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Thermal, Acoustic, and Hygrothermal Properties of Recycled Bovine Leather Cutting Waste-Based Panels with Different Compositions

Francesca Merli [1], Costanza Vittoria Fiorini [2], Marco Barbanera [3], Giorgia Pietroni [3], Francesco Spaccini [4] and Cinzia Buratti [1,*]

[1] Department of Engineering, University of Perugia, Via G. Duranti 63, 06125 Perugia, Italy
[2] DIAEE Department of Astronautical, Electrical and Energy Engineering, "Sapienza" University of Rome, Via Eudossiana 18, 00184 Rome, Italy
[3] Department of Economics, Engineering, Society and Business Organization, University of Tuscia, 01100 Viterbo, Italy
[4] CIRIAF, Centro Interuniversitario di Ricerca sull'Inquinamento e sull'Ambiente "M. Felli", University of Perugia, Via G. Duranti 67, 06125 Perugia, Italy
* Correspondence: cinzia.buratti@unipg.it; Tel.: +39-075-5853993

Abstract: This study aims to investigate the thermo-acoustic and hygrothermal properties of innovative panels made of leather waste produced by a bag factory from a circular economy perspective. Their performance was compared with other residual-based insulation panels. The leather scraps were chipped and three boards were fabricated by means of a mold with different compositions, adding adhesive glue. In order to improve the sustainability aspects, a sample was assembled by using a water-based polyurethane glue, in addition to the two panels with vinyl glue. Panels were tested for thermal, acoustic, and hygrothermal performance. Thermal conductivity values in the range of 0.064–0.078 W/mK at 10 °C were measured depending on the composition and the adhesive. A slight thermal performance deterioration occurs when using the natural water-based glue. The samples were characterized by good performance both in terms of sound absorption coefficient (Noise Reduction Coefficient NRC = 0.21–0.28) and Transmission Loss, up to 59 dB values. Water vapor resistance factor values in the 35–48 range were obtained, close to the values of standard materials, such as expanded polystyrene and polyurethane.

Keywords: bovine leather cutting waste; thermal properties; acoustic performance; hygrothermal properties

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1. Introduction

In modern society, energy efficiency and environmental issues are becoming central topics. The construction and building sector can be considered as a major contributor of a large part of carbon dioxide emissions and pollution [1]. Today's society is developing quickly and the demand for buildings and their comfort levels are increasing. Heating, air conditioning, and ventilation consumption accounts for about 50% of a building's energy demand [2] and the application of thermal insulation is the main technical measure to reduce energy use and cost [3]. In addition to the thermal aspect, it is important to consider the acoustic one closely related to people's well-being [4,5]. Noise from road traffic, construction sites, or social activities is an unavoidable problem caused by rapid urban development and it is a major source of environmental pollution affecting people's health, especially in densely populated areas [6]. To ensure maximum comfort inside a building, noise reduction techniques and materials should also be investigated [7]. Moreover, the humidity tightness of the building envelope is another very important aspect; it is necessary to balance the breathability of the walls, in order to ensure a longer life of the materials by avoiding condensation problems.

Therefore, several synthetic, natural, and innovative materials have been deeply investigated in the literature and launched on the market. Examples are plastic, mineral and sheep wool, wood, cellulose, vacuum insulation boards, and aerogel [8–12]. Presently, more and more researchers are focusing on thermal and sound insulation materials based on the concepts of environmental friendliness, recycling, and circular economy. Industries generate a large amount of solid waste. Increased restrictions, recent increases in costs, and decreases in the number of disposal sites are encouraging the development of alternative treatments. The reuse of different types of waste can significantly contribute to sustainability.

Textile scraps are very interesting materials for building envelope applications [13,14]. Polyester is the most widely used fiber in apparel and textiles. Trajković et al. [15] used polyester cutting waste during apparel manufacturing to produce thermal and acoustic insulation materials. Good insulation properties were found with thermal conductivity in the 0.0520–0.0603 W/mK range and the noise reduction coefficient (NRC) in the 0.55–0.75 range. Wool has been one of the most used animal fibers in the world since ancient times, due to its excellent thermal insulation properties. In particular, sheep wool, which has a thermal conductivity of 0.034 W/mK measured in panels with a thickness of 40 mm [16], also has excellent sound-absorbing properties: the absorption coefficient is about 0.9 at around 800–2000 Hz [17]. Cotton is one of the most cultivated non-food agricultural products, mainly used in apparel production, and in the last few years, some companies have started commercial production of insulation materials from cotton waste. Bonded Logic incorporated commercially produced Echo Eliminator™ Acoustic Panels from recycled cotton fibers, with NRC in the 0.8–0.9 range [18]. Zach et al. produced thermal and acoustic insulation materials by combining recycled cotton, polyester, and flax fibers, obtaining an average thermal conductivity of the tested samples equal to 0.043 W/mK [19].

