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Recovery of Cork Manufacturing Waste within Mortar and Polyurethane: Feasibility of Use and Physical, Mechanical, Thermal Insulating Properties of the Final Green Composite Construction Materials

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Abstract: The valorization of industrial waste is a hot topic toward circular economy and sustainability. Several wastes have been proposed as resources for different production processes; however, others are still disposed to landfill or waste-to-energy plants. For the first time, this work suggests a sustainable alternative to managing cork waste from bottle caps manufacturing; this is generated by a local company at about 220,000 m^3/year. The powder waste has a 0.063–1 mm particle size and is mainly composed of cork, polyurethane adhesive, and paraffin. Its valorization is proposed as filler in construction materials such as lime-based mortar (1–4 wt%) and polyurethane (5–15 wt%). Thermal, spectroscopic, and physical characterizations are performed on the cork waste, and mainly result in a low apparent density (340 kg/m^3) and high-water absorption (177%). Cork properties allow consideration of extra water in the mortar mix and improve lightness without significantly affecting compressive, bending strength, and thermal insulation. Cork waste in polyurethanes promotes a color change, slightly increases the density (up to 12.5%), and still results in producing a thermally insulating material (<0.06 W/mK). Considering the promising results, this study demonstrates the feasibility of using the manufacturing waste from cork bottle caps to produce green construction materials, thus upgrading it from waste to secondary raw material.

Keywords: waste; recycling; cork; cork bottle caps; mortars; polyurethane; sustainability; green construction materials; thermal insulation; mechanical properties

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1. Introduction

The ongoing population growth, which deals with economic, social, and sustainability issues, has resulted in a significant increase in global solid waste generation, which is currently about 17 billion tons and is estimated to become 50 billion tons by 2050 [1].

Therefore, it becomes necessary to seek different solutions and new applications of waste materials, especially in the construction sector. This would reduce the environmental impact of the construction sector, which is responsible for producing 35% of global CO_2 emissions and 45–65% of waste disposed in landfills each year. [2]

Over the last few decades, attempts have been made to reduce the volume of waste in landfills by converting waste into secondary raw materials [3]. The principal construction materials that could benefit from introducing these secondary raw materials are mortar, concrete, and polyurethane.

Currently, the most widely reused waste in mortar and concrete is construction and demolition waste (CDW), which seeks to solve the problem of natural aggregates exploitation and the increasing production of CDW [4]. Glass waste [5], polymer waste [6], and natural fiber waste [7,8] are also involved in improving sustainability and lightness to the final product.

To avoid expensive disposal, several hazardous wastes which can be flammable, chemically reactive, or corrosive [9] are currently reused as a substitute for natural aggregates or cement (SCM) [10–13]. This also results in decreased demand for cement and natural resources and reduced CO_2 emissions [4]. In addition, it was displayed that some waste can promote the compressive strength of cementitious materials under elevated temperatures [14].

In the literature, some work has focused on the use of virgin cork and very few on cork waste in mortar and concrete. As a result of adding a lightweight material into a cementitious matrix, the introduction of cork is known to decrease the mechanical performance of the finished product. However, it has been shown not to affect it excessively, and the final products remain classifiable according to the strength classes of UNI EN 998-2 [15,16]. Density decreases and thermal conductivity depends on the sample curing conditions, thus on the presence of residual water within the cork [15,17].

On the other hand, lignocellulosic fibers, walnut shells, cork, and other organic waste have been considered inside the polyurethane matrix to create thermal insulating construction materials [18–21]. The main characteristic of polyurethane is thermal insulation [22], and all the additional materials in the mix should not change this characteristic.

In this framework, this paper proposes for the first time the valorization of cork waste from the manufacturing of cork bottle caps within both lime-based mortar and polyurethane. Thus, this work aimed to fill the research gap of cork waste recycling, reduce the large volume of waste in landfills or waste-to-energy plants and provide sustainability to the most important construction materials.

Cork is a natural material from the bark of the oak Quercus Suber L. and is mainly present in the western Mediterranean Sea area, including Sicily, Sardinia, Maremma of Grosseto, Corsica, Spain, and Portugal. The cork production involves about 20,000 km^2 in the Mediterranean area for an annual extraction of 300,000 t [23]. Portugal is the first cork producer with about 52% of world production, followed by Spain (25%) and then all the other countries of the Mediterranean basin including Italy with 6%, concentrated for 4% in Sardinia [23].

Its primary use is in manufacturing wine bottle caps: globally, about 12 billion caps are produced per year [23]. In Italy, particularly in the Emilia Romagna region, and precisely in Reggio Emilia and Parma, cork factories have an ancient tradition.

Cork extraction and processing include several phases. The decortication is carried out between May and June when the bark is softer, and the damage to the plant is prevented. It is a delicate operation entrusted to specialized workers that allows the extraction of large sheets of bark called planks. The planks are then stored for seasoning and selected based on aesthetics. Cork planks with defects are milled and directly transformed into agglomerates for the building industry or agglomerated wine caps. On the other hand, cork planks without defects are used to produce high-quality corks caps. [23] During the cork trimming and cutting phase of high-quality cork caps, a large quantity of cork residue is generated, which is also used to produce agglomerated cork caps (micro agglomerated caps or pieces for sparkling wine caps).

The production of agglomerated cork caps first involves the selection of virgin cork residue (around 79 wt%) depending on granulometry, then the mixing with a binder and other additives (around 21 wt%, i.e., polyurethane glue and paraffin). The mixture is compressed and extruded at 95–105 °C to obtain the polymerization of the polymeric component. Alternatively, the mixture could be pressed and die-cut. Subsequently, a cutting phase occurs and provides agglomerated cork cylinders. Agglomerated caps may differ because of the residue granulometry, type of glue, compressive strength, etc. [24]. Finally, the caps are smoothed, and a large amount of fine cork waste is produced during this phase. This is usually delivered to waste-to-energy plants and not reused in cork manufacturing or other industrial processes.

