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Article

Thermal and Acoustic Performance of Gypsum Plasters Mixed with Different Additives: Influence of Bio-Based, Synthetic, and Mineral Fillers

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Abstract: Due to the high impact of the building sector on the environment, a growing interest focuses on insulating materials able to ensure good thermo-acoustic performance for the building envelope, in a sustainable and circular economy perspective. In this context, Moroccan natural gypsum was mixed with natural waste local materials. Thermal and acoustic properties of the samples were measured; they were compared to those of synthetic- and mineral-based gypsum plasters, manufactured with the same technique. A Small Hot Box apparatus was used for thermal characterization, whereas acoustic performance was investigated by means of a Kundt’s Tube. Natural and synthetic additives involved a reduction in density and an improvement in thermal performance. Conductivity values in the 0.181 – 0.238 W/mK range were obtained, depending on the type of natural additive, with respect to 0.275 – 0.323 W/mK of mineral-based gypsum plasters. The acoustic measurements showed that all the composites have similar performance in terms of acoustic absorption, whereas high Transmission Loss values were obtained for the natural additives (TL = 35 – 59 dB). Petiol of Palm and Stipa Tenacissima were found as the materials able to improve both thermal and acoustic properties.

Keywords: Moroccan gypsum; natural additives; synthetic and mineral additives; bio-based gypsum plasters; thermal conductivity; acoustic performance

1. Introduction

The building sector is one of the largest consumers of energy in the world and generates greenhouse gases and CO₂ emissions [1]. This prompts the scientific community to develop different solutions and strategies to solve the problem and bring significant improvements in the construction industry, taking effective and sustainable actions against climate change. Therefore, reducing the impact of the building sector has become a key point in the sustainable development of building materials, to develop high-performance thermal and acoustic insulation biomaterials able to improve the comfort of buildings reducing the environmental impact. In this context, bio-composite materials are a first step towards the integration of an ecological, green, and sustainable approach. The incorporation of vegetable aggregates in building materials resulted in several benefits in addition to the environmental impact reduction. Positive effects of bio-composites can be obtained in terms of thermal insulation, mechanical, and hygro-thermal properties [2–6].

Plasters have an important role in the opaque building envelope performance. In the literature several studies focus on the addition of natural or waste materials in plasters, resulting in a great environmental benefit, although they are characterized by a slightly improvement in thermal and acoustic performance with respect to conventional solutions [7–15]. Gypsum plaster is one of the most standard materials used by many ancient civilizations in construction applications, due to its aesthetic and thermo-acoustic qualities [16]. Gypsum plaster is an economical and easily exploitable

product, offering a good thermo-acoustic comfort, as well as its protection against fire (Lushnikova & Dvorkin, 2016). Several authors tried to improve its physical, thermal, mechanical, and hygrothermal properties by adding additives. Among the synthetics, polystyrene was widely used. A complete study of the mechanical, thermal, and acoustic properties of gypsum-plasters mixed with EPS was carried out by Bouzit et al [17], taking into account the different contents and diameters of the polystyrene balls. By increasing the size of the beads, the maximum value of flexural strength occurred with a lower percentage of polystyrene. The thermal values were 0.191 and 0.116 W/mK for 10% and 30% weight content of smaller polystyrene beads (3-mm diameter), respectively. Transmission loss values ranged up to 45 dB, with a reduction of only 2-3 dB compared to pure samples, due to the reduction of density. Similar results were found in Bicer et al. [18] for a composite material based on expanded polystyrene and tragacanth gum in different percentages.

Better thermal performance was obtained in plasters with the incorporation of granular aerogel, reaching values of thermal conductivity of 0.05 W/mK (90% weight of granules in the mixture) [19–21].