In global tanned leather processing, 25% is solid waste [20]. Furthermore, 75% of used leather articles are thrown away [21]. These obsolete leather solids contain more than 85% collagen fiber and about 3% of Cr_2O_3, and the rest is made up of polymer resins and organic fillers, which not only threaten the environment, but also cause some waste of resources. In order to reduce environmental impact, it is necessary to find an effective method that can be recycled without pollution [22,23]. Italy is the top country in leather production; with over 135 million m^2 of hides, it represents around 65% of European and 13% of world production [24]. The small and medium enterprises specialized in the processing of bovine hides are located above all in the provinces of Vicenza and Pisa. Together with those specialized in the tanning of sheep and goat skins, they cover exports representing over 2/3 of the total turnover.

In this context, the authors in the last year started a study on the thermal, acoustic, fire, and hygrothermal properties of boards fabricated with recycled leather waste [25]. Thermal conductivity in the 0.100–0.110 W/mK range at 33–35 °C was obtained. Concerning the acoustic performance, NRC = 0.46 for a thickness of 18 mm and NRC = 0.20 for a thickness of 28 mm were measured, with transmission loss values in the 25–33 dB and 25–42 dB range, respectively. Moreover, leather board showed good moisture transfer (water vapor resistance factor in the 27–30 range), rejecting the attacks of fungi and bacteria. These aspects, together with thermal stability and wettability performance, confirmed the potential of the application of this material in buildings, with a low environmental impact.

In the present paper, an extended experimental campaign was carried out on three panels composed by different leather scraps deriving from the manufacturing of bags. Two panels were assembled with different types of leather waste glued with vinyl binder. Furthermore, a water-based polyurethane glue was used to fabricate a further sample with a higher sustainability perspective. The thermal, acoustic, and hygrothermal properties of recycled leather waste panels were investigated and they were compared with those of other insulation materials.

2. Materials and Methods

An Italian company which fabricates leather bags in the province of Viterbo provided finished bovine leather cuttings. In general, these wastes are disposed to landfill involving severe environmental damage. After being chipped by means of a hammer chipping machine with a 5 mm sieve, leather cuttings were manually mixed with glue at room temperature. Three different mixtures were prepared and the following samples were fabricated:

- P1, leather cutting waste panel with vinyl glue, composed of leather scraps (1.3 kg), vinyl glue (0.65 kg, 50% by weight of the leather), and distilled water (0.27 kg, 20% by weight of the leather). The glue consists of polyvinyl alcohol emulsifying agent in liquid form [25];
- P2, leather cutting waste + internal support leather materials (infustiture) panel with vinyl glue, composed of leather scraps (0.455 kg, 35% of the total waste), infustiture (0.845 kg, 65% of the total waste), vinyl glue (0.65 kg, 50% by weight of the leather + infustiture matrix), and distilled water (0.24 kg, 18% by weight of the leather + infustiture matrix). The infustiture was composed of 50% of polyurethane and 50% of polyamide, as declared by the company. The vinyl is the same type as in the panel P1;
- P3, leather cutting waste panel with ECHO PU 249/F glue [26], composed of leather scraps (1.3 kg), ECHO PU 249/F glue (0.65 kg, 50% by weight of the leather), and distilled water (0.24 kg, 18% by weight of the leather). The glue used is a water-based polyurethane adhesive.