The local company that supplied the cork waste under investigation produces about 240 t/year of cork residue, corresponding to 218,000 m^3. Since it cannot be reinserted within the same manufacturing process, the most problematic residue is the one containing cork, polyurethane glue and paraffin deriving from the smoothing process of agglomerated bottle caps.

This work proposes a viable solution for the recycling of cork waste from the manufacturing of agglomerated bottle caps, considering its application as filler in traditional construction materials (mortar and polyurethane). This valorization approach is aimed to solve real waste management issues of the local company as well as contributing to the hot research topic on waste recycling. Since no evidence on cork waste has been registered by literature, this work assesses for the first time its properties and influence on those of the matrixes in which it could be used as filler.

To this aim, a thorough preliminary characterization of the cork waste was performed, and morphological, chemical, physical, and thermal properties were investigated. Then, increasing cork waste content was considered in the construction materials to maximize the recycling effectiveness. Namely from 5 wt% to 15 wt% (from 35 vol% to 62 vol%) within polyurethane and from 1 wt% to 4 wt% (from 16 vol% to 65 vol%) within lime-based mortar. Optical microscopy, colorimetry, apparent density, thermal conductivity, three-point bending, and compressive strength tests were carried out to detect the influence of cork addition on the properties of commercial polyurethane and mortar. A curing of 35 days and drying at 50 °C for 12 h were considered for mortar samples to avoid the influence of cork humidity during testing.

2. Materials and Methods

2.1. Cork Waste

A cork waste from the manufacturing process of agglomerated cork bottle caps of a local company (Italsughero, Montecchio Emilia (RE), Italy) was considered. Precisely, this powder waste is generated during the smoothing phase of agglomerated cork caps and is directly collected through a cyclonic air-filtering system. The involved cork waste has a particle size distribution of 0.063 mm < d < 1 mm and contains polyurethane glue and paraffin which are industrially used as binders and additives for cork particles, respectively.

Simultaneous thermal analysis (STA 449 F3 Jupiter, Netzsch-Gerätebau GMBH, Selb, Germany) was considered for thermal characterization and composition description of cork waste. This technique allowed to merge differential scanning calorimetry (DSC) and thermal gravimetric analysis (TGA) in the same measurement. Thus, it allowed relating each thermal transformation of the material to the corresponding mass loss. The test was performed considering a heating ramp from 0 °C to 900 °C at 10 °C/min heating rate, a silicon furnace (heating up to 1500 °C), and aluminum crucibles. Data analysis was carried out through Proteus software (Netzsch Proteus Thermal Analysis, Proteus 6.1.0, Netzsch-Gerätebau GmbH, Selb, Germany), resulting in a combined plot of DSC, TGA and the first derivative of TGA (DTGA). The composition of cork waste was also detected through attenuated total reflectance Fourier transformation infrared spectroscopy (ATR-FT-IR analysis, FTIR VERTEX 70, Bruker Optics, Germany). A transmittance range between 400–4000 cm^{-1}, 32 scans, and a resolution of 4 cm^{-1} was considered, while FTIR spectra were analyzed through OPUS software (OPUS 6.5, Bruker Optics GmbH, Selb, Germany).

A laser particle sizer (Malvern Mastersizer 2000, Malvern Panalytical Ltd., Malvern, UK) was used to determine the particle size distribution of cork waste particles. Optical microscopy (LEICA EZ4D, Leica Microsystems) with 8×, 20× and 35× magnitude was considered for the morphological description of cork waste particles.

According to UNI EN 1079-6 2013, water absorption (WA) and apparent density (ρ_{rd}) values were evaluated by performing a 24 h water absorption test. Finally, real density was measured through a helium pycnometer (Micrometrics Accupyc 1330, Micrometrics Instruments).

2.2. Mortars

The first attempt to valorize the cork waste involved its use as filler within mortars. A commercially available mortar (GeoCalce Antisismico G by Kerakoll S.p.A., Sassuolo, MO, Italy) for historical buildings restoring and improving seismic resistance was considered. The binder is mainly composed of natural hydraulic lime NHL 3.5 and Portland cement. While the aggregate has a particle size between 0 mm and 2.5 mm, and a silicate-carbonate mineralogic composition. The technical data sheet points out an optimized effective water (w_{eff})-to-mortar ratio of 0.204, and a fresh mortar flow diameter of 180 mm (UNI EN 459-2:2002). Moreover, it certifies the mortar as belonging to the M15 strength class (UNI EN 998-2).

Increasing cork content was added from 1 wt% to 4 wt% of mortar, which in terms of volume corresponds to much higher percentages, 16 vol% and 65 vol%, respectively.

The mix design of the four lime-based mortars containing cork waste is reported in Table 1 compared to the reference commercial one. As displayed in Table 1, a contribution of effective water (w_{eff}) was set to reproduce an effective water-to-mortar ratio equal to 0.204. At the same time, extra water (w_{extra}) was also considered to compensate for the amount of water absorbed by the cork, as assessed by the WA test on cork waste.

Table 1. Mix design of mortars containing increasing content of cork waste (from 0 wt% to 4 wt%).

Cork (wt%)	Cork (vol%)	Cork (g)	Mortar [1] (g)	Effective Water [2] (g)	Extra Water [3] (g)	Superplast. (g)
0	0.00	0.00	8041.04	1604.40	0.00	0.00
1	16.15	69.31	6931.04	1413.93	122.88	29.6
3	48.49	162.04	5431.49	1108.02	288.88	49.84
4	64.66	196.05	4901.29	999.86	347.58	44.96

[1] NHL3.5 + Portland cement + natural aggregate, commercial powder mix. [2] water available for the binder hydration. [3] water added to compensate for water absorption by the cork.

