

ACOUSTIC PROPERTIES OF POLYPROPYLENE COMPOSITES REINFORCED WITH STONE GROUNDWOOD

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Currently, acoustic isolation is one of the problems raised with building construction in Spain. The publication of the Basic Document for the protection against noise of the Technical Building Code has increased the demand of comfort for citizens. This has created the need to seek new composite materials that meet the new required acoustical building codes. In this paper we report the results of the newly developed composites that are able to improve the acoustic isolation of airborne noise. These composites were prepared from polypropylene (PP) reinforced with mechanical pulp fibers from softwood (*Pinus radiata*). Mechanical and acoustical properties of the composites from mechanical pulp (MP) and polypropylene (PP) have been investigated and compared to fiberglass (FG) composites. MP composites had lower tensile properties compared with FG composites, although these properties can be improved by incorporation of a coupling agent. The results of acoustical properties of MP composites were reported and compared with the conventional composites based on fiberglass and gypsum plasterboards. Finally, we suggest the application of MP composites as a light-weight building material to reduce acoustic transmissions.

Keywords: Mechanical pulp; Tensile properties; Acoustical properties; Polypropylene; Composites

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INTRODUCTION

Over the past two decades, great attention has been dedicated to the exploitation of natural fibers as reinforcement for plastics, thereby replacing synthetic fibers (Habibi *et al.* 2008; Rahman *et al.* 2009). Natural fibers are used as suitable reinforcing material to satisfy environmental aspects, and they are now rapidly emerging as a potential alternative to synthetic fibers in engineering composites (Lopez *et al.* 2012). Natural fibers can be described as renewable, nonabrasive, cheaper, abundant, and showing less health and safety concern during handling and processing compared to the fibers most often being used in composites at present. In our research group, several lignocellulosic fibers such as hemp, jute, sisal, flax, alfa, abaca, pine, and stone ground wood fibers have been explored as reinforcement in composite materials (Vallejos *et al.* 2006; Méndez *et al.* 2007; Mutjé *et al.* 2007; Vilaseca *et al.* 2010; Lopez *et al.* 2012).

The mechanical wood pulp (MP) used in this research work was a stone groundwood pulp, which is a fibrous material, commonly prepared from softwoods, in a

process that can reach 98.5wt% yield (Lopez *et al.* 2012). The most common applications of MP are in the production of printing paper, newsprint, boards, and packaging papers. Mechanical wood pulp is frequently used in paper formulations together with recycled fibers (Sundholm 1998). Thanks to these applications, the existence of mechanical pulp in the global market is guaranteed, and it has a very economical price between 0.3 and 0.4 € /kg (Lopez *et al.* 2011). Moreover, due to its fibrous morphology, mechanical fiber has found applications as a reinforcing element of polypropylene and polyethylene (Mendez *et al.* 2007; Lopez *et al.* 2011 and 2012). In this respect, it was also reported that natural fibers can be used as a substitute for wood-based raw materials and explored as filler for composite systems suitable for acoustic applications (Huda and Yang 2009). In that work, feather and jute fibers were used as a reinforcement element for light-weight composites with good acoustic properties. Composites studied in this work, have a density below the materials used as acoustic lightweight isolation solutions ($< 20 \text{ kg/m}^2$). However, to the best of our knowledge, there is no research work published on the use of mechanical wood fibers (MP) as reinforcement for the light-weight building material designed for acoustical applications.

At the present time, acoustic isolation is one of the problems raised with building construction in Spain. The sources of noise found both inside and outside the buildings, which generate higher levels of noise, are more and more numerous (domestic electrical appliances, systems for reproducing sound, traffic, *etc.*). And with the recent publication of the Basic Document for the protection against noise of the Spanish Technical Building Code (Real decreto 1371/2007), the exigencies for the comfort of citizens have increased. This has created the need to seek new composite materials that meet the new required acoustical building codes.

These compound compositions show characteristics of sound-proof layers. There are mathematical models to describe the acoustic behavior of these sound-proof layers based on elastic properties or properties related to bending (Alba *et al.* 2003, 2004). The acoustical behavior of the absorbent materials cannot be described from the same properties (Delany and Bazley 1970, King *et al.* 2012). In this case, the properties to be considered are viscosity, tortuosity, and porosity based on the distribution of fibers that compose the absorbent material.

