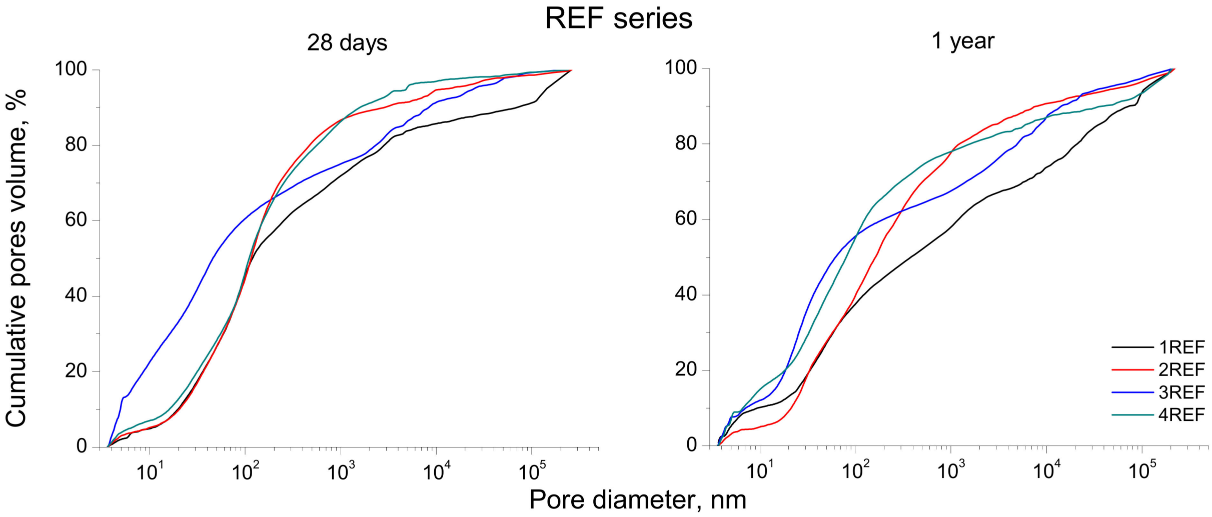

Figure 3. Cumulative volume of pores in percentage versus pore size curves for reference series.

The pore size distributions at that age were very similar for the 2REF and 4REF series, showing lower cumulative percentages of pores with sizes under 100 nm compared with the 3REF mortars, although they had around 90% of pores volume with diameters less than 10 µm. On the one hand, the results of the 2REF series could be justified regarding the filler effect of the limestone addition [30,31,58], which also contributed to the increase in the proportion of finer pores, although its lack of activity [29] would entail the lower pore refinement of these 2REF series in comparison with those with slag. Additionally, the less refined microstructure in the short term for series with fly ash than that observed for 3REF mortars would be a consequence of the delay in the development of the pozzolanic reactions of fly ash in comparison with slag hydration [12,61]. Slag can directly react with setting water [25,55], and fly ash needs enough portlandite produced along the hydration of clinker in order to start its pozzolanic reactions [12]; thus, more time is generally needed for observing the positive influence of fly ash in the pore network than for slag. For the 1REF series, at 28 days, the pores distribution curve until 100 nm was very similar to those noted for the 2REF and 4REF series, although for pore diameters greater than the abovementioned value, the growth of the curve of the 1REF mortars slowed down, which could indicate a lower microstructure refinement, which is in agreement with other works [53,55,62].

At one hardening year, a rise in the proportion of fine pores was observed overall for all the reference series in comparison with the pore size distributions observed at 28 days, which would be a consequence of the abovementioned effects at later ages of hydration [59,60,63] and pozzolanic reactions [12,24]. This increase was specifically notable for specimens with fly ash (4REF series). In the long term, the highest percentage of pores under 100 nm was noted for mortars with slag and fly ash (3REF and 4REF series); however, the 4REF mortars presented greater percentages of pores smaller than 1 µm, which would suggest that they have a more refined microstructure compared with the 3REF specimens. This would indicate the best performance of active additions at later ages [53], and in particular fly ash, whose pozzolanic activity notably improved the pore structure with time [12,24]. The pore size distribution curve hardly changed from 28 days to 1 year for the reference mortars with limestone, showing a less refined microstructure than the 3REF and 4REF series, which is in keeping with other authors [53–55]. In the case of the 1REF mortars, an increase in the percentage of pores lower than 20 nm was observed at 1 year compared with 28 days, which was probably produced by the formation of solids as products of clinker hydration [55,56], although its pore structure was the least refined of the reference series in the long term.