As in the previous study [25], a stainless steel mold and a cold hydraulic press machine (TOYO: Model TL30, capacity 30 Ton) were used in order to fabricate square panels (0.36×0.36 m^2). After the pressing operation for 48 h at room temperature, each leather board was demolded and it was kept in oven at 70 °C for 24 h, in order to remove excess water. Samples (0.3×0.3 m^2) were obtained cutting the fabricated boards for the thermal characterization (P1, P2, and P3, Figure 1a). Unlike the other panels, the one with water-based glue appears rather fragile, showing some small cracks on its irregular surface. After thermal measurements, cylindrical samples (two samples of 100 mm and 29 mm diameter each, named A and B) were obtained by means of a cylindrical hole cutter for the acoustic characterization; the thickness of the specimens were not very uniform due to the manual assembly. The same large specimens (P1-A-LT, P1-B-LT, P2-A-LT, P2-B-LT, P3-A-LT, P3-B-LT) were then used for the hygrothermal tests (Figure 1b). All the panels were pre-conditioned following the typical environmental conditions (temperature equal to 25 °C and relative humidity equal to 57%). The composition and the main features of the examined samples are reported in Table 1. The thickness of each sample is not very uniform; it was measured in at least 5 points in each sample and averaged. Due to the cut, also the densities of the acoustic samples are not the same. The specimens were weighted with an electronic precision balance and the density values in the 592–648 kg/m^3, 693–775 kg/m^3, and 614–680 kg/m^3 ranges were obtained for the P1, P2, and P3 samples, respectively.

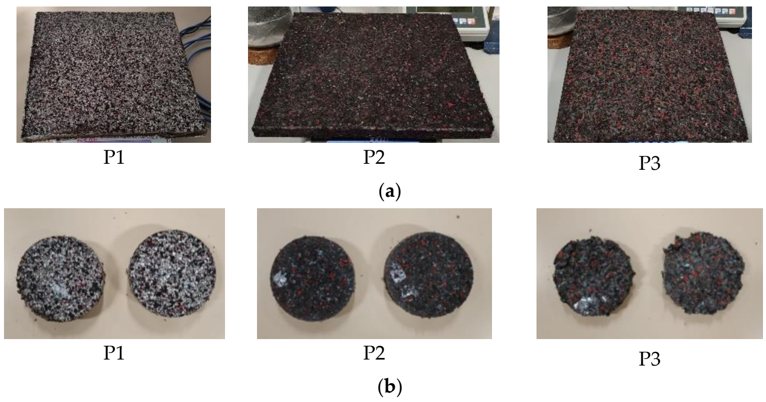


Figure 1. Leather cutting-based samples: (**a**) Thermal measurements; (**b**) Acoustic and Hygrothermal measurements.

Table 1. Main characteristics of the investigated samples.

Sample	**Composition**		
P1	1.3 kg leather scraps, 0.65 kg vinyl glue, 0.27 kg distilled water		
P2	0.455 kg leather scraps, 0.845 kg infustiture, 0.65 kg vinyl glue, 0.24 kg distilled water		
P3	1.3 kg leather scraps, 0.65 kg water-based glue, 0.24 kg distilled water		
Thermal characterization			
Sample	Average thickness [m]		
P1	0.018		
P2	0.019		
P3	0.017		
Acoustic and Hygrothermal characterizations			
Sample	Diameter [m]	Average thickness [m]	Density [kg/m^3]
P1-A-LT *	0.100	0.021	623
P1-A-ST *	0.029	0.021	641
P1-B-LT *	0.100	0.020	648
P1-B_ST *	0.029	0.021	592
P2-A-LT *	0.100	0.017	775
P2-A-ST *	0.029	0.016	693
P2-B-LT *	0.100	0.018	775
P2-B_ST *	0.029	0.017	700
P3-A-LT *	0.100	0.014	660
P3-A-ST *	0.029	0.019	671
P3-B-LT *	0.100	0.016	614
P3-B_ST *	0.029	0.020	680

* LT, large tube (100 mm diameter); ST, small tube (29 mm diameter).

Thermal, acoustic, and hydrothermal properties of the panels were investigated in order to determine a higher performance board for a building envelope, from a sustainable and circular economy perspective (Figure 2).