This research work reports the results of the mechanical and acoustical properties of mechanical pulp (MP) and fiberglass (FG) composites. The acoustical behavior of MP composites was evaluated for their application as acoustic isolation elements for single-layer and double-layers. The obtained results were compared to the acoustic solutions commonly used as light-weight building material such as gypsum plasterboard.

Theoretical Basics

There exist many models for the prediction of acoustic isolation for both airborne as well as impact noise (Brekke 1981; Davy 2009, 2010; Ookura and Saito 1978; Vinokur 1990). Some of these predictive models only allow the possibility for modeling the acoustic behavior of a single layer. Others permit study of isolation of multi-layer partitions with either air chamber or material with absorbent properties in its interior. And in some cases allow modeling solutions for perforating plates that improve many frequency problems.

For the present work, we have developed a computer application (AISLA) that studies the three situations mentioned above. This computer application takes as a starting point the prediction model reported by Ookura and Saito (1978). This model is based on coupling the acoustic impedance of various layers to obtain the overall acoustic isolation. The final parameter that indicates to us the value of the acoustic isolation is known as the sound reduction index (R). This parameter can be given as a function of the frequency or as an overall value. For that, it is necessary to introduce as input values in our computer application the characteristics of the impermeable plates and those of the absorbent acoustic materials. This computer application allows the characterization of each material separately. In addition, this application not only makes it possible to characterize the absorbent materials in a standard form from predictive models (Delany and Bazley 1970; Dunn and Davern 1986), but it also allows the introduction of specific values of the conducted tests carried out in the same laboratories of acoustic and materials by incorporating new natural or recycled materials (Ramis *et al.* 2010).

Sound reduction index

The sound reduction index (R) is obtained from the ratio of transmission $\tau(\theta)$ (transmitted energy) with respect to the incident as a function of the incidence angle on the wall test (Alba *et al.* 2001; Alba and Ramis 2003; Alba *et al.* 2003). We can use this variable to obtain a transmission coefficient in a diffuse field (τ_d) given by Eq. 1,

$$\tau_d = \frac{\int_0^{\theta_{\text{lim}}} \tau(\theta) \cos \theta \sin \theta d\theta}{\int_0^{\theta_{\text{lim}}} \cos \theta \sin \theta d\theta} \quad (1)$$

where θ_{lim} values represent the limit angle of our area, which is the major inclination with respect to the surface vector that we can get by impacting on the test wall. There has been some discrepancy with respect to the limit angle reported by different authors. For example, some authors have decided that the limit angle is 90° (*i.e.*, direction parallel to the wall test), however others indicate that it is difficult to get an incident above the 80° because of the limitations of design conditions (Ookura and Saito 1978; Trochidis and Kalaroutis 1986; Alba *et al.* 2004).

From this expression, we can obtain an estimate of the sound reduction index according to Eq. 2:

$$R = -10 \log \tau_d \quad (2)$$

Therefore, if we consider as a valid estimate of Eq. 1, then we can estimate an R value that depends on the limit angle of the incident sound.

In the case of a thin plate, infinite and elastic, with the mass per unit area m , which separates two regions of space, I and II, without any connection between them (see Fig. 1), there exist relatively simple expressions derived from an improvement of acoustical mass law (Ookura and Saito 1978; Trochidis and Kalaroutis 1986):

$$\tau(\theta) = \frac{(2\rho_o)^2}{\left(\frac{\omega^3 \cos \theta D \sin^4 \theta \eta}{c_o^5} + 2\rho_o\right)^2 + \frac{\omega^2 \cos^2 \theta (D\omega^2 \sin^4 \theta - mc^4)^2}{c_o^{10}}} \quad (3)$$