The workability of the reference mortar (flow diameter of 180 mm) was set as the target value to be also maintained for mixes containing cork. Thereby, the flow table test (UNI EN 459-2:2002, performed through Flow Table 64, ControlsGroup, Milan, Italy) was preliminary involved to evaluate the effect of filler addition on workability. In order to improve the workability of the fresh mortars containing cork waste, a commercial superplasticizer was considered following its datasheet prescriptions and as reported in Table 1. Cork contents higher than 4 wt% were not considered due to workability issues.

First, water and mortar were mixed at low speed (300 rpm) for 30 s through a high-speed laboratory mixer (RW20 DZM, IKA-Werke GmbH & Co. KG, Staufen, Germany), then the cork was added, continuing mixing for other 30 s. Subsequently, the mixing phase continued for 1:30 min at high speed (500 rpm), following the prescriptions of UNI EN 459-2:2002.

The mixes were cast into 40 × 40 × 160 mm^3 and 300 × 300 × 30 mm^3 formworks to produce samples for the three-point bending and thermal conductivity tests, respectively. Four samples for mechanical tests and three for thermal conductivity were produced for each mortar type.

According to UNI EN 196-1, samples were cured for 28 days in a climatic chamber with RH 90% ± 5% and temperature of 20 °C ± 2 °C. Moreover, additional 7 days of cure at laboratory conditions (RH 60% ± 5% and temperature of 25 °C ± 2 °C) were considered to have complete evaporation of the water potentially still present in cork particles.

Four 40 × 40 × 160 mm^3 samples for each designed mix were mechanically tested through three-point bending, while a compressive strength test was carried out on four 40 × 40 × 40 mm^3 residual samples from the three-point bending test. The tests were performed following the UNI EN 196-1:2005 and using a universal testing machine (UTM, INSTRON 5567) equipped with a load cell of 30 kN. The three-point bending test was performed at a nominal rate of 1 mm/min under displacement control mode, and a distance of 10 cm was set between the two steel supports. The compressive strength test was also carried out with the same testing parameters and using two circular steel plates.

Apparent density was geometrically evaluated as the ratio between the measured mass and the known volume (40 × 40 × 160 mm^3). The mean apparent density of each hardened mortar type was computed as the average over three measurements.

Optical microscopy (LEICA EZ4D, Leica Microsystems) allowed the description of the cork distribution within the mortar, whereas the colorimetry (CIELab method through PCE-CSM6 colorimeter, PCE Instruments) to see the color change of mortar samples at increasing cork content.

The thermal conductivity of mortars was measured through a heat flow meter (HFM Lambda, Netzsch-Gerätebau GmbH, Selb, Germany) after drying the samples for 12 h at 50 °C; the mean thermal conductivity was derived as the average over three measurements.

2.3. Polyurethanes

The second attempt to valorize the cork waste dealt with its use as filler within polyurethanes. A commercially available polyurethane was involved, and an isocyanate:polyol weight ratio of 7:5 was considered. Cork additions equal to 5 wt%, 10 wt%, and 15 wt% of reference polyurethane mix (0 wt% cork) were considered for the mix design of the polyurethane samples (Table 2).

Table 2. Mix design of polyurethanes containing increasing content of cork waste (from 0 wt% to 15 wt%).

Cork (wt%)	**Cork (vol%)**	**Cork (g)**	**Isocyanate (g)**	**Polyol (g)**
0	0.00	0.00	94.99	67.85
5	34.99	8.14	94.99	67.85
10	51.84	16.28	94.99	67.85
15	61.75	24.43	94.99	67.85

The cork waste was first mixed with polyol using a high-speed laboratory mixer (RW20 DZM, IKA-Werke GmbH & Co. KG, Staufen, Germany). Then the isocyanate was added, and the mixture was stirred for 45 s. Rapidly, the mixture was poured inside a 200 × 200 × 80 mm^3 metal sealed mold and thermally controlled during the expansion phase. Precisely, the expansion of the polyurethanes was performed for 20 min at 25 °C, and thermal control was ensured by a refrigerated/heating circulator (Argolab CB 5–30, −30 °C to +100 °C) connected to a coil placed inside the metal sealed mold.

The apparent density of polyurethane samples was computed as the ratio between the measured mass and the known volume (200 × 200 × 80 mm^3). The mean apparent density of each polyurethane type was derived as the average over three measurements.

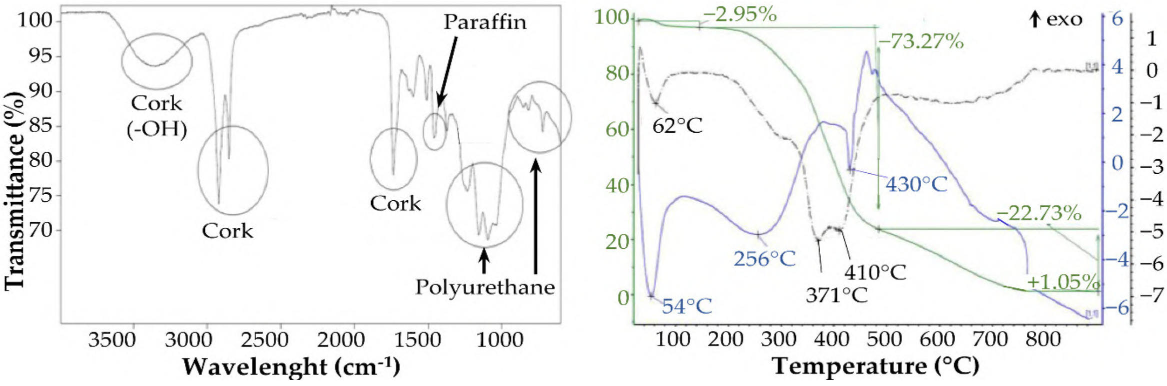
Optical microscopy (LEICA EZ4D, Leica Microsystems) allowed the description of the cork distribution within the polyurethane matrix and its influence on porous polyurethane structure. In addition, a colorimetry test (CIELab method through PCE-CSM6 colorimeter, PCE Instruments) was involved in detecting the color change of polyurethane samples at increasing cork content.

