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*Research Article*

# **A comparative study of expanded and raw cork waste composite materials**

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## **1. Introduction**

The use of cork waste in conjunction with various binders to achieve composite materials is both technically and economically advantageous for building. Indeed, these composite materials are still gaining increasing interest in recent years [1]. The approaches developed for the reuse of waste materials from various plant origins and various sources to reduce environmental pollution and thus protect public health. Wood-based composite materials combine mechanical performance and insulation while being ecologically better than traditional insulation materials.

Cork exhibits very interesting properties from low density, and elasticity to low thermal conductivity, sound insulation, and impermeability [2]. The processing industries generate considerable quantities of cork waste [3]. These by-products include raw cork granules and reproduced cork. The chemical compositions of these two corks are very similar: 0.9% ash, 9.2% extractable, 37.8% suberin, and 28.5% lignin on average [4].

Several studies [5-9] show that the reuse of cork associated with cement is very effective for thermal insulation. However, the incorporation of the volume of cork residues increases the volume of air or voids, which is accompanied by a decrease in mechanical performance. This mixture is also accompanied by a low bulk density, which exhibits a non-negligible advantage for structures [10]. Menor et al. [11] also found that the introduction of raw cork and bark into a cementitious matrix increases porosity. Tedjditi et al. [6] reported the

same results on a combination of virgin cork and bark cork. The thermal conductivity of the composites was indeed improved. In another study, cork is mixed with gypsum to make a new composite [12]. The acoustic absorption coefficient at high frequencies (7000Hz) of gypsum-cork composites was examined. The resulting composite is soundproof but reflective and is identical to the gypsum board. Matos et al. [13] demonstrated the use of cork powder as a fine alternative in self-compacting concrete mixes. This allowed researchers to assess the novel material's strength (C30/37) and durability. The results reveal that the material is strong and durable for common applications.

Some studies have investigated the impact of cork powder and superplasticizer on the mechanical properties of cement composites [14], while others have explored the use of hybrid cork–polymer composites containing sisal fibre [15] or composites prepared with a metakaolin-based geopolymer and cork waste [16]. Other studies have used phenolic aerogels and cellulose plugs to prepare nanocomposites [ 17]. Parthasarathy et al. [18] investigated the mechanical properties of cork-based composites with cork contents up to 30% and fabricated using the hand-laying technique. The results revealed that composites containing between 20% and 25% cork exhibited the most promising mechanical properties, suggesting an optimal compromise between cork content and composite performance. Svetlana et al. [19] studied composites based on virgin cork (C) and plaster (P) with varying C/P ratios. The results showed that these composites can be used as insulation materials, with thermal conductivity ranging from 0.168 to 0.460 W/(m. K), making them suitable for non-structural building applications. Alves et al. [20] conducted a comparative study of the fire resistance of geopolymer composites fabricated with different materials, including cork, expanded vermiculite, and lightweight expanded clay. The results of their research reveal that composites containing expanded vermiculite and clay exhibit the best fire resistance.

The influence of fly ash substituted for cement on the physical-mechanical and thermal characteristics of cement-cork composites was examined by Novais et al. [21] and Borges et al. [22] lining mortars based on cement and expanded cork aggregates as a sand substitute, as well as a material with various fly ash additives, have been developed. The mechanical characteristics are unaffected by the addition of fly ash. Also achieved were very low density, low conductivity, and good sound absorption [21].

The blast furnace slag was incorporated as an addition to the cementitious composite, and the authors concluded that the slag reduced the thermal conductivity by 50% [23]. Similar conclusions were obtained by Kim et al. [24]. The incorporation of 50% slag reduced the shrinkage by up to 18% [23]. Liu et al. [7] showed that the use of cork in cement mortar can reduce its density, thus increasing its thermal insulation capacity, while the increase in moisture increases the thermal conductivity of the mortar. Other work has been based on two binders. Indeed, they were cement and hydrated lime. Meanwhile, the strength, density, and thermal conductivity decrease with the introduction of cork [25]. Mounir et al. [26] mixed clay and cork to increase heat conductivity, which is twice as thick as the clay-cork composite and lighter than clay alone. A cork and white cement mixture were developed in another experiment. When compared to white cement alone, the results show a 47% increase in lightness, a 76% increase in insulating qualities, and a 40% reduction in thermal inertia parameters [9].