The pore size distributions observed for the series with natural cork are represented in Figure 4. The curve cumulative pore volumes versus pore diameters at 28 days were very similar for all these series, which would suggest that the short-term pore size distributions were mainly influenced by the natural cork aggregate, having the binder less influence. Nevertheless, at that age, the 2NCK mortars showed slightly higher percentages of pores lower than 100 nm, and this could be justified in relation to the filler effect of limestone [30,31,58], which would be more noticeable in the pore structure of natural cork mortars at the initial ages.

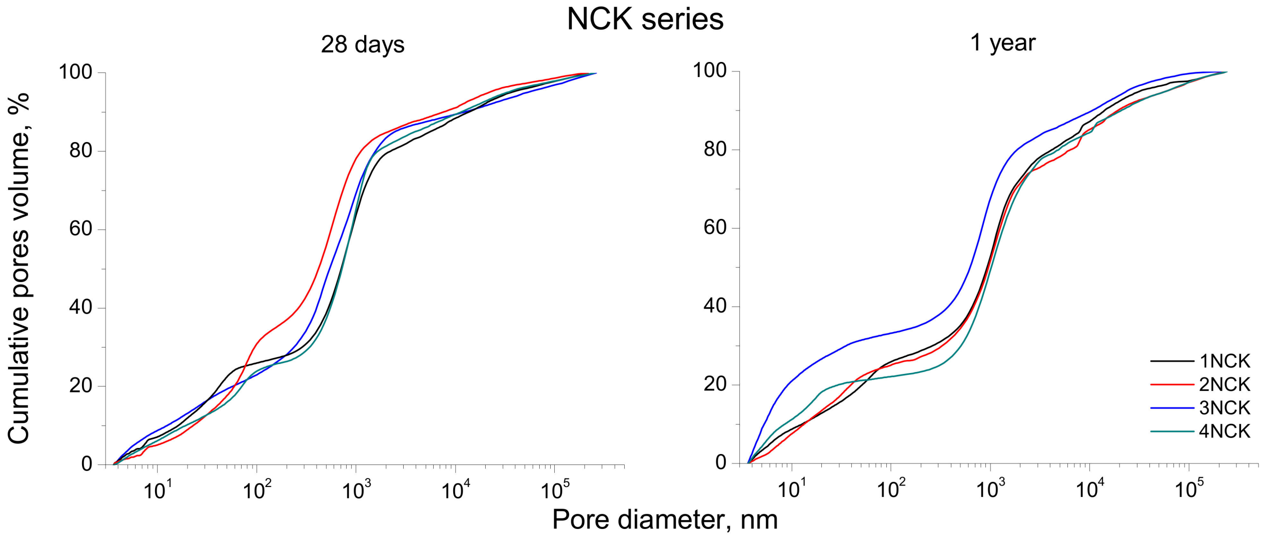

Figure 4. Cumulative volumes of pores in percentage versus pore size curves for mortars with natural cork.

At 1 year, it is noteworthy to underline that the pore structure of the natural cork series with slag (3NCK series) presented a higher refinement in comparison with the other series with this aggregate. This series contained higher proportions of pores under 100 nm and 1 µm, which were probably caused by the abovementioned influence of the hydration of the slag at later ages [25,55,57]. For the 4NCK mortars, between 28 days and 1 year, it has been observed an increase in the percentage of pores lower than 30 nm, which would show the gradual closure of the pore network because of the pozzolanic activity of fly ash [12,24], which was also noticeable when the natural cork aggregate was used. Finally, the long-term pore size distribution was very similar for the 1NCK and 2NCK series. In this result, the lack of activity of the limestone [29] could have had an influence, thus not providing any advantage in the long-term distributions of the pores by size of the natural cork mortars, compared with ordinary Portland cement with no additions, which is contrary to what happened with the active additions of fly ash and slag.