Over the years, researchers have worked on numerous gypsum plasters solutions with natural, vegetable, and recycled fibers additives. For instance, Khalil et al. [22] showed good reinforcing and thermal insulation properties of rice husk. *Pennisetum Setaceum* (a herbaceous plant) fibers were of interest for lightweight thermal insulating materials [23]. A small amount of date palm fibers (6% by weight) resulted in a thermal conductivity reduction of about 48% [24]. The addition of 2% by weight of Alfa fibers increased thermal performance of bio-sourced earth-based composites, as demonstrated by Charai et al. [25]. *Ferula Communis* collected from Ait Ishaq city (Morocco) had a positive impact on both the thermal performance and the lightness of the dough [16]. For a mass fraction of additive ranging from 0 to 8%, the thermal conductivity decreased from 0.372 to 0.263 W/mK (reduction of about 29%). The study of Saira et al. [26] contributed to the development of a gypsum composite with cork fibers and cardboard waste, showing the positive influence of increasing cork and reducing paper contents on thermal conductivity values. The good properties of cork from thermal and acoustic point of view were already confirmed in previous studies [27,28]. The size and content of peanut shells affected the thermal behavior of plasters; the thermal conductivity value was reduced by more than 50% with 20% of additive, characterized by a diameter in the 2.5 – 5 mm range [29]. Hemp and Alfa fibers had positive effects also on the sound absorption performance [30,31]. Wood waste improved thermo-acoustic properties of gypsum composites [32]. Straw is one of the most used additives: when added only 4 wt.% to the gypsum plaster, reductions in thermal conductivity and in greenhouse gas emissions up to respectively 68% and 13% were achieved in different climate zones of Morocco [33]. However all these additives resulted in deterioration of the mechanical performance.

In Morocco, there are huge extensive high-quality gypsum ores and the use of different additives to improve the thermal performance of gypsum-based plasters is the subject of a research collaboration between University of Perugia and University of Agadir since several years. Based on the preliminary results obtained in terms of physical, thermal, and acoustic properties [34], gypsum-plaster from the Safi region (South-western Morocco) was used to manufacture composites with different additives. Natural fibers were collected in the local area to fabricate bio-based gypsum plasters. In the present paper, thermal and acoustic performance of these mixtures are measured and discussed. A comparison with those obtained for plasters with the most common synthetic and mineral fiber additives is also carried out. The aim of the study is to identify the most promising solutions from a thermo-acoustic point of view.

2. Materials and Methods

Natural gypsum powder from the rocks of the Safi region (Morocco) was used to assemble the samples. Its features were deeply discussed in [17,34]. The powder (p) was mixed with water (w) (mass ratio w/p equal to 0.8) in about 90 s, in order to obtain a fluid and homogenous porous plaster, without any air bubbles. The paste was added with different components and mixed by means of a conventional stirrer Ika Rw 20 (helical moves for about 3 minutes with a speed equal to 100 rpm). The selected additives are of the following three types (Figure 1):

- synthetic: Polyester Fiber (PF), Paraffin (Pa), and Glass Wool (GW), selected among the most used commercial ones;
- natural: Stipa Tenacissima (ST) or Alfa fiber, Palm Petiole (PPa), Sisal Fiber (SF), Straw (Sw), Corn Bristles (CB), Cork (Ck), and Chip-Sawdust (ChSd), very diffused in the local Moroccan area;
- mineral: Cement (Ce), Clay (Cl), Sand (Sa), and Refractory Cement (R.Ce).



Figure 1. Safi gypsum powder and synthetic, natural, and mineral additives used in the samples.

Natural fibers were collected and shredded by hand, obtaining a homogeneous size of about 1 cm. Several tests were carried out to identify the optimal additive value among the different types to obtain stable samples; it was found to be equal to 20 wt.% of the total volume. Also a reference panel with pure hardened gypsum plaster and water (called PS 100) was assembled.

The mixture was placed in molds purposely made and left under the controlled laboratory conditions (relative humidity equal to 70% and temperature in the 23 – 25°C range) for about 3 hours. Once removed, the samples were dried in a climatic chamber at 45 ° C for 2 days, to eliminate all the interstitial water. For each composite, three square-shaped panels of 300x300 mm² dimensions and cylindrical specimens of 100- and 29-mm diameter were fabricated, in order to investigate the thermal and acoustic properties, respectively (Table 1). Despite the use of molds, the thickness of the samples was not the same, due to their different shrinkage during the drying step; the thickness of the square panels was in the 0.023-0.030 m range, slightly smaller than that of the cylindrical ones (about 0.040-0.060 m). For the acoustic tests, only specimens with similar thicknesses and density ρ were considered in the analysis, for a reliable comparison.

Table 1. Features of the examined samples.