The thermal conductivity of polyurethane samples was measured through a heat flow meter (HFM Lambda, Netzsch-Gerätebau GmbH, Selb, Germany), and the mean thermal conductivity was derived as the average over three measurements.

3. Results and Discussion

3.1. Characterization of Cork Waste

The FTIR and the STA analyses confirmed the chemical composition of the cork waste resulting in cork, polyurethane glue, and paraffin (Figure 1). Precisely, in the FTIR spectrum of Figure 1a, the characteristic peaks of cork can be identified at 3400 cm^{-1} (-OH bond), 2920, 2850 cm^{-1} (stretching) and 1737 cm^{-1} (bending) [24]. Paraffin can be detected at 1459 cm^{-1} (bending) [25], and polyurethane glues at 1235, 1156, 1094 (bending) and 721 cm^{-1} (bending-rocking) [26].



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

Figure 1. Composition of cork waste through ATR FT-IR analysis (**a**) and STA analysis (**b**). In Figure 1b are displayed the following curves: TGA (green line), first derivative of TGA (DTGA, black dashed line) and DSC (blue line).

On the other hand, the presence of cork, paraffin, and polyurethane glue was confirmed by thermal analysis. Considering DTGA (Figure 1b, black dashed line), the complete degradation of cork can be observed at around 371 °C and 410 °C [24]. The other exothermic peaks (between 600 °C and 800 °C) can be attributed to the degradation of paraffin [27] and polyurethane glue [26]. Moreover, the TGA curve (continuous green line) showed a mass loss of around 76.22% related to cork degradation [28] and a mass loss of 22.73% attributable to the degradation of paraffin and polyurethane glue [26,29]. Contrary to expectations, a greater content of glue and paraffin (>21 wt%) was observed. This is attributable to the fact that, during the production of agglomerated cork caps, the glue and paraffin tend to concentrate on the cap surface, which is the part subject to smoothing and from which the cork waste is generated. So, the cork waste under investigation displayed a slight difference in composition compared to the original agglomerated cork cap.

As reported in Figure 2, the cork waste displayed a bimodal particle size distribution with main peaks at 150 µm and 600 µm. A fine particle size of cork waste can also be observed (0.063 mm < d < 1 mm).

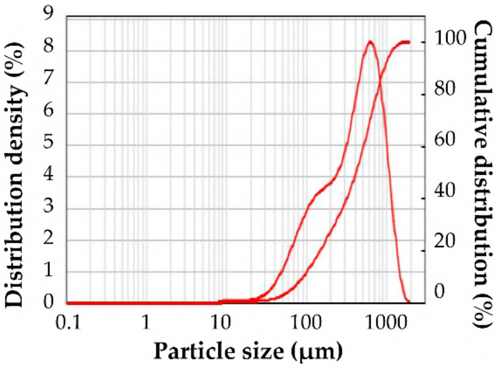
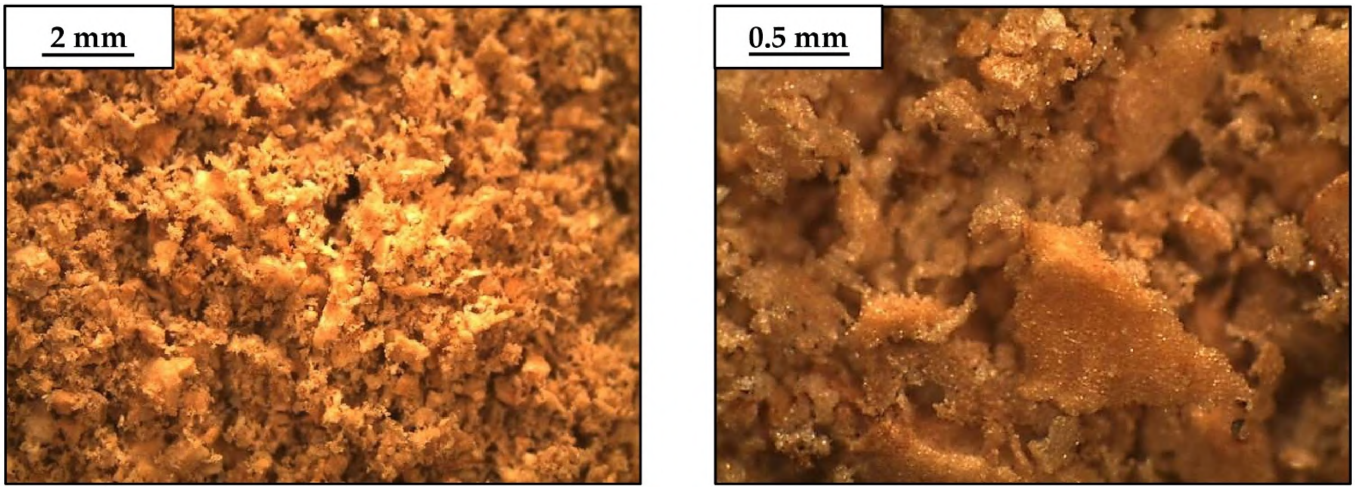


Figure 2. Particle size distribution of cork waste.