The cumulative volume of pores in percentage versus pore size curves noted for mortars with expanded clay can be observed in Figure 5. The pore network was more refined in the specimens with slag (3ECL series) at 28 days, as suggested by the greater proportion of pores with diameters less than 100 nm compared with the other series with expanded clay. This could be caused by the hydraulic activity of slag [25,55], explained before, which was more remarkable in the short term for the expanded clay series than for those with natural cork. In addition, at that age, smaller differences between the 1ECL, 2ECL and 4ECL mortars in the cumulative volume of pores in percentage versus pore size curves were noted, although the greater proportion of pores under 30 nm observed for the 4ECL mortars was noticeable, which would show the incipient effects of the pozzolanic reactions of fly ash [12,24] in the microstructure.

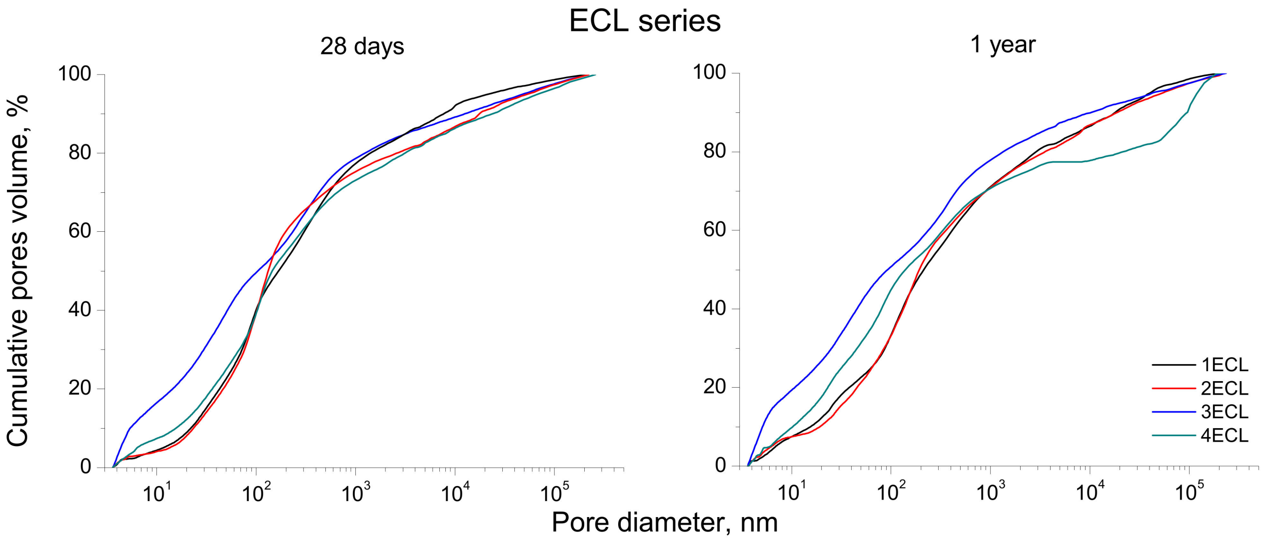

Figure 5. Cumulative volumes of pores in percentage versus pore size curves for series with expanded clay.

At one hardening year, the specimens of the 3ECL series still showed the highest microstructure refinement, with greater percentages of pores smaller than 10 nm, 100 nm, 1 µm and 10 µm compared with the other series with expanded clay. Furthermore, from 28 days to 1 year, a noticeable increase in the percentage of fine pores was observed for these samples with slag. A similar evolution was also noted for the natural cork mortars, as previously explained, so this result obtained for those mortars with expanded clay would confirm the adequate performance in the long-term microstructure development of combining a binder with a high content of slag with the lightweight aggregates studied in this work, being mainly associated with the abovementioned slag hydration [25,55].

This was likewise noted for the fly ash series, which showed a notable growth in the percentages of pores under 100 nm between both testing ages. This tendency again coincided with that observed for the natural cork mortars with this addition, although it was more remarkable when expanded clay was used, and it may be a consequence of the overall reduction in the pore sizes caused by the new solid phases formed in the pozzolanic reactions of fly ash [12,24], already discussed. Regarding the 1ECL and 2ECL series, their cumulative pores volume versus pore diameter curves were relatively similar at 1 year, and their pore structure was overall less refined than that obtained for the mortars with fly ash and slag, with a lower proportion of pores finer than 100 nm. This was also noted for natural cork mortars, in which it was linked to the long-term effects of active additions [12,24,25,55], in comparison with ordinary Portland cement, and particularly with the inert character of limestone addition [29].