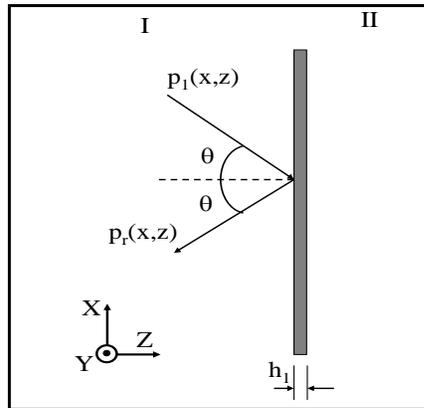


Fig. 1: Thin and impermeable plate separating two spaces

In the case of a diffuse field, *i.e.*, by applying (Eq. 1),

$$\tau_d = \frac{\int_0^{\theta_{lim}} \frac{(2\rho_o)^2 \cos \theta \sin \theta d\theta}{\left(\frac{\omega^3 \cos \theta D \sin^4 \theta \eta}{c_o^5} + 2\rho_o\right)^2 + \frac{\omega^2 \cos^2 \theta (D\omega^2 \sin^4 \theta - mc^4)^2}{c_o^{10}}}}{\frac{\sin^2 \theta_{lim}}{2}} \quad (4)$$

where D is the rigidity at the plate bending, η is the factor of total losses, c_o is the speed of propagation of sound in air, ρ_o is the density of the air, and ω is the angular frequency. This last expression allows us to solve the problem for thin impermeable plates.

Sound reduction index: Case of multi-layered partitions with absorbent material inside the air chamber

The new formulae for the calculation of the resonance and limit frequencies when the absorbent material was introduced in the air chamber are the following,

$$LLf = 43 \sqrt{\frac{m_1 + m_2}{m_1 \cdot m_2 \cdot d_{abs}}} \quad (5)$$

where m_1 and m_2 are the surface densities of each impermeable plates, and d_{abs} is the width of the air chamber and absorbent.

$$ULf = \frac{43}{\cos \theta_l} \sqrt{\frac{m_1 + m_2}{m_1 \cdot m_2 \cdot d_{abs}}} \quad (6)$$

$$f_L = \frac{c_0}{2 \cdot d_{cam}} \quad (7)$$

In this case, the computer application allows the user to choose the model corresponding to the absorbent material as well as to characterize the absorbent material through the absorption coefficient or the resistance to the flow by bands of eighths or global. Thus, the corresponding equations used are the following:

$$\tau(\theta) = \left| \frac{p_r}{p_i} \right|^2 = \left| \frac{p_{11}}{p_i} \right|^2 = \left| \frac{p_{32}}{p_i} \right|^2 \cdot \left| \frac{p_{31} \cdot p_{21} \cdot p_{11}}{p_{32} \cdot p_{22} \cdot p_{12}} \right|^2 \quad (8)$$

where:

$$\left| \frac{p_{32}}{p_i} \right|^2 = \left| \frac{2 \cdot Z_{32}}{Z_{32} + \frac{\rho \cdot c}{\cos \theta}} \right|^2 \quad (9)$$

$$\left| \frac{p_{31}}{p_{32}} \right|^2 = \left| \frac{Z_{31}}{Z_{32}} \right|^2 = \left| \frac{Z_{31}}{Z_{31} + Z_{m2}} \right|^2 = \left| \frac{Z_{22}}{Z_{22} + Z_{m2}} \right|^2 \quad (10)$$

$$\left| \frac{p_{21}}{p_{22}} \right|^2 = \left| \frac{\cosh \varphi_1}{\cosh(q_1 \cdot d_{abs} + \varphi_1)} \right|^2 \quad (11)$$

$$\left| \frac{p_{11}}{p_{12}} \right|^2 = \left| \frac{Z_{11}}{Z_{11} + Z_{m1}} \right|^2 \quad (12)$$

and:

$$\varphi_1 = \operatorname{arc} \coth \left(\frac{Z_{12} \cdot q_1}{\gamma_1 \cdot Z_0} \right) \quad (13)$$

$$Z_{12} = Z_{22} + Z_{m2} \quad (14)$$

$$Z_{22} = \frac{\gamma_1 \cdot Z_0}{q_1} \coth(q_1 d_{abs} + \varphi_1) \quad (15)$$

$$\gamma_1 = \alpha + j\beta \quad (16)$$

$$q_1 = \gamma_1 \cdot \sqrt{1 + \left(\frac{k}{\gamma_1}\right)^2} \cdot \sin^2 \theta \quad (17)$$