The authors [27] investigated the effect of cork granule size on the thermal conductivity of composites in the literature. The results demonstrated unequivocally that the size of the granules does not affect the thermal conductivity of compositions derived with constant granular mass fractions. Panesar and Shindman [28] studied the influence of cork size on mechanical characteristics and found that the finest cork sizes had the best mechanical properties. Karade et al. [29] reached the same finding with smaller cork diameters.

In contrast to the literature cited, there is a lack of experimentation on drying shrinkage and sound insulation. The influence of the expanded cork particle content on compressive strength, tensile strength, thermal conductivity, sound absorption, and shrinkage was investigated. To maximize recycling efficiency, high content of cork waste and a wide range of cork sizes were used. In addition, a comparison with raw cork was also presented.

#### **2. Materials and mix proportions**

#### **2.1 Cement**

An ordinary Portland cement type CEMII 42.5 was used, and the chemical analyses were carried out by energy dispersive X-ray spectroscopy (EDX). The results are shown in Table 1. The density of the cement used was 3.21 kg/m<sup>3</sup> its specific surface area was  $308.5 \text{m}^2/\text{g}$  and its compressive strength at 28 days was 42.66MPa.

Table 1. Chemical properties of the used cement (%)

SiO <sub>2</sub>					$AL_2O_3$ Fe <sub>2</sub> O <sub>3</sub> CaO MgO K <sub>2</sub> O Na <sub>2</sub> O SO <sub>3</sub> CaO Insoluble	Loss on ignition
18.78	49	3.7 63.06 0.88 0.47 0.21 2.43 0.85			0.74	3.98

#### **2.2 Waste Cork**

The waste cork used was sourced from a site in the eastern region of Algeria. Industries in the region produce approximately 1,900 tons of cork granules and 5,600 tons of cork powder per year [3]. Two types of waste cork were used: raw and heat-treated (expanded). All the waste cork was thoroughly cleaned of impurities prior to use. The particle size of the cork was analyzed through granulometric screening in accordance with NF B57-011, as shown in Figure 1.



Fig. 1. Granulometric curves of expanded and raw cork

Expanded cork waste endorses a higher bulk density than raw cork (Table 2). This density is significantly lower than that of many lightweight aggregates currently utilized in lightweight concrete. Therefore, we can produce this kind of concrete considering its interesting physical properties and thermal insulation.





The cork has a density much lower than most aggregates used for lightweight concrete (about 300  $kg/m<sup>3</sup>$ ) and is comparable with that of group 1 for both perlite and vermiculite according to the limits of the ASTM C332 standards [30]. Expanded waste cork has slightly higher hygroscopic moisture content than raw cork. However, raw cork favourites high water absorption after 24 hours of immersion (about 154%). Thus, the water absorption rate of the cork granules was very high due to their hydrophilic nature.The procedure begins with cleaning the cork to remove impurities. The cork granules are then mixed dry with the cement and water is added gradually to the mixer. A range of granules is used, with all composites produced using a single fraction with a size of 2-6.3 mm. Three compositions (1:1, 1:2, and 1:3) are proposed for the best volume ratio of cement to cork, and five different water-cement ratios (W/C: 0.30, 0.35, 0.40, 0.45, and 0.5) are studied (Table 3). The experiment uses two types of cork: expanded and raw. The test specimens are cast and compacted on a laboratory vibration table and then cured for 14 days in a chamber at 20  $\pm$  2 °C and 100% relative humidity. Afterward, they undergo air curing at 20  $\pm$  2 °C and 65  $\pm$  5% relative humidity until they reach 28 days of age. Hardened density, compressive, and flexural strength tests are conducted with different E/C ratios, while shrinkage, thermal conductivity, and sound absorption tests are performed with a W/C ratio of 0.5.

Table 3. Mixtures suggested



# **3. Test Procedures**

# **3.1 Hardened Density, Compressive and Flexural Strength Tests**

Hardened density tests were conducted on 40x40x160 mm<sup>3</sup> prismatic specimens after 28 days of age. Flexural strength was measured on 40x40x160 mm<sup>3</sup> prismatic specimens using a three-point test in accordance with NF EN 196-1 after 28 days, by applying a constant-speed load until failure. The compressive strength was determined by applying a load to failure, using the two specimens obtained. The average of three specimens per batch is used to determine the compressive and flexural strength.