Code	Composition	Type of additive	Dimension of samples	Thickness [m]	Density [kg/m ³]
PS 100	Safi Plaster 100%	---	0.3 x 0.3 m ²	0.030	960
			ϕ 100 mm	0.050	920
			ϕ 29 mm	0.054	960
PS 80-PF 20	Safi Plaster 80% - Polyester Fiber 20%	synthetic	0.3 x 0.3 m ²	0.025	1022
			ϕ 100 mm	0.045	1032
			ϕ 29 mm	0.047	957

PS 80-Pa 20	Safi Plaster 80% - Paraffin 20%	natural	0.3 x 0.3 m ²	0.030	932
			φ 100 mm	0.050	930
			φ 29 mm	0.056	920
PS 80-GW 20	Safi Plaster 80% - Glass Wool 20%		0.3 x 0.3 m ²	0.026	871
			φ 100 mm	0.040	930
			φ 29 mm	0.039	935
PS 80-ST 20	Safi Plaster 80% - Stipa Tenacissima 20%		0.3 x 0.3 m ²	0.026	900
			φ 100 mm	0.049	959
			φ 29 mm	0.052	818
PS 80-PPa 20	Safi Plaster 80% - Petiol of Palm 20%		0.3 x 0.3 m ²	0.026	905
			φ 100 mm	0.051	999
			φ 29 mm	0.045	818
PS 80-SF 20	Safi Plaster 80% - Sisal Fiber 20%		0.3 x 0.3 m ²	0.027	1098
			φ 100 mm	0.049	1073
			φ 29 mm	0.060	923
PS 80-Sw 20	Safi Plaster 80% - Straw 20%		0.3 x 0.3 m ²	0.029	810
			φ 100 mm	0.050	869
			φ 29 mm	0.052	841
PS 80-CB 20	Safi Plaster 80% - Corn Bristles 20%		0.3 x 0.3 m ²	0.028	969
			φ 100 mm	0.042	1050
			φ 29 mm	0.046	920
PS 80-Ck 20	Safi Plaster 80% - Cork 20%		0.3 x 0.3 m ²	0.028	845
			φ 100 mm	0.049	812
			φ 29 mm	0.049	930
PS 80-ChSd 20	Safi Plaster 80% - Chips- Sawdust 20%		0.3 x 0.3 m ²	0.023	1069
			φ 100 mm	0.043	940
			φ 29 mm	0.045	860
PS 80-Ce 20	Safi Plaster 80% - Cement 20%		0.3 x 0.3 m ²	0.024	1329
			φ 100 mm	0.043	1278
			φ 29 mm	0.042	1236
PS 80-CI 20	Safi Plaster 80% - Clay 20%	0.3 x 0.3 m ²	0.027	1182	
		φ 100 mm	0.047	1235	
		φ 29 mm	0.049	1114	
PS 80-Sa 20	Safi Plaster 80% - Sand 20%	0.3 x 0.3 m ²	0.026	1266	
		φ 100 mm	0.046	1189	
		φ 29 mm	0.048	1147	
PS 80-R.Ce 20	Safi Plaster 80% - Refractory Cement 20%	0.3 x 0.3 m ²	0.026	1382	
		φ 100 mm	0.044	1406	
		φ 29 mm	0.045	1328	

In general, the insertion of the synthetic and natural additives in the plaster matrix did not result in significant changes in density values with respect to the reference panel (Table 1); the mineral materials involved an increase in density up to about 32% (with 20% of refractory cement).

According to the heat flow meter method, the thermal performance of the samples was evaluated by means of the Small Hot Box experimental apparatus available at the Department of Engineering (University of Perugia) [35]. The tests (lasting at least 2 hours each) were carried out in steady state condition. The temperature inside the hot chamber was set at 45°C, to ensure a difference between the air temperature in the box (hot side) and in the laboratory room (cold side) higher than 20°C, according to [36]. Thermal resistance of each panel was calculated by measuring the heat flux through the sample (with a heat flow meter, ± 5% precision) and its hot and cold side surface temperatures (with thermo-resistances, ± 10% precision). The relative uncertainties type B of each test were calculated in compliance with JCGM 100:2008 [37].

Sound absorption coefficient (α) and Transmission Loss (TL) at normal incidence of each panel were measured in a conventional Kundt's tube (Brüel & Kjær, model 4187), by means of the two and four-microphone configurations (1/4 inch microphones Brüel & Kjær, model 4187), according to ISO 10534-2 [38]. Both large (100 – 1600 Hz, 100-mm diameter cylindrical sample) and small tube (400 –

6400 Hz, 29-mm diameter cylindrical sample) configurations were considered to investigate the entire frequency range (100 – 6400 Hz). The transfer function and the two-load methods were used to measure sound absorption and insulation properties, respectively.

A detailed description of the thermal and acoustic calculation procedures can be found in [39].

3. Results

3.1. Thermal properties

The thermal conductivity of the tested samples is reported in Table 2. Data measured at an average temperature in the 31.2 – 32.8 °C range were reported at the standard temperature of 10°C, according to ISO 10456 [40].