$$k_1 = \frac{2 \cdot \pi \cdot f}{c_0} \quad (18)$$

$$Z_0 = R + jX \quad (19)$$

$$Z_{12} = Z_{11} + Z_{m1} \quad (20)$$

$$Z_{11} = \frac{\rho \cdot c}{\cos \theta} \quad (21)$$

$$Z_{m1} = \eta_1 \cdot 2 \cdot \pi \cdot f \cdot m_1 \cdot \left(\frac{f}{f_{c1}}\right)^2 \cdot \sin^4 \theta + j \cdot 2 \cdot \pi \cdot f \cdot m_1 \cdot \left(1 - \left(\frac{f}{f_{c1}}\right)^2\right) \cdot \sin^4 \theta \quad (22)$$

$$Z_{m2} = \eta_2 \cdot 2 \cdot \pi \cdot f \cdot m_2 \cdot \left(\frac{f}{f_{c2}}\right)^2 \cdot \sin^4 \theta + j \cdot 2 \cdot \pi \cdot f \cdot m_2 \cdot \left(1 - \left(\frac{f}{f_{c2}}\right)^2\right) \cdot \sin^4 \theta \quad (23)$$

where γ_1 and Z_0 can be obtained with different models and theories of the characterization of the acoustic absorbent materials (Delany and Bazley 1970; Miki 1990; Allard and Champoux 1992), or with others empirical models based on experimental tests (Ramis *et al.* 2010; Del Rey *et al.* 2011, 2012).

EXPERIMENTAL

Materials

Composites materials were prepared from polypropylene (PP) (Isplen PP090 G2) delivered by Repsol-YPF (Tarragona, España) as polymeric matrix. We used mechanical pulp (MP) of softwood (*Pinus radiata*), known as stone ground fiber, from Zubialde S.A. (Aizarnazabal, España) as natural fiber reinforcement. Fiberglass produced by Vetrotex (Chambery, France) and provided by Maben S.L. (Banyoles, España) was used as synthetic fiber reinforcement. The principal features of the reinforcement fibers were reported in our previous works (López *et al.* 2011, 2012).

Methods

Preparation of the composites

Polypropylene composite materials were prepared with 20, 30, 40, and 50% (wt/wt) of mechanical pulp fibers; and with 20, 30, and 40% (wt/wt) of fiberglass. The components were mixed inside a Brabender plastograph internal mixer. The mixing procedure was carried out at 180°C for 10 minutes, and the rotor speed was about 80 rpm for the mechanical pulp fibers and 20 rpm for fiberglass. The obtained mixtures were pelletized with an Agrimsa Pelletizer. The pellets were then dehumidified with an oven at 80°C during 24 h.

Afterwards, the pellets were injection-moulded into a Meteor-40 injection machine (Matey & Solé) to obtain tensile specimens. The injection moulding temperatures were in the range of 175 to 190°C. The first and second pressures were 120 and 37.5 kgf/cm², respectively.

Mechanical characterization of the composites

Processed materials were placed in a conditioning chamber (Dycometal) at 23°C and 50% relative humidity during 48 hours, in accordance with ASTM D618, prior to testing. Afterward, composites were assayed for tensile properties by using a universal testing machine (Instron™ 1122) with load cell of 5 kN, using a cross head speed of 2 mm/min. The Young's modulus was obtained through the use of an extensometer, according to the ASTM D790. The specimens were tested and the results presented are the average of at least five samples.

Acoustic characterization of the composites

We conducted several tests to obtain the sound absorption coefficient in normal incidence according to the UNE-EN-ISO 10534-2, and the specific resistance to flow of composite materials using the method of Ingard and Dear (1985), which is not a standardized test but is a widely used method in the research field to characterize materials with acoustic aim. These two magnitudes are used to characterize an acoustic absorbing material to airborne noise when the composite material is inserted between two partitions. In Fig. 2, we present the experimental equipment used for these tests.