## **3.2. Shrinkage Test**

The drying shrinkage tests were conducted on  $40x40x160$  mm<sup>3</sup> prismatic specimens in accordance with NF P15-433. The specimens were removed from the mold after 24 hours. They were stored in a climate chamber at 20  $\pm$  2 °C and 60  $\pm$  5% relative humidity and the measurements were taken up to 90 days of age. The results of three samples were averaged to provide the final results.

## **3.3 Thermal Conductivity Tests**

The thermal conductivity  $(\lambda)$  was measured using a CT-Meter, in accordance with the NF E993-15 standard. The tests were performed on 200x100x40 mm<sup>3</sup> specimens after 28 days of age. Prior to testing, the specimens were kept in an oven at 105°C for 24 hours and in a desiccator for one day. The determination of  $(\lambda)$  was conducted between two samples and the average of the three results obtained was used as the final value.

## **3.4 Sound Absorption**

There have been few investigations on the sound absorption of cement-cork composites. The sound absorption coefficient  $(\alpha)$  is obtained by the dual microphone model 4206, according to EN ISO 10534-22 (see figure 2). This apparatus can be used at low (90 to 1800 Hz) and high frequencies (800 to 6500 Hz) for small samples. The tests were performed on cylindrical specimens of 29- and 100-mm diameter and 50 mm height at the age of 28 days. Six frequencies of 125 Hz, 250 Hz, 500 Hz, 1000 Hz, 2000 Hz, and 4000 Hz were used. The sound absorption coefficient is the ratio of the absorbed sound energy to the incident sound energy. The value of  $(\alpha)$  is between 0 and 1. It tends towards 0 for a smooth, hard, and heavy wall, and is therefore very reflective. It is equal to 1 for an open window.



Fig. 2. Equipment used to measure α (Double Microphone System Model 4206)

# **4. Results and Discussion**

## **4.1 Hardened Density**

Figure 3 illustrates the important effect of cork granules on density. Indeed, with the increase in cork volume, the density decreases. It is greater when the W/C ratio is equal to 0.30. For one volume of cork, the density of the composite with expanded cork granules is 1572 kg/m<sup>3</sup>; it is 776 kg/m<sup>3</sup> for two volumes of cork, and it becomes  $614 \text{ kg/m}^3$  for three volumes of cork. This effect is explained by the demand for water by cork and by its low density compared to cement, in addition to the very fine cellular structure that characterizes cork. Similar to lightweight aggregates, cork has a very high porosity (85% voids) [31]. This porosity has negative effects on the durability of composites.

The gradual increase in the W/C ratio readjusts the difference in density between the three compositions. This suggests an increase in the water absorption capacity of the composites. The density of an expanded cork composite with a Cement/Cork ratio of 1/1 decreases as the W/C ratio increases (Fig. 2a). On the other hand, in the composite with Cement/Cork ratios of 1/2 and 1/3, the density increases until the W/C ratios reach 0.45 and 0.5 respectively.



Fig. 3. Variation of hardened density with the W/C ratio: (a) expanded cork, (b) raw cork

Compared to the composite with raw cork aggregates, it can be seen that the density of the composite (M50) with expanded cork and  $C/L$  ratio of 1/1 was 1359 kg/m<sup>3</sup>, i.e. about 1.004 times that of the composite with raw cork (Fig. 3.b). However, the composition of M35 with raw cork was  $1581 \text{ kg/m}^3$ , which is about 1.008 times that of the composite with expanded cork. The difference in density between the two composites was not significant. This could be because raw cork aggregates have a higher water absorption capacity than expanded cork. This water will then be available for hydration to continue for a longer period, resulting in a denser structure, better compaction, and a higher density. In addition, the high density of the expanded cork particles was responsible for the reduction in the difference in hardened density. The comparison of Figure 3 (a) and Figure 3 (b) shows that the maximum density of the different composites is obtained for the same W/C ratio, except for the two M30 compositions with the two cork aggregates used in this investigation (C/L:1/1). For all formulations, the density remains relatively low (<1800 kg/m<sup>3</sup>). Therefore, the results obtained for the three composites are consistent with lightweight concrete [32]. The mixture 1/1 with expanded and raw cork could be classified as load-bearing lightweight concrete (1350kg/m3). In contrast, mixtures with Cement/Cork ratios of 2 and 3 are classed as medium and low strength lightweight concrete, respectively [33].