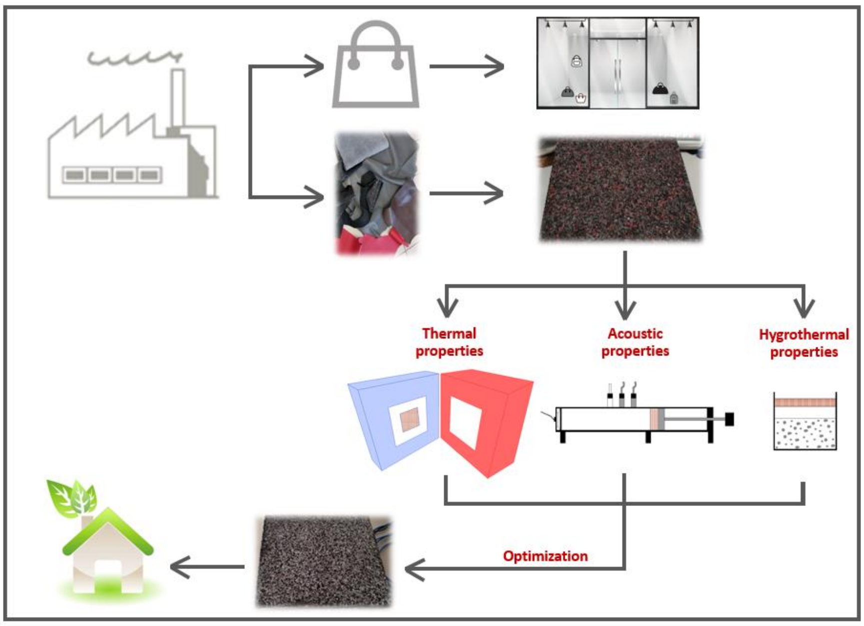


Figure 2. Scheme of the research.

Thermal properties were measured by means of a Small Hot Box apparatus, available at the Laboratory of Environmental Control, Department of Engineering—University of

Perugia. A detailed description of the device is available in [27]. Each test was performed with the heat flow meter method for a duration at least 2 h, maintaining stationary conditions both for temperatures and for thermal flux. Four thermo-resistance probes installed on each side of the sample and a thermal flux meter model HP01—Hukuseflux on the center of the panel's hot side were used in order to measure the mean hot and cold surface temperature difference ΔT_s and the mean heat flux Φ, and to determine the thermal resistance R. The thermal conductivity λ was calculated from the measured thermal resistance and the thickness of the specimen. For each test, the relative uncertainties (type B) were calculated, in compliance with JCGM 100:2008 [28]. The experimental data were measured at an average temperature of the sample in the 31–35 °C range. In order to compare the board's performance with the ones of standard and other residual-waste heat insulating panels, the measured data were reported at a standard temperature of 10 °C following the ISO 10456 [29]. A temperature conversion coefficient equal to 0.0033 1/K was considered, as recommended by the Norm for similar materials.

Sound absorption coefficient (α) and Transmission Loss (TL) at normal incidence were measured by means of an impedance tube (Brüel and Kjær, model 4206; 1/4 inch microphones Brüel and Kjær, model 4187) [25]. Transfer function method was used in order to measure α-values, using a two-microphone configuration, according to ISO 10534-2 Standard [30]. Cylindrical samples with large (100 mm) and small (29 mm) diameters for frequencies in the 100–1600 Hz and in the 400–6400 Hz range, respectively, were considered for the measurements. The obtained α-trends were combined with a software to cover the entire range of frequencies (100–6400 Hz). In order to easily compare the acoustic performance of the samples, the Noise Reduction Coefficient (NRC) was calculated as the mathematical average, rounded to the nearest multiple of 0.05, of the absorption coefficient values in the one octave band at the frequencies 250, 500, 1000, and 2000 Hz. The measurements of the sound insulation performance (TL) were carried out with the two-loads method, by acquiring the sound pressure in four fixed microphone positions [31,32].

The water vapor resistance factor μ defines the resistance of the material with respect to an equally thick layer of stationary air at the same temperature (μ = 1 for air). The tests were carried out on each 100 mm leather specimen by a dry cup test containing 15 mm of silica gel with color indicator as desiccant, according to UNI EN ISO 12572 Standard [33]. The tests were carried out in the temperature- and relative humidity-controlled conditions, in compliance with [33], inside a climatic chamber (Mazzali C330G5, temperature —equal to 23 °C and relative humidity equal to 50%) available at the University of Perugia. A vapor flow through the samples is caused by a different partial vapor pressure among the test cup and the climatic chamber. The specimens were weighed once every 24 h by means of a precision balance and the water vapor transmission rate in the steady-state conditions was evaluated, according to the procedure reported in [25].

3. Results and Discussion

3.1. Thermal Properties

For each leather cutting waste panel, four tests were carried out, setting the temperature inside the hot chamber at 45 °C and 50 °C; data were processed in a period of about 2 h, with both temperatures and heat flow in steady state. In all measurements, the difference between the air temperature into the hot (inside the box) and the cold (inside the laboratory room) side was more than 20 °C.

