

## Article

# Comparative Evaluation of Gypsum-Based Plasters with Pistachio Shells for Eco-Sustainable Building

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**Abstract:** Agri-food waste represents a serious problem that can be overcome by converting it into added-value material for the production of plasters for green building; in fact, it can be used as a reinforcement additive in the building material industry. In this study, the performance of gypsum-based plasters with pistachio shell additives was evaluated. Before being used as additives for gypsum-based plasters, pistachio shells were ground at three different grain sizes in order to verify how grain size influences the performance of the material. Tests were then carried out on all the produced mortars to evaluate their chemical and physical characteristics, and interesting results regarding the mechanical resistance of some of the produced materials were obtained. The results showed that the addition of pistachio shells improved mechanical performance in all cases and that the best mechanical performance and water absorption by capillarity were achieved with the 0.5–2 mm pistachio grain size, while the best thermal conductivity was achieved with the 2–4 mm grain size. Summarizing, the best results were obtained with a pistachio shell granulometry of 0.5–2 mm, sand, and a water/gypsum ratio of 0.86–0.74.

**Keywords:** agri-food waste; biobased mortars; gypsum plaster; pistachio shells



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## 1. Introduction

We can consider as waste any material that, at the end of any production process, is useless, destined for disposal. In recent decades, the production of waste has progressively increased as a direct consequence of economic and industrial development, population growth and the expansion of urban areas. Problems relating to disposal have assumed increasing proportions, partly due to the multiplication of the types of waste produced, which are increasingly harmful to the environment and to the health of living beings. In terms of environmental balance, the increasing amount of produced waste is related to the depletion of the earth's resources, and disposal involves a significant loss of materials and energy.

Building materials are one of the main factors influencing the overall environmental impact of the construction industry due to their production, transportation, use and end of life [1]. To reduce this problem, some strategies have been identified, such as the usage of environmentally friendly materials, the use of local materials and the encouragement of circular economy practices [2]. J. Bolden et al. [3] investigated the strengths and weaknesses of the practice of supporting the construction industry in developing effective policies regarding the use of waste and recycled materials as building materials.

According to the Waste Framework Directive (2008/98/EC), “recycling” is “any recovery operation through which waste materials are reprocessed to obtain products, materials or substances to be used for their original function or for other purposes” [4]. If recycling,

reuse or other treatment operations result in “secondary raw materials” being obtained from waste, this also constitutes a “recovery” operation (Dir 2006/12/EC) [5].

To date, research has aimed at investigating the possibility of using recycled aggregates (RAs) derived from the reuse of construction and demolition waste [6] or natural materials with possible advantages, such as low environmental impacts, low energy demands, low costs, large-scale availability, biodegradability, and good insulating and mechanical properties, as additives in construction products. A. A. Sundarraj and T. V. Ranganathan, in their studies, investigated the possibility of using cellulose extracted from agro-industrial waste as a reinforcement for the building material industry [7].

Current research on biobased lightweight mortars focuses mainly on physical (workability, density, porosity, etc.) and mechanical (compressive strength, tensile strength, elastic modulus, etc.) properties. Generally, bioaggregates possess high porosity and low specific gravity, which make significant contributions to the functional properties of mortars and concrete, such as thermal insulation and sound absorption properties. Currently, various biobased aggregates have been successfully added in the mix design of mortars, for instance, pistachio shells [8–10], date seeds [11], coconut shells [12–14], bamboo [15], apricot shells [16] and seashells [17]. Other studies concerned with the realization of composites have used rice straw [18]; rice husks [19]; bagasse, i.e., the residue from the grinding and pressing of sugar cane [20]; corn cobs [21,22]; pineapple leaves [23]; coffee chaff [24]; coffee grounds [25,26]; sunflower seeds [27,28]; durian peel and coconut coir [29]; peanut shells [30,31]; walnut shells [32]; Opuntia [33]; hemp [34]; and flax fibers [35]. According to [36], the adoption of biobased building materials can imply not only a reduction in environmental impact compared to traditional materials but also a reduction in production costs.

Most of these products have not yet been industrialized and marketed, but they potentially represent a sustainable alternative for the green building sector, with performance values comparable to those of commercial products, and in some cases achieve better results, particularly with respect to thermal and acoustic insulation properties [37]. Some research has also shown how natural fibers can effectively contribute to increasing the mechanical resistance of mortars. Wu et al. demonstrated, for example, how fibers of *miscanthus*, a common grass, improved the compressive and flexural strength of cement mortar by 82.7% and 26.9%, respectively, due to reduced porosity [38]. Affan et al. [39] experimented with cement-based mortars to which wood ash from wood incineration and vegetable aggregates (sunflower plant bark and pith) were added and found that the mortar prepared with sunflower bark had a better mechanical resistance than that made with pith.

However, the outcomes are not always positive; for example, the studies by Nguyen et al. [17] found that the freeze–thaw performance of mortars with additives from marine shells was significantly lower than that of the same mortars without additives due to the dissociation of the calcium carbonate present in the shells.

In addition, biobased light aggregates are made up of carbohydrates and are porous, so they are particularly sensitive to changes in ambient humidity; as a result, they can have a negative impact on durability, especially on water-related properties, such as shrinkage from drying and resistance to freeze–thaw. However, some studies have shown that the heat treatment of biobased additives can significantly reduce the shrinkage of mortar, increasing its resistance to freeze–thaw due to reduced microcracking and porosity [40].

In general, territorial dynamics play an important role in the implementation of the circular economy, which is oriented towards the recovery and minimization of waste and must be associated with experimental studies on the characteristics and quality of local materials [6,41–44]. In particular, the cultural premises of this study refer to the shared identity between Sicily and Tunisia, focused by a recent strategic cross-border cooperation project [45]. In fact, the present experimental research focused on two materials that are easy to find in these places, applying a replicable method to other regions in the Mediterranean area.