Figure 3 shows the morphology of cork waste at different magnifications, namely 8× and 35× magnification. The powder particles own the peculiar cork color, have an angular shape, and are different in size. Particle agglomerations and a high porosity rate between them can be observed.



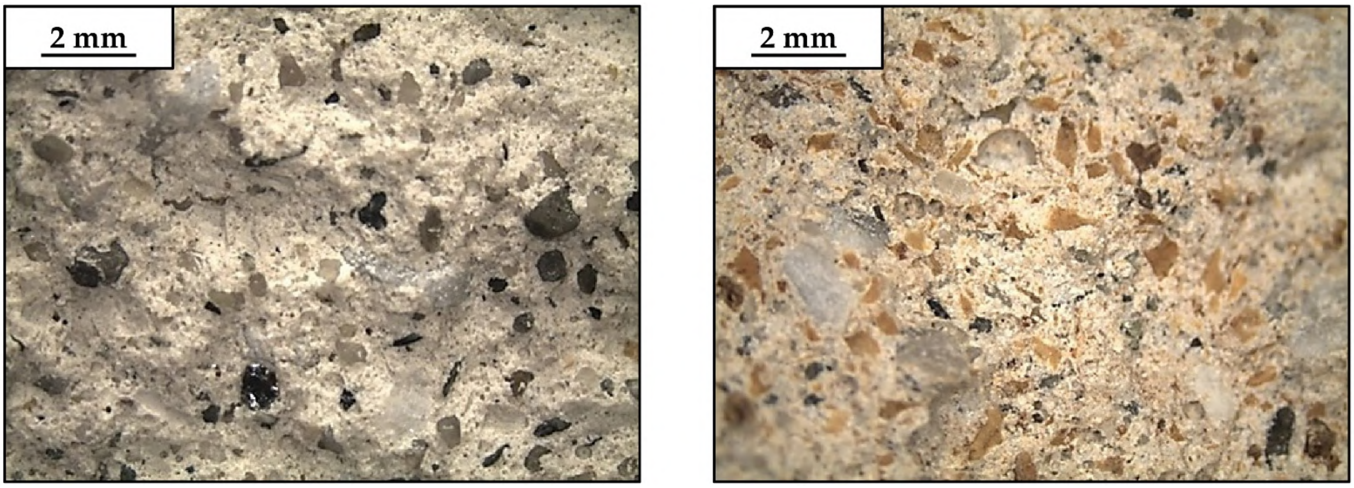
(**a**) (**b**)

Figure 3. Optical microscopy images of cork waste: (**a**) 8× magnification, (**b**) 35× magnification.

Remarkably, a water absorption of 177% was detected for cork waste, which is a consistent value considering the low particle size of the cork powder [27,30]. Moreover, an apparent density of 340 kg/m^3 and a real density of 1110 kg/m^3 were registered. These values are slightly higher than those about virgin cork from the literature [27] but are consistent considering the presence of polyurethane glue and paraffin.

3.2. Characterization of Mortars Containing Cork Waste

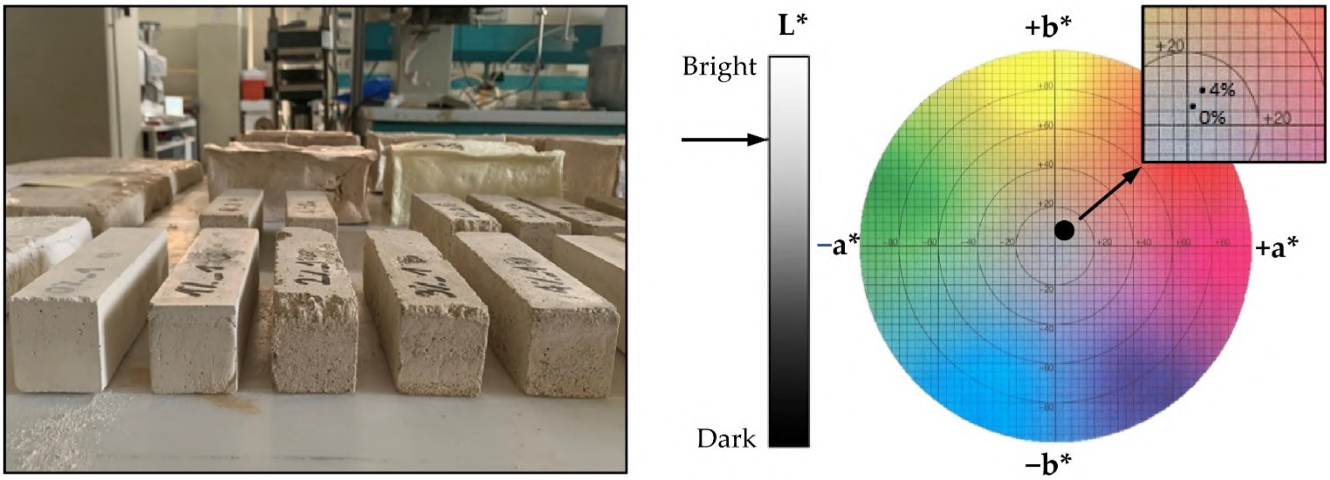
A good and homogeneous distribution of cork within mortars was detected for all the design mortar mixes through optical microscopy. A comparison between the reference mortar without cork (Figure 4a) and mortar with 4 wt% (Figure 4b) is reported as a representative comparison for the other mixes in Figure 4. In Figure 4b, the cork particles, recognizable by their characteristic color, are well distributed within the matrix and have both fine and coarse particles. Moreover, it can be observed that the maximum size of cork particles is smaller than that of natural aggregate (gray grains).