Thermal performance with synthetic additives improves when glass wool (PS 80-GW 20) and polyester fibers (PS 80-PF 20) are inserted in the mixture, with a reduction of thermal conductivity in the 40 – 44% range ($\lambda=0.144 - 0.155$ W/mK), with respect to Safi plaster (PS 100 [34]), whereas a higher value is obtained for the paraffin. The addition of natural additives involves an improvement in thermal properties ($\lambda=0.181 - 0.238$ W/mK vs. 0.257 W/mK of the reference, with a reduction of λ in the 7 – 30% range); the best results were obtained with 20% of Petiol of Palm in the mixture, close to synthetic additives ones. Conversely, the addition of mineral additives (clay, cement, refractory cement, and sand) leads to a worsening of performance, due to the higher density values; the increase in thermal conductivity varies in the 6% (with cement and sand) – 20% (refractory cement) range.

Results are consistent with the thermal properties of polystyrene gypsum-plasters measured by means of the same apparatus [17]. The samples were fabricated with the same Safi natural gypsum adding 10% and 30% by weight of EPS balls with 5-mm diameter ($\lambda= 0.216$ and 0.146 W/mK, respectively). Smaller particles involve thermal conductivity value in the 0.191 W/mK (10 wt.%) - 0.116 W/mK (30 wt.%), close to the performance obtained with Stipa Tenacissima and Petiol of Palm. Furthermore, also the study carried out by Charai et al. [25] on concrete composites incorporating Alfa fibers (ST) showed similar performance; the mixture with 10 wt.% of 3 cm fibers length was characterized by a thermal conductivity of 0.193 W/mK.

Table 2. Thermal properties of the samples.

Code	ΔT_{air} [°C]	$T_{s,\text{average}}$ [°C]	q_{average} [W/m ²]	$\lambda_{@31-33^\circ\text{C}}$ [W/mK]	$\lambda_{@10^\circ\text{C}}$ [W/mK]
PS 100	21.7	33.1	73.08	0.282	0.257
Synthetic additives					
PS 80-PF 20	26.8	31.2	76.57	0.169	0.155
PS 80-Pa 20	25.9	32.0	98.95	0.323	0.296
PS 80-GW 20	25.1	31.6	84.00	0.157	0.144
Natural additives					
PS 80-ST 20	24.7	31.4	73.49	0.205	0.188
PS 80-PPa 20	24.0	32.5	73.84	0.198	0.181
PS 80-SF 20	25.8	31.9	80.90	0.214	0.196
PS 80-Sw 20	24.4	32.6	79.38	0.225	0.206
PS 80-CB 20	23.9	32.8	83.36	0.261	0.238
PS 80-Ck 20	25.4	31.9	76.38	0.215	0.197
PS 80-ChSd 20	25.5	32.1	96.34	0.259	0.237
Mineral additives					
PS 80-Ce 20	24.4	31.6	85.30	0.300	0.275
PS 80-Cl 20	24.4	31.8	86.63	0.328	0.301
PS 80-Sa 20	23.9	31.8	82.00	0.301	0.276
PS 80-R.Ce 20	24.0	32.1	91.88	0.353	0.323

3.2. Acoustic properties

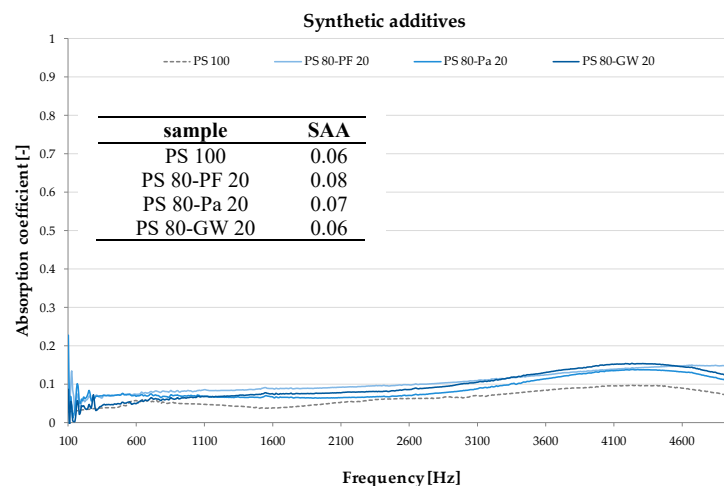
Sound absorption and insulation measurements were carried out taking into account several test specimens, with similar thicknesses and density; only the most significant measurements were considered.