Pistachio is widely used in Sicilian and Tunisian gastronomy and confectionery. Pistachio shells are a waste of highly consolidated industrial realities spread on a regional scale in Sicily. Gypsum is present in the geological texture of southern Sicily and northern Tunisia and is widespread in historical and vernacular architecture in both regions. This research, therefore, focuses on the use of pistachio shells in manufacturing gypsum-based plasters, a traditional local material [46–50] widely used as plaster due to its low cost and good fire resistance [51]. Indeed, further development of this research could include assessment of fire resistance through combustion experiments.

Pistachio shells have been used to try to overcome the limitations presented by gypsum, such as low mechanical resistance, high water absorption and low thermal insulation properties [52]. Several tests were carried out on the composed materials to verify their mechanical and physical properties, and the results of the comparative investigations showed interesting results in terms of mechanical resistance.

## 2. Materials and Methods

### 2.1. Method and Phases

For the development of this research, gypsum-based plaster mortars with the addition of pistachio shells were tested as ground plaster and internal and/or external finishing plaster; all samples were made according to the UNI EN 13279-2 standard [53].

The adopted method was based on the drafting of summary records of all the activities carried out. Filing allows for a homogeneous collection of data and easy archiving and consultation of the latter and the results obtained.

In particular, for each aggregate, a record card was created containing the following information:

- Operators;
- Material;
- Grain size;
- The manufacturer who supplied the aggregate;
- The specific weight of the aggregate;
- Preparation methods;
- Notes.

Regarding the mix designs, record cards were also filled that contained the following information:

- Sample name;
- Realization date;
- Operators;
- Composition of the mix design;
- Percentage by weight of binder, aggregates and water;
- Workability of the dough;
- Photos of the realized samples;
- Notes.

Examples of record cards are shown in Figure 1.

After the material selection and the mix design definition, different sizes of mortar samples were produced. After the necessary seasoning, the samples were tested in the laboratory to evaluate their performances, such as mechanical resistance, thermal insulation and behavior over time.

During the production phase, tests were carried out to quantify the correct water/binder ratio, as indicated in the UNI EN 13279-2 standard [53], in order to guarantee adequate workability characteristics of the mortar and certain solid-state performances in mechanical, chemical and physical terms. Through the use of pistachio shells, we also attempted to increase the efficiency and performance of the basic mixture which constituted the matrix by analyzing its new characteristics.



Figure 1. Record cards: (a) aggregates; (b) mix designs.

The project's activities were divided into five phases:

1. Identification of raw materials;
2. Production of mortars;
3. Basic characterization;
4. Aging tests;
5. Comparative tests.

In the first phase, the materials to be used for the production of the mortars were identified; in particular, we chose to use gypsum from the Trapani area manufacturers in Sicily and pistachio shell agricultural waste from local Sicilian production based in Bronte (CT), Sicily.

The second phase involved the production of samples with aggregates of different granulometries in order to verify how these influenced the workability and spreadability of the plasters, followed by a curing phase in which the samples were conserved for 7 days at a temperature of  $23 \pm 2$  °C with a relative air humidity of  $50 \pm 5\%$  and subsequently dried to a constant mass at a temperature of  $40 \pm 2$  °C as per the UNI EN 13279-2 standard [53]. After curing, in the third phase, preliminary tests were carried out to evaluate the performance characteristics of the produced mortars. In the fourth phase, the durability and behavior of the materials over time were evaluated by subjecting the samples to artificial aging cycles and repeating some of the characterization tests. The last phase involved carrying out tests on post-aging samples to verify whether there were any variations in the performance characteristics of the produced materials.

## 2.2. Gypsum

The term *gypsum* generally indicates both the natural mineral and the industrial product derived from it and, sometimes, also indicates the chemical compound of these two substances.

Gypsum is an inorganic compound that has sedimented in some deposits in remote geological times. In nature, this mineral is found in two different forms, namely, anhydrous calcium sulphate ( $\text{CaSO}_4$ ) and dihydrated calcium sulphate ( $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ ). The difference

between the two forms lies in the fact that in the second case a certain quantity of water of crystallization is present in the crystals [52,54], and from a chemical point of view, the material occurs in different forms depending on the content of crystallization water.

Gypsum is a white mineral that, depending on the degree of purity and the type and quantity of impurities present, can take on shades that vary from gray to reddish [55].

If anhydrous calcium sulphate is present in a quarry, the extracted material will not require subsequent crushing to be reduced to powder—a process which must be carried out in the case of dihydrated sulphate. Furthermore, in the case of bihydrated sulphate, following grinding, there is a cooking phase for the total or partial elimination of the crystallization water present in the mineral, which determines the technical characteristics and methods of use of the gypsum.

During cooking, once it reaches 128 °C, the chalky rock loses approximately 6% of its crystallization water: this forms a semi-hydrated calcium sulphate ( $\text{CaSO}_4 \cdot (1/2)\text{H}_2\text{O}$ , which has a setting time which varies between 1 and 4 min), which when it comes into contact with water transforms back into the starting material.

Between 150 °C and 180 °C, the crystallization water is completely eliminated and the soluble anhydrite  $\text{CaSO}_4$  ( $\alpha$ ) begins to form, the setting time of which is approximately 20 min: above 180 °C, the speed of this transformation is increased significantly. If cooking is carried out above 250 °C, insoluble anhydrite  $\text{CaSO}_4$  ( $\beta$ ) is formed. At higher temperatures, the formation of insoluble anhydrite continues, and at 1200 °C,  $\text{CaO} + \text{SO}_3$ , i.e., calcium monoxide (quicklime) and sulfuric anhydride, is formed.

After cooking, there is a second grinding process necessary to obtain a homogeneous product that is easily usable and which guarantees perfect implementation. Generally, the cooked gypsum is finely ground, with grain sizes between 150 and 600  $\mu\text{m}$ .