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

Figure 4. Optical microscopy images at 8× magnification of mortars containing 0 wt% (**a**) and 4 wt% (**b**) of cork waste.

The introduction of increasing cork content involved a color variation in mortars. This phenomenon was visually and macroscopically observed (Figure 5a) and also confirmed through the colorimeter test (Figure 5b and Table 3).



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

Figure 5. Color variation of mortar samples induced by the addition of cork waste: visual macroscopic evidence of mortars containing increasing cork content from left to right (**a**) and colorimetry values of representative samples containing 0 wt% and 4 wt% of cork (**b**).

Table 3. Colorimetry test results for mortars.

Cork Content (wt%)	a*	b*	L*
0	1.86	6.79	83.22
1	2.16	6.90	82.26
3	2.87	11.17	74.52
4	3.09	11.29	74.35

Table 3 shows the results obtained by the colorimetry test in terms of a*, b*, L* values, where:

- a* > 0 predominance of red; a* < 0 predominance of green,
- b* > 0 predominance of yellow; b* < 0 predominance of blue,
- L*: 0 = dark, 100 = bright [31].

It can be observed that at increasing content of cork, mortars increase in yellow component (increasing positive a* values), in red one (increasing positive b* values) and become darker (decreasing L* values). Representative color comparison between the samples containing 0 wt% and 4 wt% of cork is graphically reported in Figure 5b within the CIELAB color space.

The thermal conductivity test showed that, although the cork is regarded as an insulating material [24], the introduction of increasing cork content did not make the mortar an insulator. Indeed, thermal conductivity values above 0.1 W/mK, limit not to be exceeded for the definition of thermally insulating material according to UNI EN 998-1, were recorded for all mortar mixes (Figure 6). The mortars containing up to 3 wt% of cork displayed similar thermal conductivities to the reference mortar (0 wt%). Interestingly, the thermal conductivity of mortars tends to increase with cork addition and increased by 12% for the mortar with 4 wt%. This phenomenon resulted from the promotion of binder hydration by the progressive and slow release in time of the water absorbed by cork (w_{extra}), which contributed to reducing the matrix porosity with hydration products. So, even if an insulating material was added, the mortar with more cork (4 wt%) was more thermally conductive than the other. On the other hand, the reduction of matrix porosity stands as a highly valuable consequence of cork addition, and that could contribute to the prevention of chemical degradation at equal or enhanced lightweight conditions.

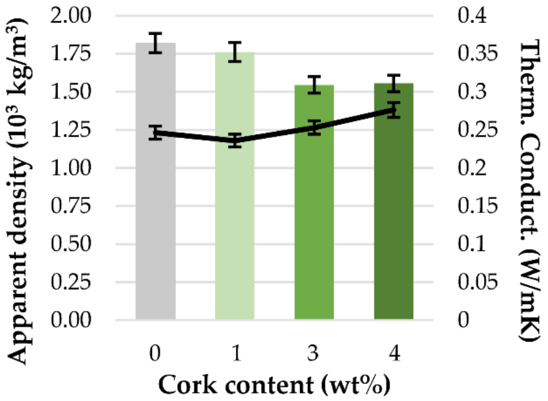


Figure 6. Correlation between apparent density (bar chart) and thermal conductivity (black line) of mortars containing increasing cork content. The error bars stand for standard deviation error.

Since the cork is much lighter than mortar (−87%), as expected it was observed a gradual decrease in the apparent mortar density with the increase of cork content (Figure 6). So, even though adding higher cork content increased thermal conductivity due to the lower porosity of the lime-based matrix, the designed mortars were lighter because the cork was an extremely lightweight material. Despite this, the designed mortars can not be defined as lightweight mortars because they all exceeded the apparent density threshold value of 1100 kg/m^3 according to UNI EN 206-1.

As expected, the increase of cork percentage in mortars involved a strength decrease for both three-point bending and compressive strength tests (Figure 7). This has also been observed by literature and is mainly attributable to the poor mechanical strength and stiffness of cork [15,16]. However, the decrease in mechanical properties was not as pronounced as expected. This might result from the promotion of the binder hydration phenomenon by a slow but progressive release of the water absorbed by the cork (w_{extra}). Remarkably, this could be an interesting phenomenon for mortar application in dry conditions; indeed, it would involve better binder hydration, contrast the water evaporation, and avoid shrinkage cracking [16,32].