#### **4.2 Compressive and Flexural Strengths**

The compressive strength data of composites with expanded and raw cork aggregates as a function of W/C ratios are presented in Figure 4. As expected, we noticed a decrease in compressive strength with increasing expanded and raw cork and, consequently, a lower cement volume. In addition, the increase in W/C ratio for a Cement/Cork ratio of 1 (C/L:1/1) decreases the compressive strength. However, for higher cement/cork ratios (C/L:1/2 and 1/3), excess water will be partly absorbed by the cork and released at later ages and hence acts as internal curing which enhances long-term hydration and increases the compressive strength. A similar tendency is observed for flexural strength (Figure 4). However, the cork aggregates retard cement hydration [34]. As indicated by [35] the level of hemicellulose is responsible for the increase in setting time and, consequently, the decrease in compressive strength. For example, the compressive strength of composites with expanded cork for a W/C ratio equal to 0.30 with one volume, two volumes, and three replacement volumes of cork was 15.02 MPa, 1.49 MPa, and 0.80 MPa, respectively, corresponding to a decrease in compressive strength of 90.08% and 94.67%. This effect was steadily the case for all the specimens at all the W/C ratios, but with less variation when the W/C ratio increases. This is because cork aggregates appeared as air voids where very small granule particles would not contribute to the strength of the composite. This conclusion is similar to the findings of other authors [6, 25]. This is also due to the high amount of cork used instead of sand [29]. Comparing our results with those of composites with larger aggregates, we can claim, for the same volume of expanded cork, an increase in compressive strength [23]. This is due to the use of CEM II 42.5 cement and the reduction of cork size. Other works using smaller sizes confirm this result [28, 29].



Fig. 4. Variation of compressive strength with W/C ratio: (a) expanded cork, (b) raw cork

Composites with raw cork show similar behavior about compressive strength. However, the use of raw cork causes a decrease in compressive strength compared to cementitious composites with expanded cork. As an example, the compressive strength of M30, M35, M40, M45, and M50 with a  $C/L:1/1$  ratio of composites with expanded cork was 1.28, 1.07, 1.35, 1.43, and 1.33 times higher than composites with raw cork. This superiority in compressive strength was maintained with the increase in the W/C ratio. As noted earlier, the volume of the cork affects the flexural strength (Figure 4). The flexural strength of the M30 expanded cork composite increased from 0.27 MPa to 3.13 MPa depending on the cork volume, a decrease of 91.37%. In contrast, the M30 with raw cork decreased by 88.42 %. When comparing the two composites, the formulations with raw cork were slightly higher than with expanded cork. This is due to the elastic nature of raw cork. However, the expanded or reproduction cork changed processing. For a W/C ratio of 0.3 and a Cement/Cork ratio of 1, mixtures with raw and expanded cork can reach 11.76 MPa and 15.02 MPa, respectively. These strengths do not meet the ASTM C330 criteria for lightweight structural concrete [36], which requires a 28-day compressive strength of 17 MPa and a maximum hardened density of 1850 kg/m<sup>3</sup>. The lowest compressive strength of the mix with a  $C/L: 1/1$  ratio was 7.77 MPa. All these mixtures can be classified as medium-strength lightweight concrete [32]. The compressive strength of these mixtures shows that they can be used as blocks and floor covering. The thicknesses are to be decided on a case-by-case basis by applying the current thermal regulations. The strengths found were higher than those of cement-sand-cellulose fibers and the composite made with cement-soil-sand-durian fibers [34]. The other formulations  $(C/L: 1/2$  and  $1/3$ ) belong to the class of non-structural lightweight concrete (Rc < 7 MPa) [32]. They can be used for horizontal or vertical plate elements to improve thermal comfort. Following the density and compressive strength, only the C/L: 1/3 mixture can be classified as a low-density lightweight concrete. Overall, when compared to the agro-composite materials identified in the literature [37], these composites demonstrated competitive mechanical characteristics.