The results of the thermal tests carried out in the various environmental conditions are shown in Table 2. The panel characterized by the best thermal properties is P2; the average thermal conductivity at 10 °C calculated according to [29] is equal to 0.064 W/mK (R = 0.293–0.299 m^2K/W, as the test conditions vary). The P1 panel with leather waste and vinyl glue has intermediate performance (λ = 0.072 W/mK, R = 0.230–0.250 m^2K/W). The addition of water-based glue to leather cutting waste (P3) leads to a deterioration in thermal properties; the thermal conductivity increases by about 8% and 22% with respect to the use of vinyl glue (P1) and the addition of infustire (P2), respectively. However, it should be noted that sample P3 is characterized by a more irregular surface and with slight lesions. For each measure, the relative uncertainty type B $\dot{u}$(R) was calculated according to [28]; values in the 2.8–4.5% range were found, in the same order of magnitude of the instrument measurement error (equal to 3–6%, estimated with several preliminary calibration tests [27]).

Table 2. Comparison of thermal conductivity of the investigated samples and other standard and innovative materials.

Sample	Hot Side Test Condition [°C]	Φ [W/m²]	ΔT_s [°C]	λ @31–35 °C [W/mK]	R@10 °C [W/mK]	$\dot{u}$ (R) [%]	λ_{mean} @10 °C [W/mK]
P1 (s = 0.018 m)	45	57.41	13.55	0.076	0.254	3.6	0.072
	45	56.41	13.13	0.077	0.250	4.5	
	50	69.88	16.01	0.079	0.249	3.7	
	50	70.70	16.29	0.078	0.230	4.2	
P2 (s = 0.019 m)	45	53.90	15.00	0.068	0.298	2.0	0.064
	45	55.08	15.36	0.068	0.299	2.8	
	50	70.01	19.05	0.070	0.294	4.0	
	50	67.93	18.42	0.070	0.293	3.9	
P3 (s = 0.017 m)	45	55.87	11.40	0.083	0.220	3.5	0.078
	45	55.89	11.30	0.084	0.218	4.0	
	50	66.86	13.28	0.086	0.216	2.9	
	50	65.94	13.02	0.086	0.215	2.7	

The reduction in chipped leather pieces involves an improvement in thermal performance; thermal conductivity value decreases by 22 (for P3 panel)–36% (for P2 panel) compared to the leather cutting waste LCW panel fabricated by means of a hummer chipping machine with a 30 mm sieve [25] (Table 3). However, the panels perform worse with respect to standard insulating materials, i.e., expanded and extruded polystyrene, which shows a thermal conductivity in the 0.030–0.040 W/mK range. However, the great added value of the panels is given by their sustainability and low environmental impact. In this perspective, the obtained thermal performance is close to that of other natural waste materials, such as wood in both fiber and chipped form, rice husk, and coffee chaff-based panels [27,34–37]. Other mineral and natural fiber-based panels, such as kenaf, cork, paper, and sheep wool have good thermal properties (λ = 0.034–0.052 W/mK). Conversely, thermal performance is better when compared to a granulated rubber waste panel (λ = 0.127 W/mK) [37], despite the texture being similar.

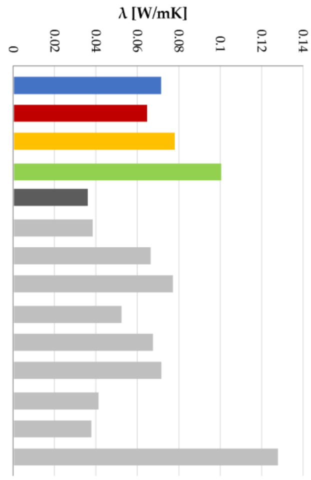
3.2. Acoustic Properties

Several measurements for each sample were carried out and average absorption coefficient (α) and Transmission Loss (TL) values were deemed for the sample with most similar thickness and density. α- and TL-trends (combination of the large and the small tube measurements for the absorption coefficient, 100–6400 Hz) are shown in Figures 3 and 4, respectively.

The panel with higher density (P2) has the best performance both in terms of sound absorption and insulation properties; NRC is equal to 0.28 and the TL is in the 38–59 dB range. At low frequencies, the leather cutting waste panel with vinyl glue (P1) is characterized by a α-peak value of about 0.22 at 500 Hz; when the mean thickness of the sample decreases slightly (i.e., P2 and P3), the peak of the absorption curve is moved to higher frequencies (about 600 Hz), as expected. Water-based polyurethane glue used in the P3 panel involves an increase in the absorption coefficient (up to 0.28) with respect to the sample with similar density (P1).