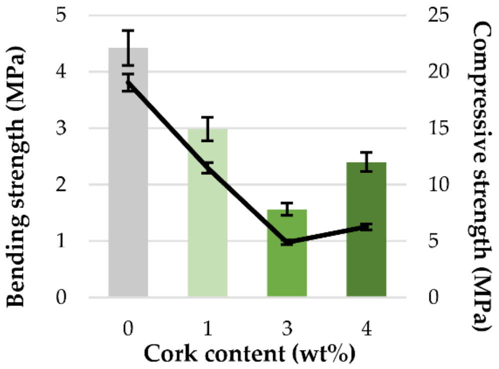
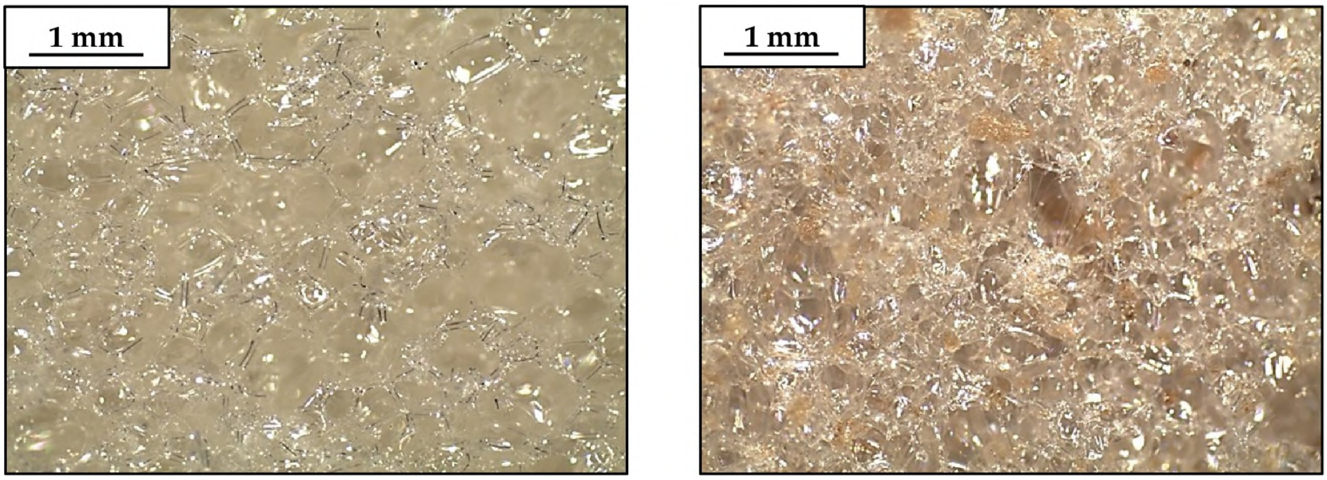


Figure 7. Mean failure strength from mechanical testing of mortars containing increasing cork content: three-point bending (bar chart), compressive strength (black line). The error bars stand for standard deviation error.

Despite the presence of high cork content (Table 1), all mortar mixes demonstrated good mechanical performance. Referring to results from the compressive strength test and prescriptions by UNI EN 998-2, different strength classes were assigned to the involved mixes. The reference mortar without cork was classified in the M15 class, to be used for structural recovery or anti-seismic reinforcement. The mortars containing 1 wt% and 2 wt% of cork were classified in the M10 strength class, so they were considered to have a good mechanical strength; these can be used both for seismic and insulating applications. The mortar containing 4 wt% of cork was classified in the M5 class, so it could be primarily used for lightweight and insulating applications.

3.3. Characterization of Polyurethanes Containing Cork Waste

Cork particles, easily identifiable by their characteristic color, were homogeneously distributed within the polyurethane matrix as displayed by optical microscopy (Figure 8b). The highly porous structure of polyurethane, which makes it an insulating material, is recognizable both for the reference sample (Figure 8a) and the sample containing 15 wt% of cork (Figure 8b). However, it was also evident that by increasing the percentage of cork, some of the pores generally filled with air were filled with cork grains.



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

Figure 8. Optical microscopy of polyurethane samples at 20× magnification with 0 wt% (**a**) and 15 wt% (**b**) of cork waste.

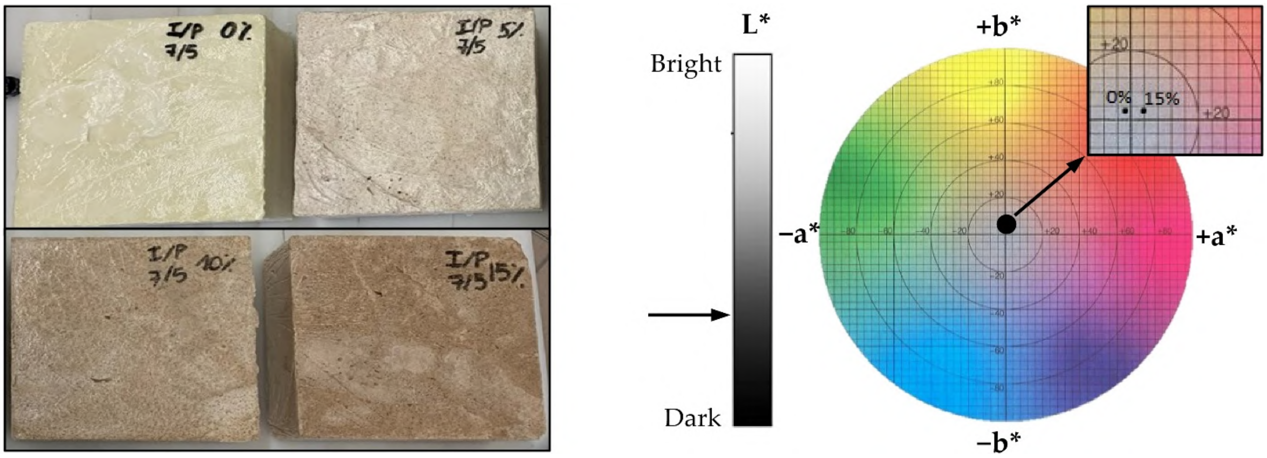
As stated for mortars (Section 3.2), the introduction of increasing cork content involved a significant color variation in polyurethane panels. Figure 9 reports visual and macroscopic evidence (Figure 9a) and colorimetry graphical results (Figure 9b).

The colorimetry results (also reported in Table 4) mainly display a variation of the red color (a* value) above the other that remain almost unvaried (b* and L*).

Table 4. Colorimetry test results for polyurethanes.

Cork (wt%)	a*	b*	L*
0	−0.44	3.86	32.04
5	2.58	4.11	33.85
10	4.20	4.45	30.47
15	3.66	3.81	30.62

Increasing cork content involved a slight increase in the apparent density of polyurethanes (Figure 10). Although cork is a very light material, a slight increase in density was observed since cork fills the pores of the polyurethane matrix (Figure 8b), replacing the lighter air. However, since cork was very light, a very negligible density increase was displayed (≈6 kg/m^3) between the reference polyurethane and the one containing 15 wt% of cork.