The average normal incidence absorption coefficient trends of each sample are reported in Figure 2 in the 100-5000 Hz frequency range (combination of the large and small tube measurements), depending on the additive type in the mixture. The addition of additives improves the sound absorption properties compared to pure Safi plaster (PS 100, dotted line in Fig. 2 [34]) for all the three groups of additives. This behavior is hardly emphasized with synthetic additives, especially glass wool GW, paraffin Pa, or polyester fibers PF (Fig. 2 (a)). A maximum improvement of 0.02 of the Sound Absorption Average SAA [41] value (from 0.06 to 0.08) is observed for Polyester Fiber.

Better performance with respect to synthetic additives were obtained with natural ones, such as Petiol of Palm (PS 80-PPa 20), Sisal Fiber (PS 80-SF 20), and Stipa Tenacissima (PS 80-ST 20), characterized by lower densities (Fig. 2 (b)). In terms of SAA, values in the 0.10 – 0.15 range were obtained, double or more with respect to the value of the reference sample (0.06).

The insertion of mineral additives (Fig. 2 (c)) in the composites leads results comparative to natural additives, with a sound absorption peak value of 0.25-0.28 at about 900-1000 Hz, especially with 20% of sand (PS 80-Sa 20, SAA=0.15).

The Transmission Loss (TL) values measured with large tube configuration (100–1700 Hz) are shown in Figure 3; the measurements carried out on the small samples (small tube configuration, 400 – 6400 Hz) are not shown because not significant. Additives which involve an increase in the densities, also improve the sound insulation properties. The samples with Stipa Tenacissima ST and Petiol of Palm PPa are characterized by higher sound insulation performance, with TL values up to 59 dB (Fig. 3 (b)). For composite with 20% of straw Sw, TL-values are slightly lower than the pure Safi gypsum plaster PS 100 (TL values are in the 32-41 dB range for PS 100 with respect to 30-40 dB for PS 80-Sw 20), but it is also characterized by a lower density. Even with cement (PS 80-Ce 20) and Sisal fiber (PS 80-SF 20) the properties are lower, despite having higher densities, probably due to an imperfect mixing of the material during the samples preparation. Both synthetic and mineral additives, in general, involve a slightly improvement in the sound insulation properties with respect pure plaster (up to 4-5 dB).



(a)

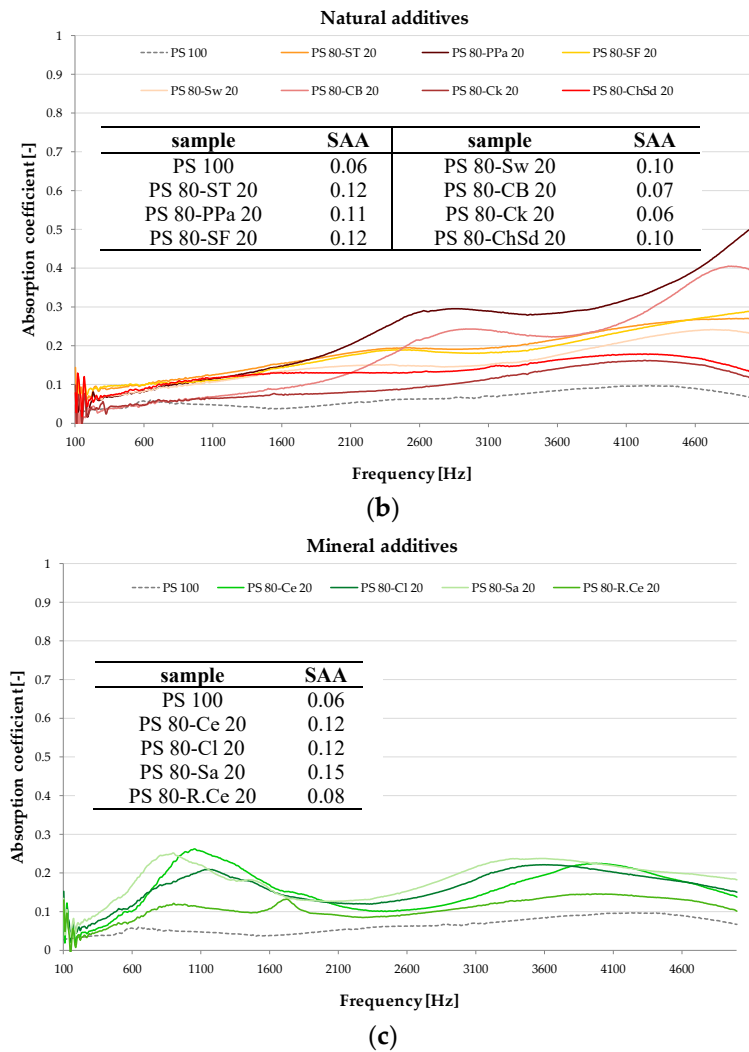
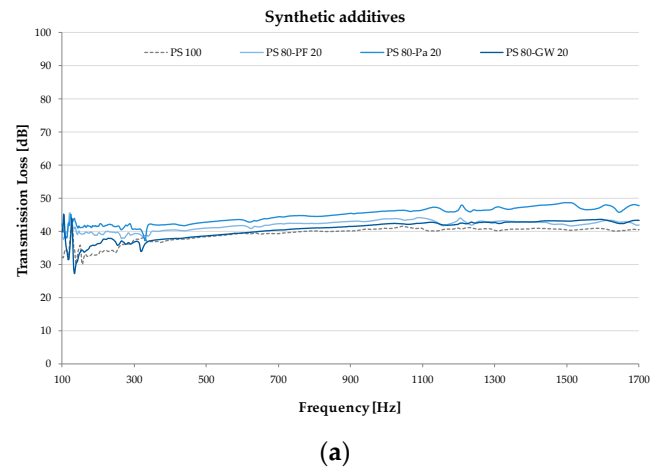