Table 3. Thermal properties of the samples: comparison with other materials.

Panel	λ @10 °C [W/mK]
P1	0.072
P2	0.064
P3	0.078
LCW [25]	0.100
expanded and extruded polystyrene [25]	0.035
kenaf [34,38]	0.038
wood fiber [27,34,35]	0.065
chipped wood [36]	0.071–0.084
cork scraps * [37]	0.052
rice husk * [37]	0.066
coffee chaff * [37]	0.071
wastepaper * [37]	0.034–0.047
sheep wool * [16,35]	0.036–0.038
granulated rubber * [37]	0.127



* data calculated at a mean surface temperature of 10 °C, according to [29].

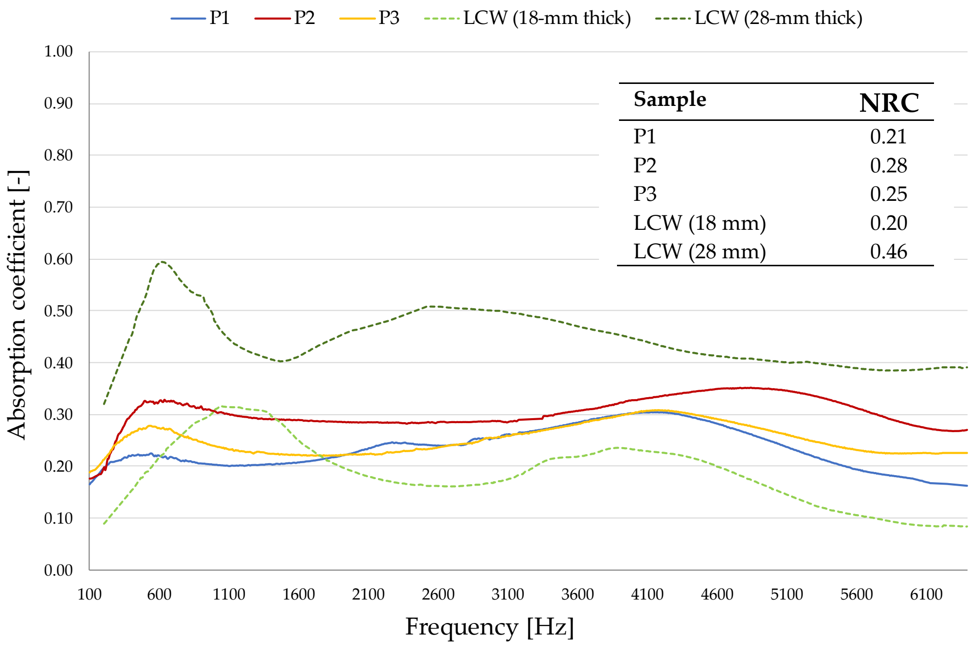


Figure 3. Sound absorption coefficient at normal incidence vs. frequency: influence of chipped leather pieces size [25].

The two trends are overlapped at frequencies greater than 2000 Hz, whereas P3 has α-values higher than P2 over 4500 Hz. The sound insulation performance of P3 panel is of the same magnitude of P1. In fact, despite TL = 30–41 dB for P1 and TL = 30–38 dB for P3, the mean thick value of the panel with natural glue (P3, equal about 17 mm) is lower, involving a decrease in Transmission Loss, according to the literature [31]. Considering the results obtained in the previous study [25], an increase of NRC in the 5–30% range is obtained with respect to the 18 mm thick panel. In the 200–1600 Hz range, transmission loss levels are significant: for the P2 board, they also exceed the values measured for the 28 mm LCW panel with higher-size leather pieces (TL = 25–42 dB) [25]. The obtained acoustic absorption properties are consistent with those of mineral and natural fiber-based panels (NRC = 0.20–0.31) with similar thicknesses (16–20 mm), and with NRC value of 17 mm granulated rubber board characterized also by similar texture and density. Conversely, less thick panels (10 mm) of coffee chaff and glued wastepaper perform better [25]. Glued wood fiber has a similar NRC value (0.25), but for much higher panel thickness (30 mm) [27,34,35].