The construction plasters found on the market are made up of semi-hydrated plaster or soluble anhydrite or a mixture of them.

The wall plaster used in this study was a powdered product based on hemihydrate gypsum classified according to the European standard EN 13279-1 [56] as A1/A2/A3/7.

Table 1 shows the technical data sheet characteristics of the wall gypsum used to produce the samples.

**Table 1.** Technical data sheet characteristics of the wall gypsum.

Technical Features	
Appearance	White powder
Powder specific gravity	$\approx 650 \text{ kg/m}^3$
Granulometry	$\leq 0.3 \text{ mm}$
pH	$\approx 7$
Consumption	Variable, depending on use
Seizing time	$\approx 7 \text{ min}$
Workability time	$\approx 6 \text{ min}$
Drying time	$\approx 8 \text{ days}$
Insulation from direct airborne noise	NPD
Heat resistance	NPD
Fire resistance	Class A1
Mixing water	$\approx 50\%$ ( $\approx 16.5 \text{ l}$ per 25 kg)

Legend: NPD: no performance declared. Data expressed at  $22 \pm 1 \text{ }^\circ\text{C}$  with relative humidity at  $50 \pm 5\%$ . Lower temperatures lengthen maturation and hardening times; higher temperatures reduce maturation and hardening times.

### 2.3. Pistachio Shells

According to FAOstat (2020) data [57], in 2018, 90% of pistachios were produced by the United States, Turkey and Iran, with a production of 1,239,007 tons/year. In Italy, in total, around 300 thousand tons of nuts are produced per year, and, according to ISTAT data from 2017 [58], pistachio areas in Italy do not exceed 4000 hectares, for a production of just under 4 thousand tons, concentrated almost exclusively in Sicily. At present, pistachio

shells do not have an industrial use or significant economic value, and this is why they are burned or disposed of in landfills [59].

The pistachio shell makes up between 51% and 69% of the weight of the fruit [60]. Furthermore, the weight loss of the pistachio shell is approximately 75–80% during the heating phase due to its high cellulose structure [61,62]. Pistachio shells are a biomass composed mainly of cellulose, hemicellulose and lignin; they have a rather regular shape; and the size is close to that of the pellet. The pistachio shells used in this work were ground using a high-power electric grinder (2.5 kW) with a high number of laps due to their high elasticity and mechanical resistance: 36,000 r/min for 1 min. Following the grinding process, the product obtained was sieved in order to separate the different grain sizes obtained. Three different grain sizes were produced:

- Pg pistachio shells with dimensions ranging from 2 to 4 mm;
- Pf pistachio shells with dimensions of 0.5–2 mm;
- Ps pistachio shells with dimensions < 0.5 mm.

The products obtained from grinding are shown in Figure 2.



**Figure 2.** Pistachio shell grain sizes: (a) Pg shells with dimensions ranging from 2 to 4 mm; (b) Pf shells with dimensions of 0.5–2 mm; (c) Ps shells with dimensions <0.5 mm. Figures report the metric reference to 1 cm.

#### 2.4. Sample Preparation and Mix Design

As regards the preparation of the specimens and the production procedures, a mixer compliant with the UNI EN 13279-2 standard [53] was used which was equipped with a steel cup and a mixing blade connected to an electronic control system for speed and rotation.

To determine the consistency of the fresh mortar, the shake table method was adopted in accordance with UNI EN 13279-2 [53]. The test determines the consistency of mortar by measuring its spreading. The equipment consists of a circular table with a shaft mounted on a sturdy frame through which it is possible to apply vertical shocks of standard energy defined by the stroke of the shaft.

The mortar is positioned inside a bronze cone, placed in the center of the circular table and compacted with a pestle to settle it in the truncated cone specimen, ensuring uniform filling. Once the cone has been raised after a period of between 10 and 15 s, 15 shocks are applied in 15 s, and at the end, the spreading of the mortar, i.e., its diameter expressed in millimeters, is measured. For the good workability of the mortar so that it can be used for plasters, it is necessary to adopt a spreading which can vary from 160 to 165 mm (with a tolerance of  $\pm 5$  mm), as prescribed by the UNI EN 13279-2 standard [53].

According to the above-mentioned standard and according to the classification of the analyzed mortar, since it is a masonry plaster, for all the mix designs created, we stuck to spreading values between 160 and 165 mm.

The mixtures were prepared in steel molds, the walls of which were appropriately treated with a release agent. Once an adequate degree of resistance was reached, the specimens were removed from the molds. They were subsequently placed in a curing environment maintained at a temperature of  $23 \pm 2$  °C and a relative humidity of no less than  $50\% \pm 5\%$  for 7 days, as in real operating conditions.

Subsequently, they were dried to constant mass at a temperature of  $40 \pm 2$  °C. After drying, the samples were allowed to cool to room temperature.

All the samples were classified with a characteristic nomenclature, and the samples that underwent an aging process and on which tests were carried out before and after exposure to physical–mechanical and hydrothermal stress conditions were cataloged and filed.

To evaluate each mix, the samples shown in Table 2 were created.