Figure 2. Absorption coefficient at normal incidence vs. frequency: gypsum plasters with synthetic (a), natural (b), and mineral (c) additives.

Compared to polystyrene [17], the sound absorption performance of natural additives is improved. 30 wt.% and 10% of the smallest EPS balls ($d = 3 \text{ mm}$, $\rho = 700 - 840 \text{ kg/m}^3$) have peak values of α equal to 0.19 and 0.17 at high frequencies (about 4200-4400 Hz), whereas the natural additive peaks are in the 0.25-0.30 and in the 0.30-0.40 range at mid and high frequency, respectively. TL-values were similar with synthetic and mineral additives ($TL = 30 - 45 \text{ dB}$, depending on the percentage and size of the EPS balls). Bio-additives have values even 12-13 dB higher.



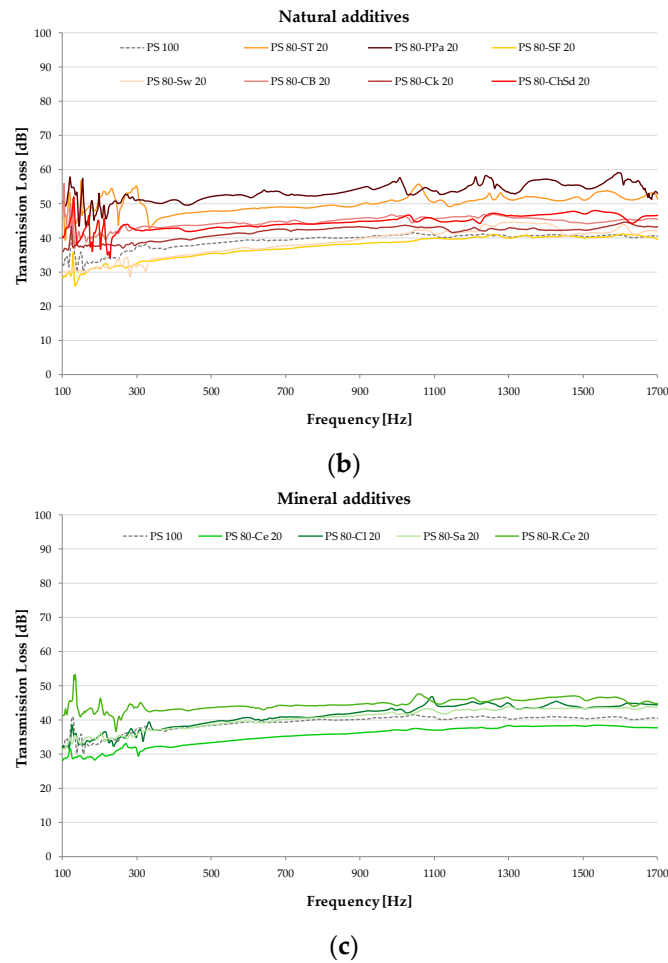


Figure 3. Transmission Loss at normal incidence vs. frequency: gypsum plasters with synthetic (a), natural (b), and mineral (c) additives.

4. Discussion

The decrease in plasters' density due to the additives improves thermal and sound absorption performance, resulting in a decrease in thermal conductivity and an increase in SAA values, according to linear trends. However, there is a decrease in Transmission Loss, as expected from the Mass Law.