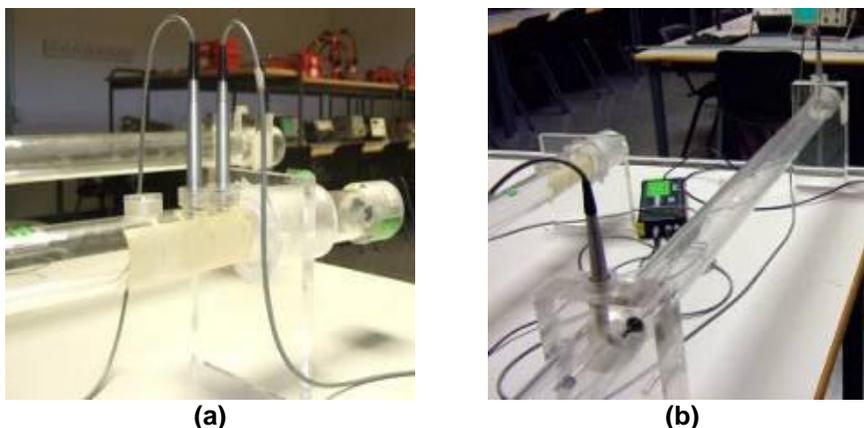


Fig. 2. Experimental equipments for measurement of (a) sound absorption coefficient (UNE EN ISO 10534-2:2004), and (b) the specific resistance to flow (Ingard & Dear method)

Prediction of the acoustic insulation

The final parameter that indicates to us the value of the acoustic insulation is the sound reduction index (R). This parameter can be given as a function of the frequency or as an overall value. To do this, a computer application (AISLA) has been developed that not only makes it possible to characterize the absorbing materials on a standardized basis from predictive models (Delany and Bazley 1970; Dunn and Davern 1986), but also allows one to introduce specific values of the tests conducted in the same laboratories of acoustic and materials (Ramis *et al.* 2010; Del Rey *et al.* 2011, 2012). The theoretical basics of AISLA computer application are described in the Introduction.

RESULTS AND DISCUSSION

Tensile properties of MP or FG composites are summarized in Table 1. It can be seen from the table that FG composites had better tensile strength in comparison to MP composites, even at lower reinforcement percentage of fiberglass. It can also be seen that tensile strength of FG composites increased dramatically with the reinforcement percentage of fiberglass. However, the tensile strength of MP composites slightly increased from 28.5 to 31.5 MPa with an increase of the fiber content from 20% to 50%. This is can be explained by the poor adhesion between the two components of the composite, due to the polar nature of natural fibers and non-polar groups characteristic of polypropylene matrix.

Table 1. Tensile Properties and the Critical Frequency of Composite Materials

| Materials | %, FG or MP content (w/w) | Tensile strength (MPa) | Young Modulus (GPa) | Elongation at break (%) | Critical Frequency (Hz) |
|-----------|---------------------------|------------------------|---------------------|-------------------------|-------------------------|
| PP + FG | 20 | 50.7 | 4.5 | 3.1 | 2371 |
| PP + FG | 30 | 58.5 | 5.9 | 3.0 | 2071 |
| PP + FG | 40 | 67.1 | 7.4 | 2.4 | 1845 |
| PP + MP | 20 | 28.5 | 3.3 | 3.4 | 2595 |
| PP + MP | 30 | 28.5 | 4.4 | 2.4 | 2247 |
| PP + MP | 40 | 29.5 | 4.9 | 1.9 | 2141 |
| PP + MP | 50 | 31.5 | 6.3 | 1.3 | 1883 |

FG: Fiberglass, MP: Mechanical Pulp.

In relation to Young's modulus, it is evident that the composites' stiffness increased linearly with the fiber content for both mechanical pulp fibers and fiberglass. It is well known that the rigidity of the composites is generally affected by the dispersion and the amount of the reinforcement (Karmaker and Youngquist 1996; Vilaseca *et al.* 2010). According to this, the observed lineal tendency of this property with the fiber content is representative of a good dispersion degree of the reinforcement inside the composite. These results are in good agreement with the expected increase of materials' stiffness and the reduction of the capacity to sustain the plastic deformation (Vilaseca *et al.* 2010). In relation to the elongation at break, it can be shown that this property decreased with increasing proportion of either FG or MF reinforcement. Major reduction was found for 50 wt% mechanical fiber content. Hence, the stiffening of the matrix due

to the addition of natural fibers caused lower deformability of the matrix; thereby, the elongation at break decreased. Consequently, higher fiber content can cause failure, which results in reduction in elongation at break for higher fiber content.

The tensile properties of MP composites were really low when the coupling agent was not used. This behavior indicates that mechanical pulp fibers from *Pinus radiata* can be used only as filler in wood-like plastic composites due to the low mechanical properties compared to FG composites. In order to increase compatibility of MP with the polypropylene, modification of the matrix polymer and treatment of mechanical pulp itself are potential options that may significantly improve the mechanical properties of the MP composites. In this respect, maleic anhydride modified polypropylene has demonstrated better compatibility with MP fibers when compared to PP, leading to significant increase in mechanical properties (Lopez *et al.* 2012).