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

Figure 9. Color variation of polyurethane samples induced by the addition of cork waste: visual macroscopic evidence of polyurethane containing, from left to right, 0 wt%, 5 wt%, 10 wt%, 15 wt% of cork (**a**) and colorimetry values (**b**).

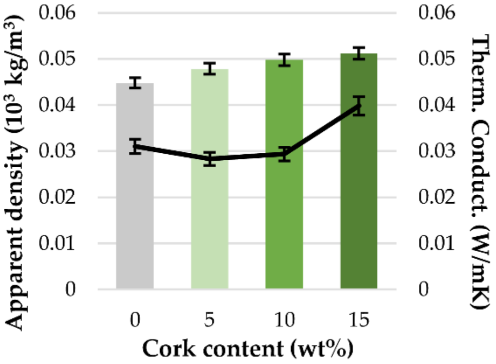


Figure 10. Correlation between apparent density (bar chart) and thermal conductivity (black line) of polyurethanes containing increasing cork content. The error bars stand for standard deviation error.

Thermal conductivity measurements were performed primarily to set if the addition of cork within the polyurethane mix could affect the definition of insulating materials. Figure 10 displays similar thermal conductivity values between polyurethanes containing 0 wt%, 5 wt%, 10 wt%. In contrast, an increase of around 33% was observed for the sample with 15 wt% of cork (0.03981 W/mK) compared to that with 10 wt%. In fact, the insulating capacity of the polyurethane related to the high air volume was affected by higher cork content, which substituted air inside the pores. However, since cork is an insulating material itself, this increase in thermal conductivity (0.03981 W/mK) did not affect the definition of polyurethane as a thermally insulating material. Thus, all polyurethane mixes can be considered insulating materials due to thermal conductivity lower than 0.06 W/mK, which is the characteristic thermal conductivity of a standard thermally insulating polyurethane.

In conclusion, it is possible to add up to 10 wt% of cork and obtain an insulating material with an equal thermal insulating capacity to the reference one. Still, it is also possible to add 15 wt% of cork, obtaining a material that can be defined as an insulator as well.

Following these promising results, future works will investigate the effect of cork waste addition on durability, temperature-dependent mechanical properties, and flame resistance of mortar and polyurethane samples.

4. Conclusions

The continuous growth of the world population is the main cause of generating a large volume of waste from industrial processes. Among these, cork waste from the manufacturing of bottle caps has not found any valorization application and is currently delivered to waste-to-energy plants. In order to face this problem, this work proposes its valorization as filler within lime-based mortars and polyurethanes, with the double objective of valorizing the waste and providing sustainability to traditional building materials.

The cork waste under investigation was characterized through FTIR and STA and resulted in a mixed composition of cork (about 76%), paraffin and polyurethane glue (about 24%). The cork displayed a particle size distribution of 0.063 mm < d < 1 mm and a low apparent density of 340 kg/m^3. High water absorption (about 180%) was observed and considered for cork application within mortar.

The addition of increasing contents of cork in mortars, i.e., from 1 wt% to 4 wt% (16 vol% to 65 vol%), gradually decreased the three-point bending and compressive strength of the reference mortar (0 wt%). However, the decrease in mechanical performance was less than expected, thanks to a continuous hydration promotion. In fact, the water absorbed by the cork during the mixing phase was later released over time and promoted better hydration of the binder. Therefore, better mechanical properties and less porosity of the final product were observed than expected. The apparent density of mortar mainly decreased with higher cork content, i.e., from 1820 kg/m^3 (0 wt%) to 1560 kg/m^3 (4 wt%). In conclusion, green mortars possibly suitable for the masonry restoration were obtained, with a reduced density by 15%. Lightness is a major property of restoration materials because it allows reduced loads applied to the pre-existing structure.

The peculiar hygroscopic property of cork could be exploited for applications in dry environments, where too fast water evaporation, which could lead to the creation of cracks and early degradation, could be avoided by the release of water over time by cork.

Cork contents up to 15 wt% (62 vol%) were considered within polyurethanes. Apparent density and thermal conductivity of polyurethanes containing cork slightly increased by 6 kg/m^3 and 0.01 W/mK, respectively. This is due to the fact that the cork fills the porosities that are usually filled with air and make polyurethane a highly lightweight thermal insulator. However, the thermal conductivity of these samples (0.03–0.04 W/mK) satisfied the requirements for the definition of thermally insulating material and standard insulating polyurethane (<0.06 W/mK).

For both mortar and polyurethane, the addition of the cork waste also caused a color shift toward red-yellow color, which could be regarded as a positive aesthetic factor.

In conclusion, this preliminary investigation allowed to identify possible alternative solutions to waste-to-energy of cork waste, thus making a so-far-linear economy circular and moving towards a more sustainable industry. The encouraging results displayed that it is feasible to valorize manufacturing cork waste as secondary raw material for construction materials and involve desired properties such as sustainability, lightness, coloring, etc., without affecting mechanical and thermal insulating performances. The companies involved in cork waste management issue can take advantage of these sustainable practices and valorize their manufacturing waste from an economic and sustainability point of view. Even though policies generally do not encourage the effective use of secondary raw materials in construction materials by strongly limiting their content or application use, this work points out positive performance that could be considered for real applications. Moreover, this study displays for the first time the properties of cork waste from the manufacturing of cork bottle caps and the influence of cork waste use in traditional construction materials. Therefore, it seeks to fill the research gap on cork waste recycling and contribute to the first valorization solutions.

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