Fig. 5. Variation of flexural strength with W/C ratio: (a) expanded cork, (b) raw cork

#### **4.3 Correlations Between Flexural Strength and Compressive Strength**

The outcome of a correlation matrix between the flexural strength and compressive strength of the test results is in (Figure 6). Moreover, we have got empirical relationships of the type  $R_f = K$  $(RC)^R$  between compressive and flexural strengths for lightweight concrete [38]. The experimental results of the mechanical resistance are in agreement with this expression. Among these relationships, the one proposed by Bonzel (formula 1), underestimates the flexural strength compared to the composites examined in this study. On the other hand, the formula proposed by Narayanan and Ramamurthy [39] (formula 2) overestimates the flexural strength compared to the composites with expanded cork. However, it remains lower than the composites with raw cork. The results show that it is possible to predict a better flexural strength of composites with expanded and raw cork with equations (3) and (4) with a coefficient of determination of  $R^2 = 0.97$  and 0.99, respectively.



As illustrated in Figure 5, the flexural strength of composites with raw cork particles improves as the compressive strength increases, resulting in increased flexural strength. Therefore, the use of composites containing raw cork will contribute more to the flexural elements. The maximum flexural strength of M35 for the composite with raw cork,  $(C/L: 1/1)$  is 3.2 MPa and M30 is 3.13 MPa for the composite with expanded cork. It is conclusive that all the composites have a compressive strength greater than the flexural strength and the ratio between them Rc/Rf is always greater than 2.



Fig. 6. Correlation between flexural and compressive strength

#### **4.4 Shrinkage**

Figure 7 shows the results of the drying shrinkage of composites with expanded and raw cork for 90 days. The highest shrinkage is that of C/L: 1/3 (M50) with raw cork which was 4647 μm/m at 90 days. Most of the shrinkage started at an early age, as reported in the literature [40, 41]. The shrinkage of the three expanded cork composites C/L: 1/3, C/L: 1/2, and C/L: 1/1 was 8%, 12.41%, and 13.48%, respectively, lower than that of the raw cork composites. This suggests the use of raw cork aggregates in the mixtures induces greater shrinkage. There is an improvement in the bond between the matrix and the expanded cork aggregates compared to raw cork aggregates. Approximately 24.81% of the shrinkage of the C/L:1/3 composition with raw cork happened at an earlier time (7 days) and reached over 81% at 14 days. A similar result has already been published, confirming this trend (89%) but with the size of a different granule and cement [23].

We observed an increase in shrinkage with increasing cork content, which could be due to the higher porosity and water absorption of raw cork. This could be due to the higher porosity and water absorption of the raw cork, as cork aggregates do not resist forces like mortar aggregates. In this case, shrinkage is also caused by the volume of cement used. It is important to state that cork composite is sensitive to the desorption phenomenon, leading to a significant shrinkage which will stabilize between 28 and 90 days where only a slight increase in shrinkage has been observed. This dimensional variation has significantly lower for hydrophilic aggregates that can reach 10 mm/m [38]. Meanwhile, the observed shrinkage has higher at 33% and 55%, respectively, than the one from wood-based panels and wood chip composites [42].



Fig. 7. Evolution of shrinkage by days: (a) expanded cork, (b) raw cork

#### **4.5 Thermal Conductivity**

Table 4 lists the thermal conductivity values from the different composites M50 with an expanded and raw cork at 28 days. This table also gives the density and compressive strength values obtained for the three mixes studied. The thermal conductivity decreases with increasing cork content. This is due to the formation of a more porous material, and therefore less conductive. Moreover, the thermal conductivity of expanded and raw cork is lower than that of cement. The introduction of cork aggregates in cement-based composites leads to a decrease in the thermal conductivity of the composites, which has been reported by other authors [43, 44]. The lowest thermal conductivity coefficient (0.19 W/m. K) corresponds to the highest raw cork content and the lowest cement content (C/L: 1/3).

The raw cork aggregates gave low values of thermal conductivity. The mixtures: C/L: 1/1, C/L: 1/1, and C/L: 1/3 with raw cork reduced the thermal conductivity by about 17%, 20.6%, and 5% respectively. The cellular structure and high void content of the raw cork aggregates were responsible for the reduction in conductivity.

The results summarized in Table 4 show that the inclusion of expanded and raw cork simultaneously decreased the thermal conductivity, density, and compressive strength of the composites. The C/L: 1/3 mixture with raw cork is comparable to that of the clay-cement-wood and clay-cork composites [26]. This thermal conductivity was slightly higher than that of polystyrene foam [45]. On the other hand, it is more insulating than autoclaved concrete, expanded clay cavity concrete, and expanded shale [46]. The thermal conductivities obtained for C/L: 1/1 and 1/2 mixes confirm the RILEM recommendations for lightweight concrete with a compressive strength greater than 3.5 MPa [47].