The critical frequency is defined as the frequency at which the airborne acoustic wavelength matches the plate bending wavelength. It is also called the coincident frequency. It is evident that the critical frequency of both composites decreased with the increase of the fiber content, as can be seen in Table 1. This gave an indication of the loss of the acoustic isolation property of both families of composites.

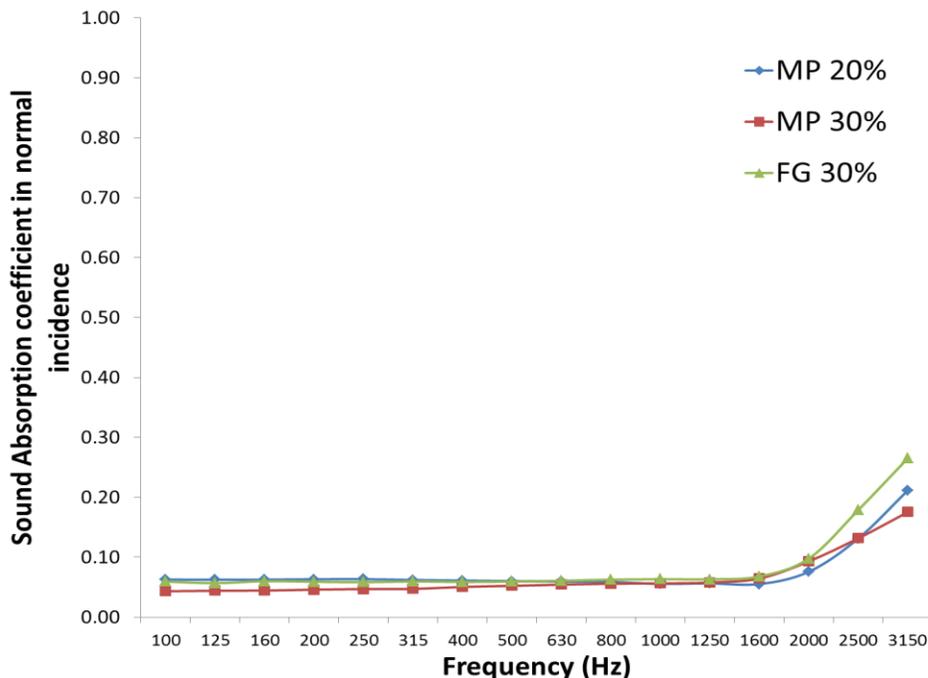


Fig. 3. Sound absorption coefficient vs. frequency of some studied composite materials

In general, the composite materials can be manufactured to show a range of acoustic properties. By modifying such properties as weight and material formulation, different acoustical properties can be achieved depending on the specific application requirements. Figure 3 shows the values of the sound absorption coefficient in normal incidence versus frequency of composite materials made with 20% and 30% of mechanical pulp and 30% of fiberglass. For composites made with 20% and 30% MP

concentration, it can be seen that the sound absorption coefficient increased slightly up to 1250 Hz for the 20% MP composite, after which the 30% MP composite showed a better sound absorption coefficient; however, above 2500 Hz, the 20% MP led to a sharp increase in sound absorption coefficient. The 30% FG composite exhibited similar sound absorption coefficient pattern up to 1250 Hz, after which the sound absorption coefficient rose more rapidly for the FG composites than the MP composites as the frequencies increased beyond 1250 Hz. On the other hand, all the composites studied showed values of the resistance to flow over 1000 kPas/m².

In Fig. 4, one can observe the values of the acoustic isolation (dB) as a function of the frequency for both MP and FG composites as a possible impermeable single-layer. In all figures, the results are compared with the acoustic isolation of gypsum plasterboard.

On the other hand, the predictions of the acoustic isolation for the double-layers with air chamber or with absorbent material in its interior are presented in Fig. 5. The thickness of the air chamber was supposed to be about 40 mm. The considered absorbent material was wool polyester RC with 20 mm thickness, 500 g/m² of surface density, and the resistance to the flow was 2 kPas/m². This material was chosen as an absorbent material because it is recognized in the Spanish Technical Building Code (Real decreto 1371/2007) and it is frequently used in the light-weight building partitions referred (Del Rey *et al.*, 2011). The double-layers were compared with the acoustic isolation of gypsum plasterboards.