Finally, in comparing the different aggregates used, it was observed that the series with natural cork presented a high proportion of coarser pores, and particularly in the range between 400 nm and 2 µm, which would indicate the high rise in their cumulative pore volume versus pore size curves in that range of pores. This was observed independently of the binders and at both testing ages, and it could be related to the effect of the natural cork aggregate. For the reference mortars and those that incorporated expanded clay, this growth of the curve up to a size of 2 µm was more progressive, and therefore, they showed greater percentages of finer pores.

3.2. Non-Steady-State Chloride Migration Coefficient

The results of the non-steady-state chloride migration coefficient are shown in Figure 6. The study of this parameter is important because chloride ions are the most important aggressive agent that can produce the development of corrosion pathologies in reinforced concrete elements. Then, it is important to assess the effects of the natural cork aggregate in combination with eco-friendly cements in the chloride ingress resistance of the analyzed materials.

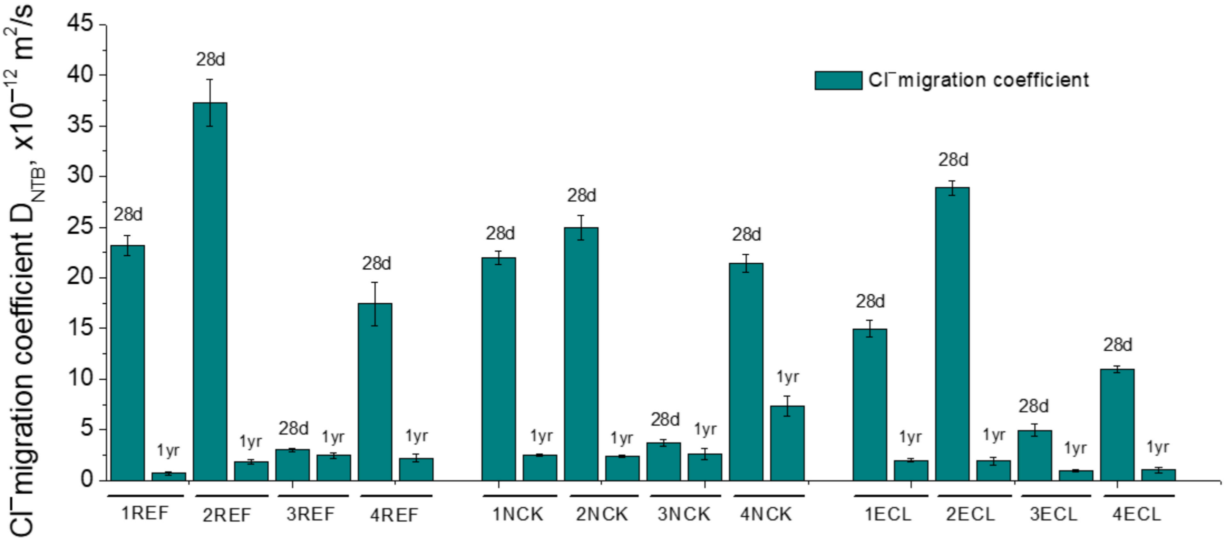

Figure 6. Non-steady-state chloride migration coefficient results for the series tested.

As can be observed in Figure 6, the binder used in the mortars played an important role in the results of this parameter. In general, relatively low migration coefficients were obtained in mortars with lightweight aggregates compared with the reference series. With regard to these reference mortars, the highest migration coefficient at 28 days was observed for the 2REF series, followed by the 1REF series, while it was smaller for the mortars with fly ash and slag, being lower for the 3REF mortars compared with the 4REF mortars. At 1 year, the migration coefficient was very similar for all the reference series, independently of the binder used, showing the 1REF mortars scarce lower values.

For the natural cork mortars, this coefficient was higher for the 1NCK and 2NCK series at 28 days in comparison with the 4NCK and 3NCK series, and the last one with slag presented the lowest values in the short term. After 1 year, the migration coefficient was higher for the 4NCK mortars, whereas small differences were noted for the 1NCK, 2NCK and 3NCK series. It is interesting to highlight that, at both testing ages, the series with natural cork showed similar or slightly higher migration coefficients than the reference mortars for each one of the binders used, with the exception of the 4NCK series, which had greater values of this parameter than the 4REF series.