Natural additives (dotted blue markers in Figure 4) improve the thermal properties of the plasters, especially when Petiol of Palm (PPa), Stipa Tenacissima (ST), Sisal fiber (SF), or cork (CK) are added in the matrix. Higher λ -values of the pure plaster (PS 100) are obtained with the mineral additives and also with paraffin (synthetic), characterized by a density of the same magnitude ($\rho = 960 \text{ kg/m}^3$). However, glass wool and polyester fiber perform better.

All additives result in sound absorption index SAA in the 0.06 - 0.12 range (Figure 5). Only sand has a SAA-value slightly higher (0.15). Among the naturals, ST, SF, PPa, Sw, and ChSd have the best behavior. However the acoustic insulation is of the same magnitude ($TL = 35 - 45 \text{ dB}$) of the one produced by mineral additives, which have higher densities (Figure 6 (a)). Higher TL values in bio-plasters were measured with PPa and ST, even though PPa has a slightly higher thickness (0.051 m, Figure 6 (b)). With the same thickness (0.049 m) ST has better acoustic insulation properties than Ck and SF: TL of SF-based plaster is lower despite its higher density.

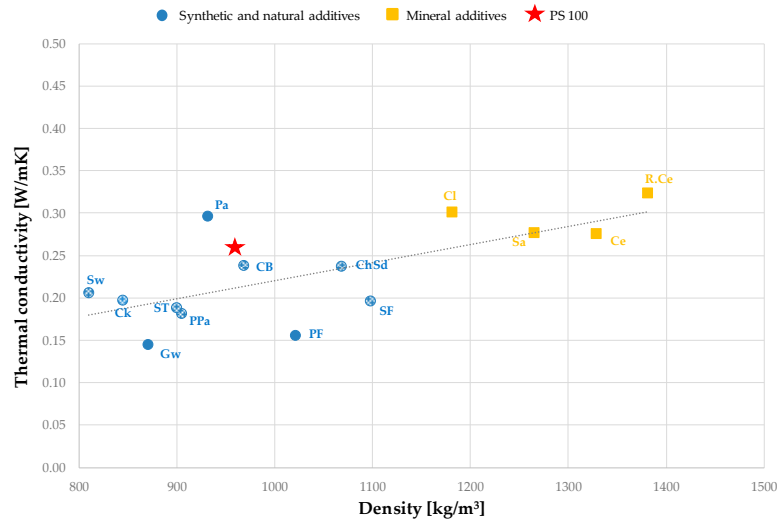


Figure 4. Thermal conductivity vs. density of the plasters with additives.

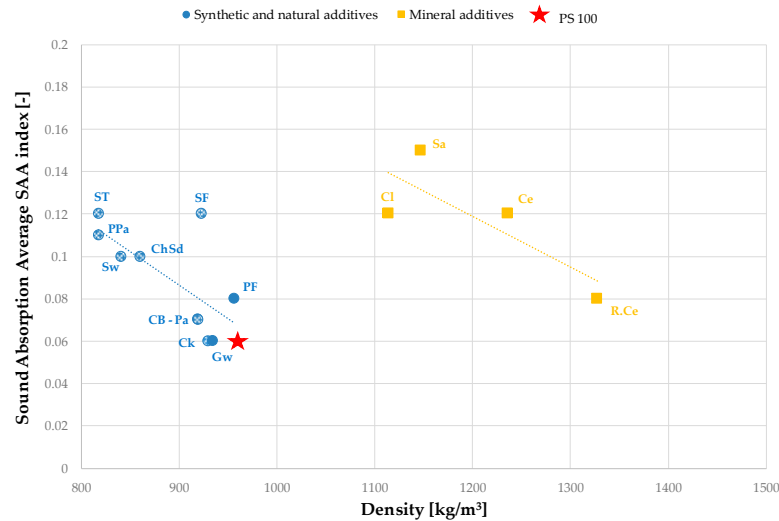
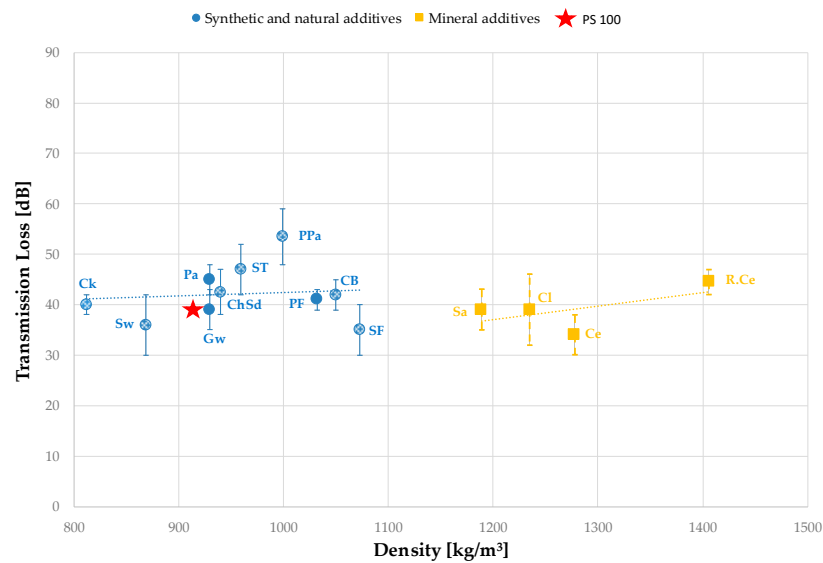