**Table 2.** List of samples made with their relative dimensions and the tests carried out on each of them.

Sample	Dimension	Test Type
GPsPg	16 × 4 × 4	Mechanical and water absorption tests
	10 × 10 × 0.5	Adhesion tests
	4 × 4 × 4	Mechanical compression and water absorption tests
	20 × 20 × 2	Thermal conductivity tests
GPgPflfS	16 × 4 × 4	Mechanical and water absorption tests
	10 × 10 × 0.5	Adhesion tests
	4 × 4 × 4	Mechanical compression and water absorption tests
	20 × 20 × 2	Thermal conductivity tests
GSPf	16 × 4 × 4	Mechanical and water absorption tests
	10 × 10 × 0.5	Adhesion tests
	4 × 4 × 4	Mechanical compression and water absorption tests
	20 × 20 × 2	Thermal conductivity tests
GSPf.2	16 × 4 × 4	Mechanical and water absorption tests
	10 × 10 × 0.5	Adhesion tests
	4 × 4 × 4	Mechanical compression and water absorption tests
	20 × 20 × 2	Thermal conductivity tests
GPsLfS.2	16 × 4 × 4	Mechanical and water absorption tests
	10 × 10 × 0.5	Adhesion tests
	4 × 4 × 4	Mechanical compression and water absorption tests
	20 × 20 × 2	Thermal conductivity tests
GPsLfS.3	16 × 4 × 4	Mechanical and water absorption tests
	10 × 10 × 0.5	Adhesion tests
	4 × 4 × 4	Mechanical compression and water absorption tests
	20 × 20 × 2	Thermal conductivity tests

Legend: G: gypsum; S: sand; Pg: large pistachio shells; Pf: medium pistachio shells; Ps: fine pistachio shells; Lf: sawdust.

Some of the samples made are shown in Figure 3.

The compositions of the mix designs listed above are shown in Table 3.

To evaluate the adhesion of the mixtures, they were applied on tiles measuring  $10 \times 10 \times 0.5$  cm, as shown in Figure 4.

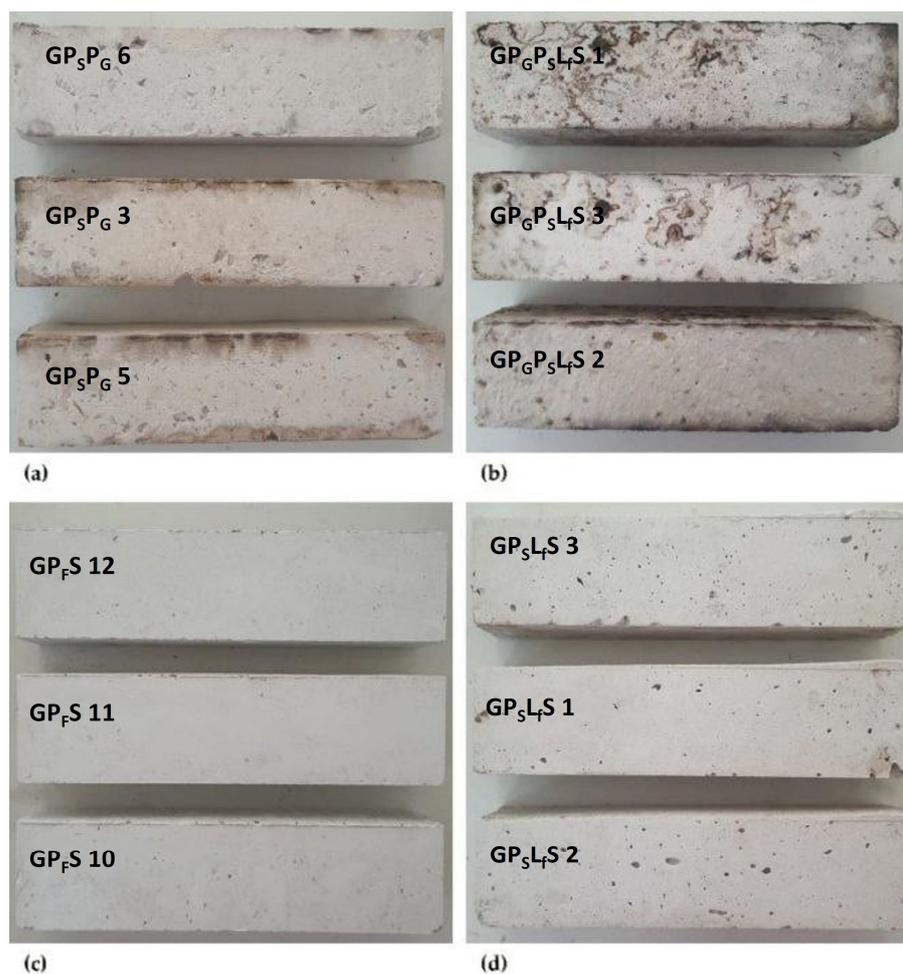


Figure 3. Example of samples made for testing: (a) GPsPg; (b) GPgPflS; (c) GSPf; (d) GPslfS.2.

Table 3. Compositions of the created mix designs.

Mix Design	Composition	Weight [g]
GS	Gypsum (G)	940
	Sand (S)	60
	Water	700
GLf	Gypsum (G)	925
	Sawdust (Lf)	75
	Water	700
GLf.2	Gypsum (G)	800
	Sawdust (Lf)	200
	Water	700
GSLf	Gypsum (G)	800
	Sand (S)	150
	Sawdust (Lf)	75
GSLf.2	Water	700
	Gypsum (G)	800
	Sand (S)	50
GPsPg	Sawdust (Lf)	150
	Water	700
	Gypsum (G)	600
	Pistachio shells (Pg) (2–4 mm)	260
	Pistachio shells (Ps) (<0.5 mm)	140
	Water	500

Table 3. Cont.