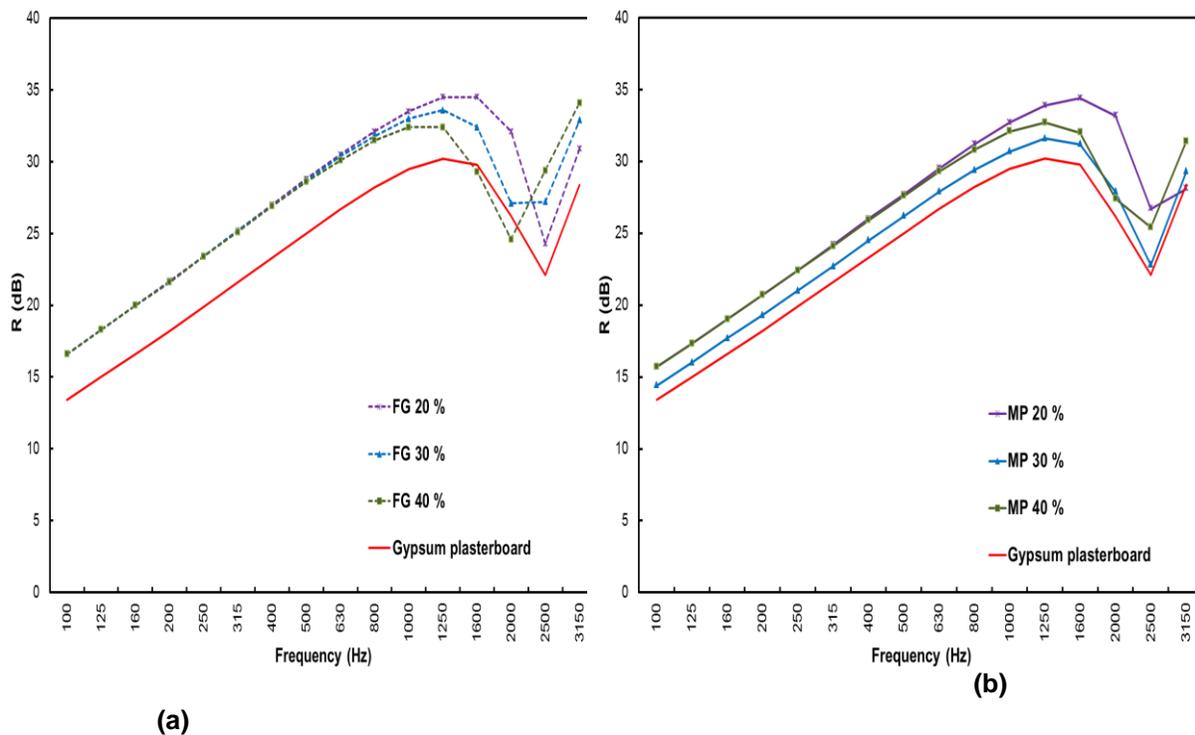
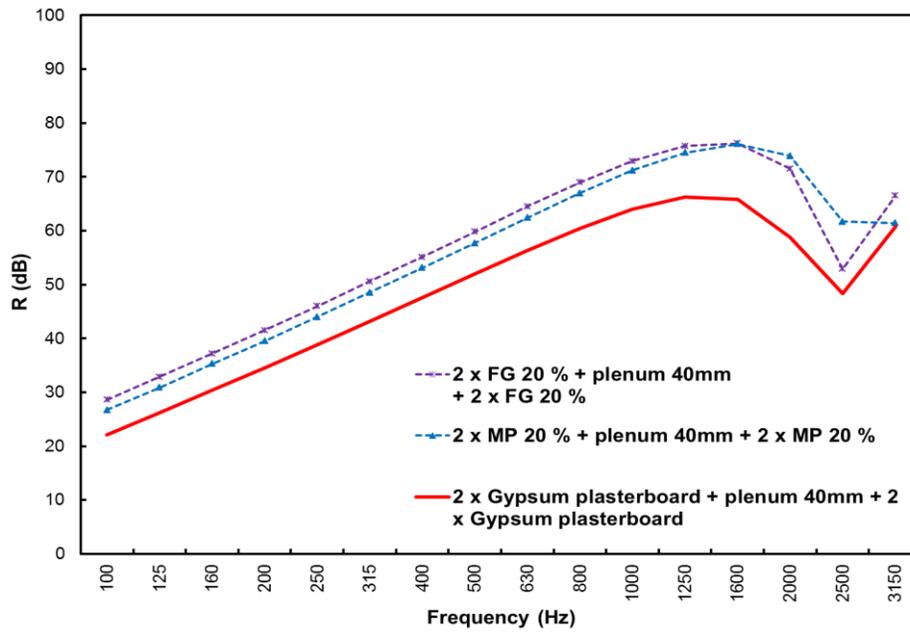
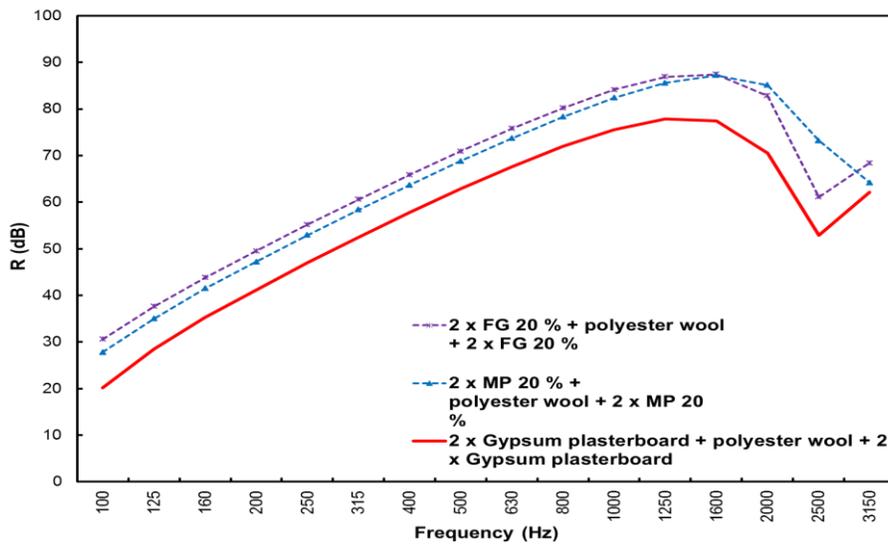