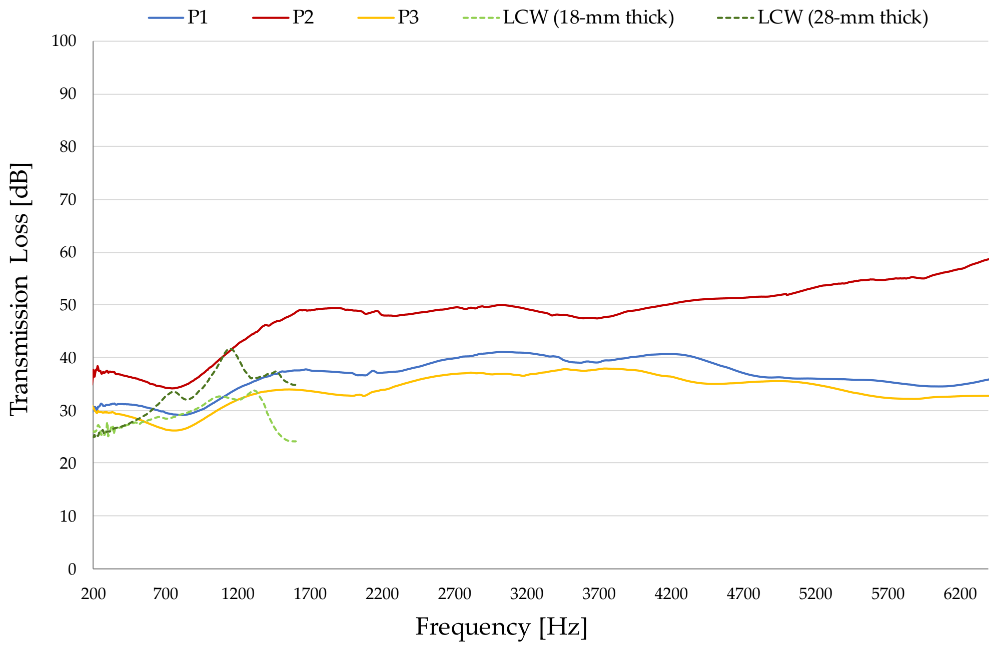


Figure 4. Transmission Loss at normal incidence vs. frequency.

3.3. Permeability Properties

The water vapor resistance factor μ of each sample was calculated when the permeability test stability was achieved (8–21 days from the beginning of the experimental campaign), according to the standard prescriptions [33]. For calculation, a water vapor permeability of air equal to 2.05×10^{-10} kg/(msPa), with a measured barometric pressure at 23 °C inside the apparatus of 985 hPa and a water vapor pressure difference across the sample equal to 1404 Pa were assumed. G-values in the 2.88–3.16 $\times 10^{-9}$ range were calculated and mean μ-values of 35, 44, and 48 were obtained for the P1, P2, and P3 panels, respectively (Table 4). Water-based glue involves an increase of about 37% in the permeability performance. Higher water vapor resistance values were obtained with increasing density for all mixtures (Figure 5). For P1 and P3 panels, μ increases by about 15% as the density rises by 4–7%, respectively. Furthermore, resistance value increases of about 13% for boards with the addition of the infustiture (P2-A and P2-B) were characterized by similar density values (775.2 and 774.6 kg/m^3 respectively). Water vapor resistance of the material is improved when the leather cutting wastes are smaller; lower μ-values (in the 27–30 range for 18 mm boards with density equal to 570 and 650 kg/m^3, respectively) were obtained in the previous study [25]. Permeability performance is close to that of conventional insulating materials, such as the expanded polystyrene (μ = 20–80) and polyurethane (μ = 30–100). Conversely, the panels had higher moisture transfer properties with respect to many traditional fibers (i.e., wood (μ = 3–10), cork (μ = 5–10), hemp, corn, and coir (μ = 1–3), expanded vermiculite and perlite (μ = 5–8), glass and stone wool (μ = 1–5), sheep wool (μ = 3), and glued waste and wool paper (μ = 2–4)) [25,39].

Table 4. Water vapor resistance factor of the panels.