Mix Design	Composition	Weight [g]
GPgPfLfS	Gypsum (G)	700
	Pistachio shells (Pg) (2–4 mm)	80
	Pistachio shells (Pf) (0.5–2 mm)	80
	Sawdust (Lf) (<0.5 mm)	80
	Sand (0.075–0.6 mm)	60
	Water	600
GSPf	Gypsum (G)	800
	Pistachio shells (Pf) (0.5–2 mm)	150
	Sand (S) (0.075–0.6 mm)	50
GSPf.2	Water	700
	Gypsum (G)	941
	Pistachio shells (Pf) (0.5–2 mm)	94
GPsLfS.2	Sand (S) (0.075–0.6 mm)	59
	Water	700
	Gypsum	800
	Pistachio shells (Ps) (<0.5 mm)	75
	Sawdust (Lf) (<0.5 mm)	75
GPsLfS.3	Sand (S) (0.075–0.6 mm)	50
	Water	700
	Gypsum (G)	790
	Pistachio shells (Ps) (<0.5 mm)	75
	Sawdust (Lf) (<0.5 mm)	75
	Sand (S) (0.075–0.6 mm)	60
	Water	700

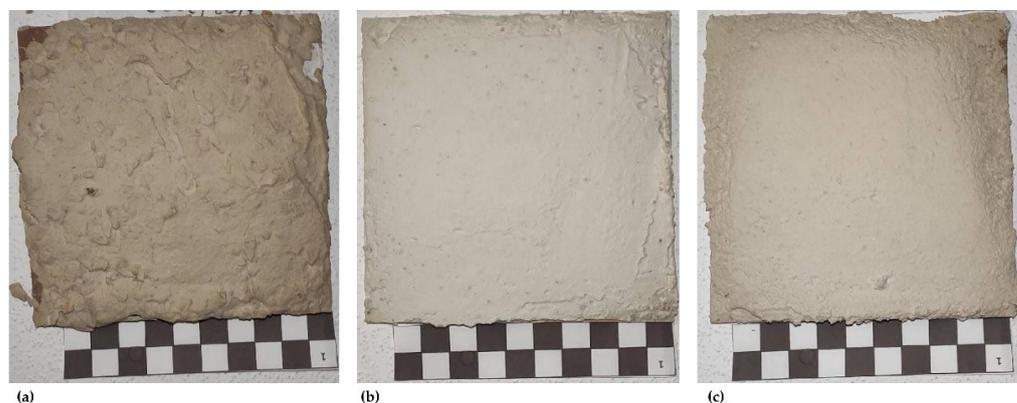


Figure 4. Examples of mortars spread on tiles: (a) GPsPg; (b) GSPf; (c) GPsLfS.2. Figures report the metric reference to 1 cm.

### 2.5. Tests Carried Out

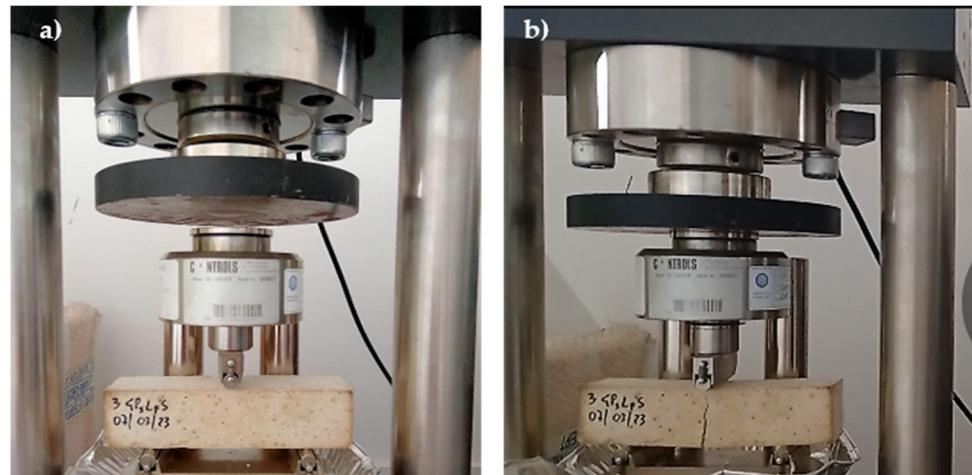
The produced samples were subjected to different tests aimed at characterizing the new mix designs and evaluating the eventual performance improvements compared to the mortars that were not prepared with additives. The mechanical resistance and the water absorption were evaluated, as well as the maintenance of the obtained results after exposure to artificial ageing.

Moreover, in order to evaluate the possible contribution of the additives to improving the energy performance of buildings, measurements were carried out for the evaluation of thermal conductivity.

### 2.5.1. Mechanical Properties

Following the 7-day curing period, all the specimens were subjected to compressive and flexion strength tests as prescribed by the UNI EN 13279-2 standard [53].

Figure 5 shows an example of a flexion test performed on the samples.



**Figure 5.** Example of a flexion test: (a) photo before running the test; (b) photo after running the test.

### 2.5.2. Water Absorption Tests

The water circulation tests by capillarity absorption were carried out before and after the artificial aging tests in order to verify both the ability of each mortar mix design to absorb water and to verify how this property was maintained over time.

The water absorption/desorption test by capillarity, as defined by the UNI 10859:2000 standard [63], is based on the measurement of the increase in the mass of a specimen of stone material in contact with deionized water per unit of surface as a function of time.

For the test, 3 samples measuring 16 cm  $\times$  16 cm  $\times$  4 cm of each type of mortar mix design were examined.

After the complete drying of the samples, they were placed in a basin, kept away from the base thanks to filter paper discs, and immersed in water to a depth of 5 mm. The water level was kept constant during the entire test.

The specimens were weighed, after swabbing with a damp cloth, at different time intervals: 10, 20, 30, 60, 240, 360, 1440, 2880 and 4320 min, with the time intervals adjusted according to the recommendations in the standard UNI 10859:2000 [63] for stone materials to assess gypsum behavior. From the weight values, it was possible to obtain the coefficient of water absorption by capillarity, which corresponds to the slope of the straight line that joins the points representing the measurements carried out at the time intervals. The test for calculating the imbibition coefficient was carried out in compliance with the CNR BU standard n. 137/92 (*Regulations on aggregates—Determination of the imbibition coefficient*) [64], and the calculation of the imbibition coefficient used the following formula:

$$C_{i_i} = \frac{m_{1,1} - m_{2,1}}{m_{2,1}} \cdot 100$$

where

- $m_{1,i}$  indicates the mass expressed in grams of wet material for the reference specimen,  $i$ ;
- $m_{2,i}$  indicates the mass expressed in grams of dry material for the reference specimen,  $i$ .