In relation to the specimens with expanded clay, similar trends were observed in comparison with those described for the natural cork and reference series. The migration coefficient at 28 days was higher for the 2ECL specimens, followed by the 1ECL specimens, while their values were more reduced for the 3ECL and 4ECL series, showing the mortars with slag the lowest coefficient. Scarce differences between the series with expanded clay were noted at 1 year, being slightly lower the migration coefficient for the 3ECL and 4ECL mortars compared with the 1ECL and 2ECL mortars.

Regarding the comparison between the coefficients noted for the natural cork and expanded clay mortars, it is relevant to point out that no substantial differences were detected between them for each binder. In the short term, the 1ECL and 4ECL series presented smaller migration coefficients than the 1NCK and 4NCK series, while this parameter was greater for the 2ECL and 3ECL mortars when compared with the 2NCK and 3ECK mortars. Additionally, in the long term, this coefficient was slightly lower for the series with expanded clay than for those with natural cork, with the greatest difference observed between the 4NCK and 4ECL mortars. These small differences at one hardening year could be related to the less refined microstructure of the natural cork mortars compared with the expanded clay and reference series, especially due to the higher proportion of coarser pores for the natural cork specimens, previously described in the results of mercury intrusion porosimetry. Despite this, the adequate long-term performance of the studied mortars with natural cork in relation to chloride ingress resistance is remarkable, and particularly in comparison with a standardized and commonly used lightweight aggregate, such as expanded clay.

The lower chloride migration coefficients at 28 days obtained for the series with slag and fly ash, independently of the aggregate used, could be a consequence of the greater pore refinement caused in the short term by these active additions [53,64–67], already analyzed in the microstructural results. Moreover, this result could also be influenced by the higher binding capacity of cements with fly ash and slag [68], due to the high content of calcium aluminates provided by these additions, which would hamper the movement of the aggressive chloride ions within the pore network of the mortars, thus decreasing their migration coefficients.

The reduction in this coefficient with time noted for all series tested would agree with the refinement with age of the pore structure observed with mercury intrusion porosimetry, produced by the development of the hydration reactions of slag and clinker hydration and the pozzolanic reactions of fly ash, as previously explained. Lastly, after a very long maturation period, a high degree of the development of these reactions [12,24,25,55] would be expected, which would result in the smaller differences in the migration coefficient observed after 1 year.

3.3. Mechanical Strengths

The results of the compressive and flexural strength are represented in Figures 7 and 8, respectively. As can be observed, both parameters were greatly affected by the aggregate used. The mortars containing natural cork had much lower strengths than the reference series and those prepared with expanded clay, for all the studied binders. This result could be expected, according to the results reported in the literature [43,69] regarding the mechanical performance of cementitious materials with cork aggregates. Additionally, the higher total porosity and greater proportion of pores with diameters greater than 100 nm observed for the natural cork mortars compared with reference ones and those with expanded clay, according to the porosimetry results already discussed, could also have influenced the reduction in the mechanical behavior of the natural cork series. Furthermore, in general, the expanded clay mortars also showed lower mechanical strength than the reference series. This result would be also in agreement with the total porosity results because it was higher for the expanded clay series than for the reference series. The low mechanical strengths observed for the natural cork mortars, combined with their adequate chloride ingress resistance, would recommend their possible use as non-structural material in civil engineering works exposed to maritime environments.

With regard to the reference series, at 28 days, the compressive strength was very similar for the 1REF, 2REF and 4REF series, and it was higher for the 3REF series. The flexural strength at that age was greater for the specimens with active additions (3REF and 4REF series) compared with the 1REF and 2REF mortars. From 28 days to 1 year, an increase in the compressive and flexural strengths was generally observed, showing the 4REF mortars the highest values of both parameters in the long term, followed by the 1REF and 3REF series. The lowest strengths at 1 year were noted for the 2REF series.

Regarding the natural cork mortars, low strengths were observed along the studied time period, regardless of the binder used, below 6 MPa for the compressive strength, and under 2 MPa for the flexural strength. The compressive strength scarcely changed with time for the natural cork series, being relatively similar for all of them, although at 1 year, its greatest values were obtained for the 4NCK series, and the lowest corresponded to the 2NCK series. The rise with time was more noticeable for the flexural strength, and particularly for cork mortars containing additions (2NCK, 3NCK and 4NCK series). In the long term, the highest flexural strength of the natural cork series was noted for the 4NCK mortars, which were closely followed by the 3NCK and 1NCK mortars, whereas the lowest flexural strength was noted for the 2NCK samples.