Sample	G [kg/s]	μ	$\mu_{average}$
P1-A-LT	3.30×10^{-9}	33	35 (P1)
P1-B-LT	3.02×10^{-9}	37	
P2-A-LT	2.88×10^{-9}	46	44 (P2)
P2-B-LT	3.08×10^{-9}	41	
P3-A-LT	3.16×10^{-9}	51	48 (P3)
P3-B-LT	3.16×10^{-9}	45	

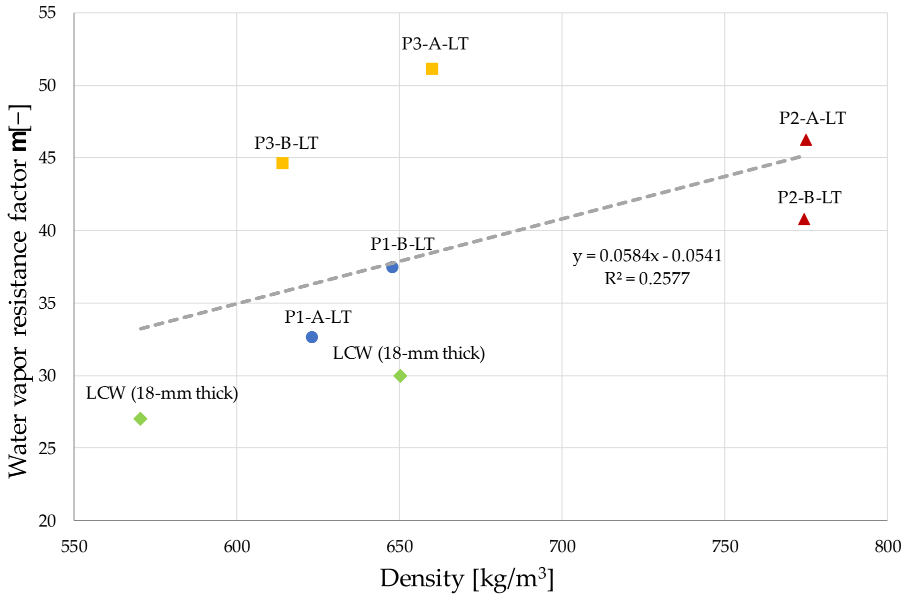


Figure 5. Water vapor resistance factor vs. density: influence of chipped leather pieces size [25].

4. Conclusions

The large amount of waste generated by the leather goods industry requires solutions in order to ensure an end-of-life for the scraps from an environmental sustainability and circular economy perspective. In this research, leather scraps deriving from the manufacturing of bags were used as secondary raw materials to fabricate panels for insulating building applications. Two panels were fabricated by means of 3 mm leather pieces glued with vinyl; internal support leather materials were added in one of the samples (P2). A water-based adhesive was used in a third board (P3). Thermal, acoustic, and hygrothermal performance was investigated in order to evaluate the suitability of the proposed solutions for building applications. It was obtained that:

- The leather cutting waste with internal support leather materials panel (P2) has a higher density and better thermo-acoustic performance; thermal conductivity is 0.064 W/mK at 10 °C, the noise reduction coefficient is 0.28, and the Transmission Loss is in the 38–59 dB range;
- The use of natural glue leads to a slight deterioration in thermal performance (λ = 0.078 W/mK). However, sound insulation properties are comparable with those of the leather cutting waste panel with vinyl glue (P1), whereas the absorption coefficient values are higher (NRC equal to 0.25 and 0.21 for P3 and P1, respectively);
- The thermo-acoustic behavior of the samples is close to that of other natural fiber-based panels with similar thickness, such as wood (λ = 0.065–0.084 W/mK), rice husk (λ = 0.066 W/mK), and coffee chaff (λ = 0.071 W/mK) boards;
- The thermal properties of the leather panels are slightly worse than those of standard insulating materials. However, their benefit is the low environmental impact;
- Excellent permeability properties were obtained for the studied samples, especially when natural glue was used; water vapor resistance factor values equal to 35, 44, and 48 were measured for P1, P2, and P3 panels, respectively;
- The reduction in the leather pieces' dimension resulted in an improvement in thermal (thermal conductivity reduction up to 36%), acoustic (NRC increase up to 30%, and TL outperforms the thickest panel with the largest pieces), and permeability properties.

The proposed panels are a valid alternative to traditional materials used in the building sector, due to their good properties combined with low environmental impact. Unlike other materials of natural origin, leather waste takes a long time to decompose. For this reason, its reuse has a greater impact in terms of environmental sustainability. This aspect will be investigated in future research through Life Cycle Assessment analysis.

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

Nomenclature

R [m^2K/W]	thermal resistance
ΔT_s [K]	mean surface temperatures
Φ [W/m^2]	heat flux
λ [W/mK]	thermal conductivity
$\dot{u}$ [%]	relative uncertainty type B
α [-]	absorption coefficient
TL [dB]	Transmission Loss
NRC [-]	Noise Reduction Coefficient
μ [-]	water vapor resistance factor

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