Figure 6 shows an example of a capillary water absorption test.



**Figure 6.** Water absorption test by capillarity.

### 2.5.3. Thermal Conductivity Measurements

Thermal conductivity measurements were carried on in order to compare the thermal properties of the various mix designs and verify whether the addition of biobased aggregates brought about improvements in terms of thermal insulation. The tests were conducted on samples measuring  $20\text{ cm} \times 20\text{ cm} \times 2\text{ cm}$  using the ISOMET 2114 thermofluximeter, from Applied Precision s.r.o., Bratislava, Slovakia (Figure 7).

The instrument is used to determine the thermal properties of different isotropic materials, including insulating ones, and is widely used in the literature for the determination of the thermal conductivity of building materials [65–69]. The ISOMET 2114 is a commercial portable microprocessor-controlled instrument for direct measurement of the thermal conductivity coefficient of materials and is equipped with two types of measurement probes: a needle probe (for unconsolidated and composite materials) and a surface probe (for solid consolidated and composite materials).

In principle, the time dependence of the thermal response to the impulse transmitted by the heat flow in the material to be measured is analyzed. The heat flow is generated by the electrical energy dissipated through the probe, which is in direct contact with the measured material being processed. The resistance-dependent temperature is detected by a semiconductor sensor, and time variation in the temperature is sampled at discrete points (the regression polynomials passing through the samples are constructed using the “least squares method”, and the coefficients of the relevant regression polynomials allow the analytical calculation of required parameters). For a measurement ranging from 0.015 to 0.700 W/m·K, the ISOMET 2114 thermofluximeter guarantees an overall relative measurement uncertainty  $\approx 5\%$  [65].

For each specimen, 10 measurements were performed, using the surface probe and a measurement range of 0.04–0.3 W/m·K, with a built-in memory and calibration constants stored in the memory.



**Table 4.** Exposure to high-humidity conditions.

Humid Conditions	Time	Drying of Samples
90% Salt spray/humidity cabinet, 100 lt	7 days	Drying in the oven at 40 °C for 2 days
90% Salt spray/humidity cabinet, 100 lt	7 days	Placed in the freezer at −20 °C for 2 days and subsequently dried in the oven at 40 °C for 2 days

A summary of all adopted methods and instruments is reported in Table 5.

**Table 5.** List of adopted methods and instruments.

Procedure Method	Instrument	Reference
Realization of samples	Portable High-Speed Grinder, 220 V–2.5 kW–36,000 rpm, Matest SpA, Treviolo, Italy	None
Shake table method	Shake table Matest Ref. E090-05/BZ/0035, Matest SpA, Treviolo, Italy	UNI EN 13279-2:2014 [53]
Testing Method	Instrument	Reference
Compressive and flexion strength tests	Controls–Model 65-L0019/8	UNI EN 13279-2:2014 [53]
Water absorption/desorption test	Measurement of the change in mass of the mortar sample ISOMET 2114	UNI EN 1015-18:2004 [63,70]
Thermal conductivity measurement	thermofluximeter, Applied Precision s.r.o., Bratislava, Slovakia	UNI ISO 9869-1:2015 [71]
Aging test	Salt spray/humidity cabinet, 100 lt; Ref. C & W Specialist Equipment Ltd. SF/AB100767796, Specialist Equipment Solutions Ltd., Aberdeen, UK	None

### 3. Results and Discussion

#### 3.1. Mechanical Properties before and after Aging

All samples were subjected to bending and compression tests as prescribed by the UNI EN 13279-2 standard [53]. Table 6 summarizes the results obtained from the tests carried out and the identification of the specimens on which they were carried out.

**Table 6.** Results of mechanical tests before aging.

Mix Design	Water/Gypsum Ratio	Compression Test	
		KN	MPa
GS	0.74	4.95	2.56
GLf	0.87	6.80	4.24
GLf.2	0.87	1.81	1.13
GSLf	0.87	5.48	3.42
GSLf.2	0.87	3.45	2.16
GPsPg	0.83	3.27	2.04
GPgPflfS	0.86	2.30	1.44
GSPf	0.75	6.60	4.14
<b>GSPf.2</b>	<b>0.74</b>	<b>10.75</b>	<b>6.69</b>
GPslfS.2	0.88	6.63	4.13
GPslfS.3	0.89	7.78	4.86

Mechanical tests were also carried out by changing only the water/gypsum ratio of the mortar to verify how the mechanical properties of the material varied. Below is the table reporting the results of the tests.

In general, it was noted that most of the biobased mix designs exhibited increased compression resistance compared to a regular gypsum mortar plaster without biobased aggregates (GS), with values of around 3 MPa, the mechanical performance being improved due to the elastic behavior of the pistachio shells.

The presence of sand in the mix designs, especially in GSPf.2, GSPf and GPsLfS.3, appeared to contribute to the increased mechanical resistance of the mortar, as the greatest granulometry variety increased the compactness and the better distribution of mechanical forces.

As regards the GPsPg sample, no further tests were carried out following the aging, as the sample showed the visible presence of mold on the surface.

The results relating to the post-aging mechanical tests are shown in Table 7, with best results in bold.