Table 4. Thermal conductivity of expanded and raw cork mixes.

#### **4.6 Sound Absorption Coefficient**

Figure 8 presents the sound absorption coefficient of different materials commonly used in construction and composites with both expanded and raw cork. As shown, no peak was recorded. However, the work of Novais et al. [21] revealed that a high peak appears when the cork percentage is over 75%, thus indicating that higher quantities of cork can enhance the sound absorption capacity of cork composites. In terms of the studied blends, all exhibited higher sound absorption capacity at high frequencies (above 1600 Hz). However, for the low-frequency range (20 to 400 Hz), the composites demonstrated a low absorption capacity. Notably, the mid-frequency range (500 and 1600 Hz) displayed a considerable improvement, and the results demonstrated that increasing the volume of raw cork in the composites enhances the sound absorption coefficient. The M50 composite with a C/L ratio of 1/3 (75% raw cork) recorded the highest absorption coefficient ( $\alpha$ = 0.45). The composite with raw cork possesses a more open porosity that allows it to connect the pores and absorb more sound waves.

On the other hand, composites containing expanded cork particles had lower sound absorption coefficients, potentially due to their impermeability following processing in the factory. The absorption capacity was also observed to depend on the volume of expanded cork introduced into the composite, and the results were relatively low. This phenomenon is similar to that of cellular concrete, which has high porosity but can only absorb 40% of the sound due to low permeability. Similarly, materials such as plaster and brick with less permeable surfaces exhibit the same behavior [47]. Furthermore, the binder utilized has a considerable impact on the sound absorption coefficient, primarily because of the less significant open porosity in this type of binder, which makes the material impermeable. Comparable results were observed with cork-gypsum composites [12]. To compare the acoustic performance of insulating materials, we employed noise reduction coefficients (NRC), which are the consecutive averages of sound absorption coefficients at four frequencies (250, 500, 1000, and 2000 Hz) [48]. The NRC of the M50 mixture (C/L: 1/3) with raw cork was 0.26, representing a 38% and 50% increase over the raw cork composites  $C/L:1/2$  and  $C/L:1/1$ , respectively. For the same volume of cork, the NRC of raw cork composites was higher than that of composites with expanded cork, plaster, raw brick, and hardwood flooring but lower than that of perforated brick and flax wool (see Fig. 7).



Fig. 8. Sound absorption coefficient  $(\alpha)$  of some materials

#### **5. Conclusions**

The significant amount of cork waste generated in the environment requires innovative and sustainable solutions for its disposal. The incorporation of cork waste into cement matrices presents a promising option for construction purposes. Our study compared the physical, mechanical, and insulation characteristics of composite materials made from both raw and expanded cork waste.

The results showed that the low density of cork results in a reduction in density and mechanical strength when added to the composites. However, the use of cork waste as the only aggregate in the composites resulted in materials with apparent densities ranging from  $614$  to  $1572$  Kg/m<sup>3</sup>. The compressive strength varied from 2 to 15.02 MPa, while the flexural strength varied from 0.27 MPa to 3.20 MPa, with a strong correlation ( $R^2 > 0.97$ ) between the two. The addition of cork also led to a significant increase in shrinkage, particularly in raw cork composites, although the method of curing used contributed to a reduction in shrinkage.

Moreover, the thermal conductivity coefficient decreased with the addition of cork, resulting in composites with thermal conductivities ranging from 0.19 to 0.47 w/(m. K). While composites with expanded cork showed better mechanical performance, those with raw cork exhibited superior insulating and acoustic performance.

In summary, the results indicate that waste cork can replace sand in the production of insulating composites that are durable, easy to use, and have low thermal conductivity. These composites have various applications, including insulation for roofs, thermal and acoustic isolation, and partition walls, among others. Therefore, the utilization of waste cork in cement matrices can contribute to a more sustainable and environmentally-friendly construction industry. However, this study can be complemented by further in-depth investigations in order to observe, analyze and assess the cork-based composites. The results raise several questions that could constitute important topics for future research.

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