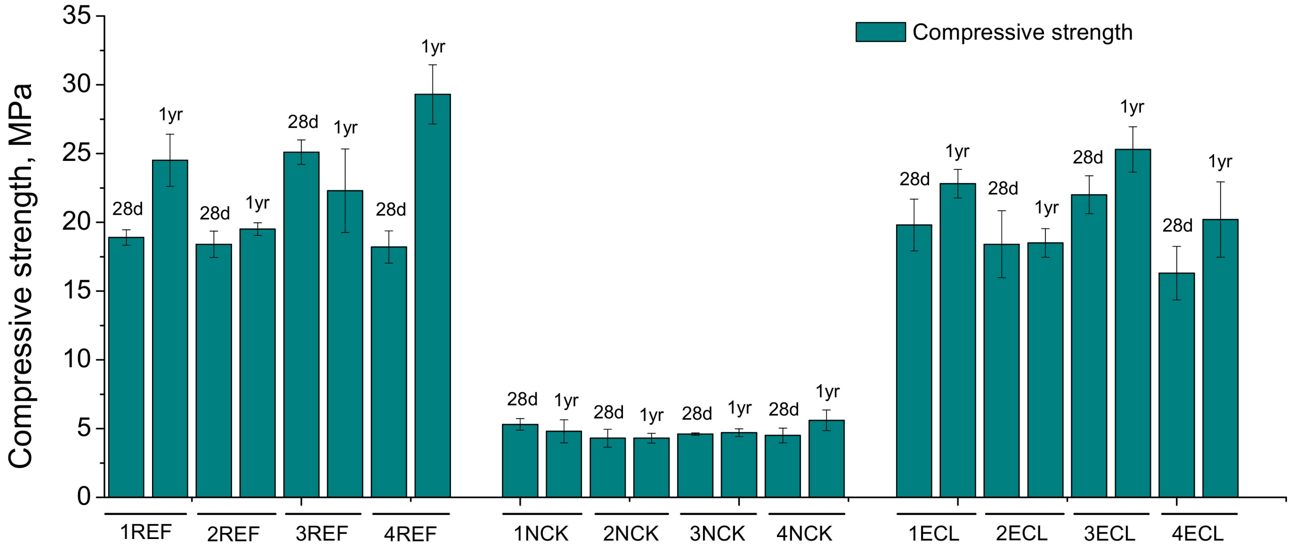

Figure 7. Compressive strength results for the analyzed series.

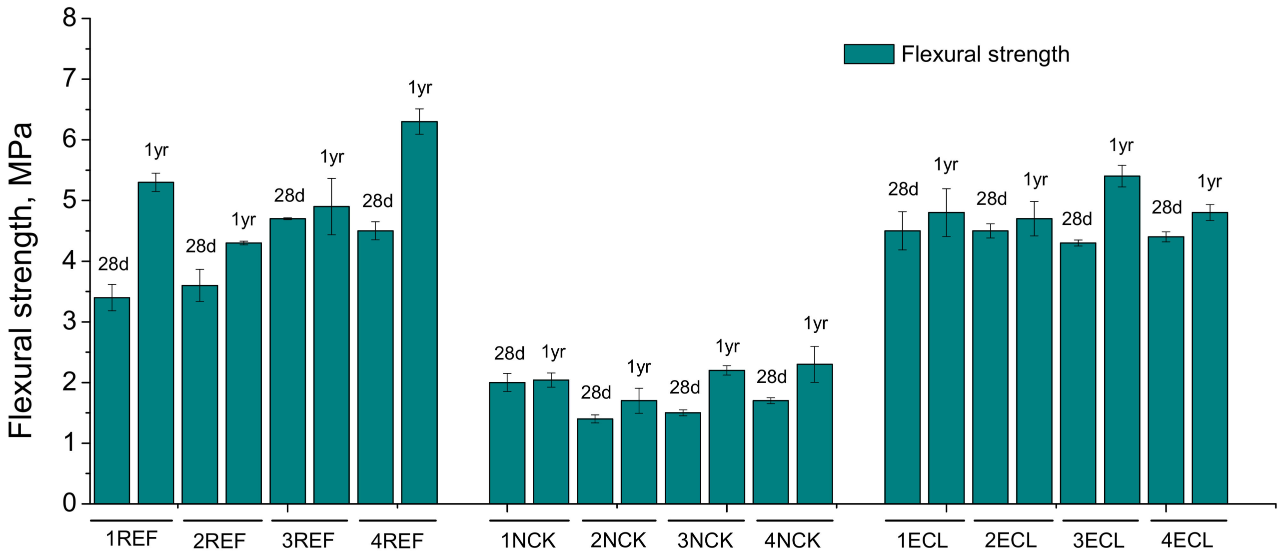

Figure 8. Flexural strength results for the analyzed series.