**Table 7.** Results of mechanical tests after aging.

Mix Design	Water/Gypsum Ratio	Compression Test after Oven		Compression Test after Freezer	
		KN	MPa	KN	MPa
GS	0.74	4.80	2.50	4.65	2.40
GLf	0.87	6.70	4.20	6.55	3.94
GLf.2	0.87	1.74	1.10	1.58	0.81
GSLf	0.87	5.43	3.35	5.24	2.83
GSLf.2	0.87	3.41	2.12	3.14	1.87
GPgPflS	0.86	2.20	1.40	2.04	1.27
GSPf	0.75	7.22	4.51	7.03	4.39
<b>GSPf.2</b>	<b>0.74</b>	<b>10.95</b>	<b>6.81</b>	<b>10.7</b>	<b>6.4</b>
GPsLfS.2	0.88	6.88	4.29	6.62	4.10
GPsLfS.3	0.89	6.88	4.29	7.74	4.84

Considering the results obtained from the mechanical tests, it was noted that aging caused no evident reduction in the mechanical compressive strength of the different materials. It was noted that the mix design that presented the best results was GSPf.2, made up of gypsum, sand and finely ground pistachio shells, this design obtaining higher compressive strengths. The best results were due to the better ratio between the aggregates, the presence of ground pistachios and sand, and a balanced water and chalk ratio.

Finally, it could be interesting to compare these mechanical resistance data with those of other mixtures of biomass aggregate mortars present in the literature to evaluate the validity of the tested mortar, although it should be underlined that there are substantial methodological differences between studies. For instance, in [51], the authors tested different samples made of gypsum (95–97.5% of weight) and crushed straw (2.5–5.0%), obtaining high variability in terms of compressive strength values (3.09–14.93 MPa). Anyway, it can be noted that the values in the previous study were generally higher than those in this study due to a higher percentage of gypsum than that adopted in the present case (40–55%).

### 3.2. Water Absorption Tests

The results relating to the capillarity absorption tests carried out before the aging tests highlight different imbibition capacities for the mortars examined. This behavior could be attributable to the different compositions of the mortars corresponding to the presence of aggregates with different grain sizes. The water absorption dynamics of the various samples present an almost similar trend for all the specimens examined. For the various types of mortar, there was a rapid increase in the quantity of water absorbed in the first 240 min. For all samples, the test ended after 4320 min, as the mass appeared to have

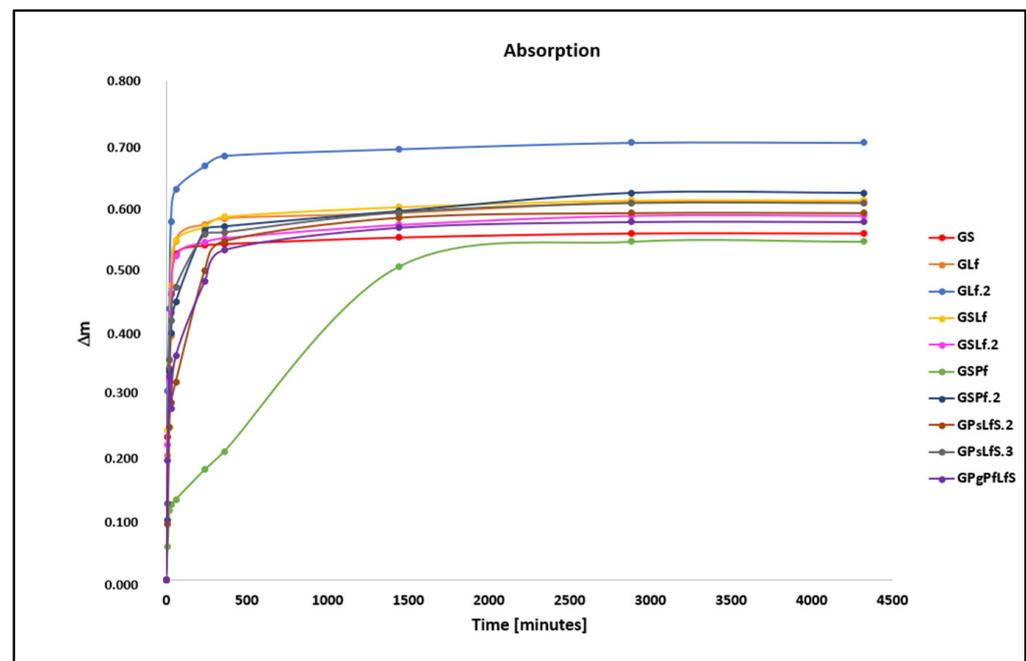
stabilized by this point. This was due to the fact that the common matrix was gypsum, a material that tends to be very water absorbent.

Table 8 reports the imbibition coefficient values for the various tested samples, with best results in bold.

**Table 8.** Imbibition coefficient values for the various samples tested.

Mix Design	Imbibition Coefficient
GS	45.54
GLf	48.42
GLf.2	80.80
GSLf	50.17
<b>GSLf.2</b>	<b>61.11</b>
GPgPflfS	41.93
GSPf	44.09
GSPf.2	44.01
GPslfS.2	48.72
GPslfS.3	51.93

Figures 9 and 10 show, respectively, the trends of the absorption and desorption curves for the various mix designs tested. The desorption curves were obtained by weighing the samples in the same time intervals used for the absorption test to check how long it took the samples to reach a constant weight. The samples reached a constant weight after approximately 2880 min. The best results for the GSPf sample were due to the greater presence of finely ground pistachios (150 g) and sand, which made the material more compact so that it did not absorb water, as well as the absence of sawdust, which in the other samples contributed to their greater hygroscopicity.