Figure 5. Sonud Absorption Average index vs. density of the plasters with additives.



(a)

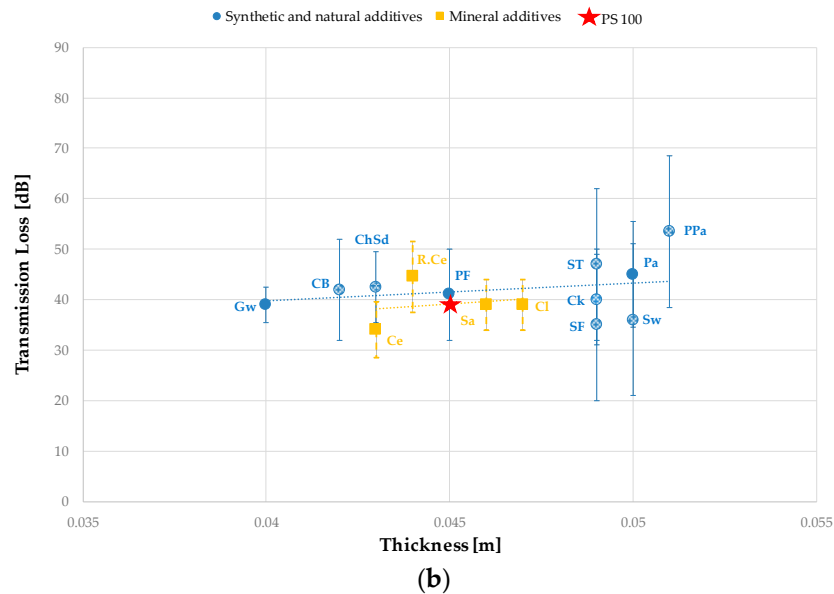


Figure 6. Transmission Loss of the plasters with additives: influence of the density (a) and the thickness (b).

5. Conclusion

Based on preliminary natural Safi gypsum rock characterization, several plasters were fabricated by adding natural, synthetic, and mineral additives. Seven natural additives (Stipa Tenacissima (ST), Petiol of Palm (PPa), Sisal Fiber (SF), Straw (Sw), Corn Bristles (CB), Cork (Ck), and Chips-Sawdust (ChSd)) were collected in the same area and were mixed with the gypsum plaster in 20% by weight. The composites thermo-acoustic performance were measured and compared with plasters with synthetic and mineral additives. The most promising composites were identified to promote the use of materials with low environmental impact in buildings refurbishment. Based on the experimental characterization, the following results were obtained:

- an increase in thermal performance with bio-additives: thermal conductivity in the 0.118 W/mK (with Petiol of Palm, PPa) - 0.238 W/mK (with Corn Bristles, CB) range, with a reduction up to 30% compared to pure gypsum plaster;
- an improvement in the sound absorption properties with all types of additives. Sound Absorption Average index SAA in the 0.06 (cork, Ck) – 0.12 (Stipa Tenacissima, ST, and Sisal fiber, SF) range were measured;
- higher sound insulation properties with ST and PPa (TL = 48 – 59 dB). However straw and Sisal fiber did not seem to affect the performance;
- the density of the samples affects the performance. The reduction in density leads to better thermal performance, with comparable sound absorption values. TL is slightly reduced but significant values were measured.

The experimental campaigns highlighted a good thermal and acoustic behavior of bio-based gypsum plasters, especially when Petiol of Palm and Stipa Tenacissima were added to the composites. However, the mechanical and hygrothermal performance of the most promising solutions will have to be investigated, in order to optimize and identify the best solutions from a thermal, acoustic, mechanical, and vapour permeability point of view.

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Conflicts of Interest: The authors declare no conflict of interest.

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