Fig. 4. Prediction of the acoustic isolation for MP and FG composites considered as an impermeable single-layer: (a) fiberglass, (b) mechanical pulp fibers. Comparison with gypsum plasterboard



(a)



(b)

Fig. 5. Prediction of the acoustic isolation for double-layers of MP and FG composites: (a) without absorbing material, and (b) with absorbing material between the two layers

In view of these reported results, it can be confirmed that the MP and FG composites can be considered a good acoustic isolating material. The values of isolation were exceeded in all frequencies of the spectrum studied for gypsum plaster-board. This material is commonly used to achieve acoustic insulation in buildings. In contrast, none of these two families of composites present properties of an acoustic absorbing material. This is reflected in the high values of the resistance to flow. Thus, due to its nature as an impermeable layer and having suitable mechanical properties, such as the critical frequency, it appears possible to use these green-composites as acoustic solutions of light-weight building material to reduce acoustic transmissions. The curves of acoustic isolation of the prepared composite materials with fiberglass and mechanical pulp are always above the curves of the gypsum plasterboards. Moreover, the isolation values of the MP composites vary with the fiber content and not for FG composites. In the case of double-layers, there is a small difference between the two families of composite materials and not between fiber percentages inside these materials. Therefore, for certain range of applications it can be concluded that these composites offer a good alternative.

CONCLUSIONS

1. Mechanical pulp (MP) can be used only as filler for the preparation of composites when no coupling agent is used. In this case, the properties are similar to those obtained in wood-plastic composites.
2. Taking into account the obtained level of sound reduction index and the critical frequency of all prepared composite materials, they can be used as a possible acoustic solution of light-weight building materials.
3. All lightweight structures with polypropylene (PP), regardless of the type of reinforcement, show isolation values as some solutions that appear in the Technical Building Code. The same conclusion is obtained when the study focuses on multi-layer partitions, which is a common format used in building.

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