**Figure 9.** Absorption curves.

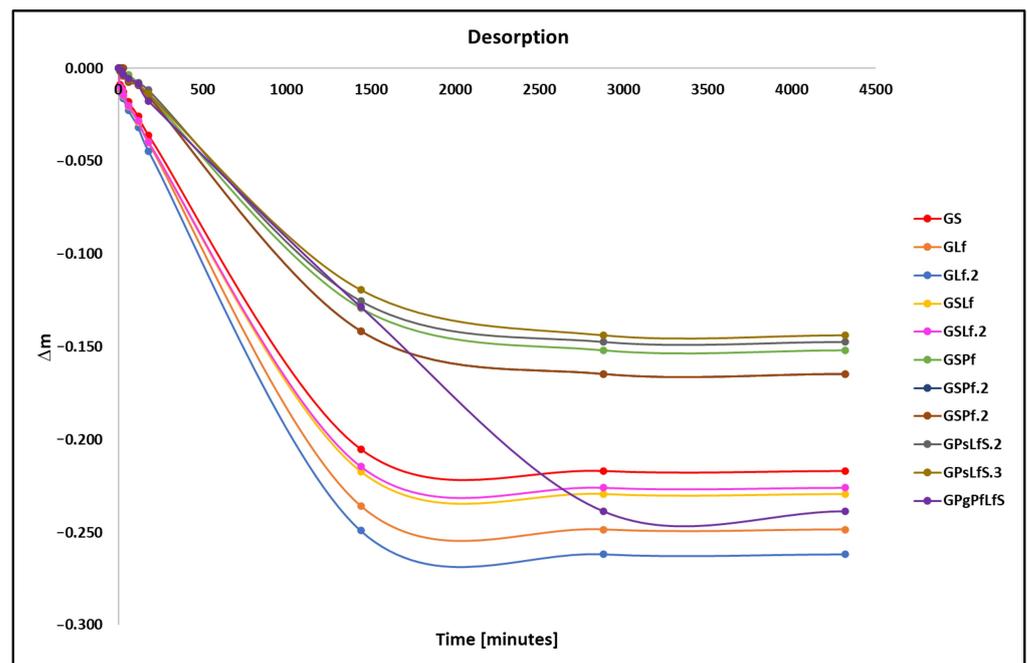


Figure 10. Desorption curves.

### 3.3. Thermal Conductivity Measurements

In general, it was noted that all the biobased mix designs reduced the thermal conductivity by over 40% compared to a regular gypsum mortar plaster without biobased aggregates, with lambda values of around 0.5–0.7 W/m·K; the thermal insulation performance was improved, as pistachio shells have a thermal insulating behavior similar to wood. In general, highly improved thermal conductivities were obtained by mixing gypsum and pistachio shells, as all values were in the range of 0.1–0.3 W/m·K, while the tested gypsum mortar without additives had a 0.53 W/m·K thermal conductivity. This range of values is comparable with the thermal conductivity values of some wood panels and insulating bricks or mortars products, as reported by Casaclima [72]. The sample that showed a lower value was GPgPflfs, composed of pistachio shells and sawdust with a grain size between 4 and 2 mm, which exhibited a seriously lower thermal conductivity of 0.152 W/m·K, which would really make a difference were it not for the fact that its mechanical resistance was also reduced compared to original gypsum plaster.

Even if plasters are applied with a small thickness so their general impact in terms of energy saving is much lower than that of structural and insulating building layers, combining these two materials shows promising prospects in the building sector. The developed mortar is even more interesting when one considers the expected associated low environmental impact thanks to its production under the circular economy paradigm, which could be investigated in further studies.

The results relating to the thermal conductivity tests are summarized in Table 9.

Table 9. Average thermal conductivity values for the various samples tested.

Mix Design	Average Thermal Conductivity [W/m·K]
GS	0.531
GLf	0.106
GLf.2	0.110
GSLf	0.191
GSLf.2	0.156
GPgPflfs	0.152

Table 9. Cont.

Mix Design	Average Thermal Conductivity [W/m·K]
GSPf	0.231
GSPf.2	0.277
GPsLfS.2	0.223
GPsLfS.3	0.214

#### 4. Conclusions

Six different mix designs of plaster mortars based on gypsum and ground pistachio shells were comparatively tested, and their performances were evaluated in terms of mechanical resistance, thermal conductivity, imbibition capacity and durability over time. Considering the results obtained from the mechanical tests, it can be stated that almost all the mix designs containing pistachio shells showed an increase in mechanical resistance compared to the mortar without additives, probably due to the characteristic high mechanical resistance and elasticity of the shells.

Moreover, it seems that aging did not cause variations in the mechanical compressive strength of the various mix designs, which maintained good mechanical resistance values: this demonstrates that the thermal stresses induced with aging do not cause loss of material, decohesion or intergranular desegregation, which could have reduced the mechanical performance in terms of compression.

As regards the capillarity absorption tests, the best behavior was shown by the imbibition dynamics of the GSPf sample, which differed from the others in that the absorption curve showed a greater slowness in absorbing water and, compared to all the other samples, an overall lower absorption capacity. These improvement data are very significant as they show that one of the most renowned problems with gypsum-based materials, namely, their high hygroscopicity, can be overcome. From the results obtained, it is possible to state that the best results in terms of mechanical performance and water absorption by capillarity were achieved with the use of pistachio shells with a grain size between 2 and 0.5 mm, in particular, GSPf.

For all the other samples tested, thermal conductivity values between 0.21 and 0.28 W/m·K were measured, confirming that all the biobased mix designs were characterized by a reduction in thermal conductivity, and the thermal insulation performance was also improved compared to the plaster without pistachios grains.

Ultimately, considering all the results obtained, it is possible to say that, on average, the best results were obtained with the use of pistachio shells with a grain size between 0.5 and 2 mm with sand and both with water/gypsum ratios of 0.86 and 0.74.

The research activities carried out demonstrate that the use of waste materials from the agro-food chain not only contributes to circular economy policies but can increase the performance of traditional mortars, reducing the need to use synthetic products and promoting eco-sustainability.

Sandoval et al. [73] reported that biobased materials are increasingly attractive for many applications, including construction, due to their lower cost, abundance, biodegradability and lower related greenhouse gas emissions, but that assessments of their processing costs and efficiency are lacking. To encourage the industrialization phase, further research could enrich this study with analysis of other properties, like durability, fire resistance and sound adsorption, and production chains and costs.

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