With respect to the series with expanded clay, the flexural strength at 28 days was very similar for all of them, while the compressive strength was scarcely higher when binder with slag was used (3ECL series), followed by the 1ECL and 2ECL mortars, showing the 4ECL series the lowest values of this strength in the short term. For most of the series with expanded clay, an increase in both strengths with the hardening time was also observed. The greatest compressive strength at 1 year was obtained for the 3ECL series, followed by the 1ECL and 4ECL mortars, whereas the smallest values were noted for the 2ECL series. The long-term results of the flexural strength were similar to those described for the compressive strength, although with lower differences between the different binders.

The improvement with time in the mechanical performance of the majority of the studied series, independently of the binder and aggregate used, could be caused by the solid-phase formation due to the progressive development of slag and clinker hydration [25,55] and fly ash pozzolanic reactions [12,24], already explained. This result would be in keeping with the lessening with time of the total porosity and chloride migration coefficient, as well as with the pore refinement noted for most of the analyzed series, previously discussed. The lower strengths shown overall after 1 year by binders with limestone, for all the aggregates studied, could be caused by the abovementioned lack of the pozzolanic or hydraulic activity of this addition [29], which would result in a smaller improvement in the long-term mechanical performance. The better performance of the binders with slag at 28 days could be due to its hydraulic activity [25,55], which makes their beneficial effects noticeable from the early ages compared with other additions with pozzolanic activity, such as fly ash [70]. Finally, as was mentioned in the microstructure characterization, the effects of fly ash are generally delayed [12,61] because sufficient portlandite is needed for the development of the reactions of this addition [12]. This product is formed during clinker hydration. For this reason, the mortars with fly ash showed flexural and compressive strengths at 1 year that were comparable to those obtained for the mortars with slag for the same aggregate, and they were even higher, in some cases, than the values noted for ordinary Portland cement mortars. The development of a relatively high strength over the very long term in cement-based materials that incorporate fly ash has also been reported by other authors [24,70].

4. Conclusions

The main conclusions obtained from this work are summarized as follows:

1. The lowest total porosity in the long term was generally obtained for reference mortars without lightweight aggregates. The total porosity differences between the series with natural cork and with expanded clay were not high, depending on the binder used.
2. The series with natural cork showed a lower refinement of the microstructure compared with the reference mortars and those with expanded clay. Furthermore, it was observed that the pore size distributions of the natural cork mortars in the short term were mainly affected by the aggregate, with the binder having less influence.
3. A decrease in the total porosity and rise in the refinement of pores with time were observed for most of the mortar series studied, independently of the aggregate used. This evolution of the microstructure would improve the other parameters analyzed, resulting in a lessening with the hardening time of the non-steady-state chloride migration coefficient and a rise in the mechanical strengths.
4. The binder used in the mortars played an important role in the results of the nonsteady-state chloride migration coefficient, especially at 28 days. It is remarkable that the studied mortars with natural cork showed similar or slightly higher migration coefficients than the reference and expanded clay mortars for each one of the binders studied at 1 year.
5. Both the compressive and flexural strengths were greatly influenced by the type of aggregate. The mortars with natural cork had lower mechanical strengths than the reference series and those prepared with expanded clay, for all the studied binders.
6. For each one of the aggregates studied, at the initial ages, the mortars with slag overall showed higher microstructure refinement, a lower chloride migration coefficient and greater compressive strength compared with the other studied binders.
7. In view of the results obtained, it is noteworthy to underline the adequate long-term performance of the studied mortars with natural cork in relation to chloride ingress resistance, and in comparison with a standardized and commonly used lightweight aggregate, such as expanded clay. This adequate chloride ingress resistance, combined with the low mechanical strengths produced by the incorporation of natural cork, would recommend the possible use of this lightweight aggregate in non-structural cement-based materials for civil engineering works exposed to maritime environments. Furthermore, the use of binders with active additions, and especially blast-furnace slag, overall improved the behavior of mortars with natural cork at later ages.

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