Influence of fire treatment on acoustic performances of vegetal wools

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RESUME Vegetal wools are performant sustainable insulation materials. However, their flammability can restrict their application, and therefore a fireproof treatment, that is environmentally friendly to respect their bio-based nature, needs to be done. This study aims to evaluate the impact of two phosphorus-based fireproof treatment methods, pulverization and impregnation, on the density and acoustic properties of hemp fiber stacks. Pulverized fibers had a higher residue amount, in the thermogravimetric analysis, which indicates a higher protection from the char layer. From the acoustic perspective, the treated fibers presented a lower sound absorption than the reference. The density can be lowered after the treatment, but no correlation with the treatment method or condition was observed.

Mots-clefs Bio-based material, vegetal wool, flammability, acoustic

I. INTRODUCTION

Bio-based materials are known to contribute to a sustainable development of the construction industry, by having a lower carbon footprint and low embodied energy when compared to traditional materials, especially when the timing of carbon storage is taken into account (Pittau et al., 2018; Zieger et al., 2020; Galimshina et al., 2022). Following this principle, the vegetal wools emerge as a sustainable alternative to insulation, due to its high acoustic and hygrothermal performances (Piégay et al., 2020, 2021; Fedorik et al., 2021). Yet, the vegetal wools are flammable (Freivalde et al., 2014; Amziane and Collet, 2017; Moussa et al., 2020). Therefore, their application is limited to the core of wall systems having outer layers that would protect the vegetal wools against fire. By reducing the flammability of the vegetal wools, their application can be expanded to acoustic insulation of indoor living spaces, such as suspended ceiling tiles or acoustic baffles.

Furthermore, it reinforces the benefits of vegetal wools as an alternative to traditional insulation materials.

Many studies have focused on the improvement of the fire properties of vegetal fibers, however, following the purpose of a sustainable development in buildings, it is logic to propose the use an environmentally friendly fireproof treatment. In this scenario, phosphorus-based treatments could be an interesting solution, as they are considered to be non-toxic, as opposed to halogenated flame retardants that are associated with environmental and health concerns (Costes et al., 2017; Hajj et al., 2018; Taib et al., 2022; Taibi et al., 2022). The phosphorus can act by changing the degradation mechanism of the cellulose, avoiding the formation of levoglucosan, a reactive substance, and favoring the dehydration and the formation of char, which is a carbonaceous layer that protects the inner layers of the fiber (Hajj et al., 2020). Also, the char protection performance can be enhanced by the association of phosphorus with nitrogen, to cause an intumescent effect when burning. This phenomenon occurs when three components are presented in the same system: an acid source (phosphorus-based flame retardant), a carbonizing agent (the natural fibers), and a blowing agent (gas release by nitrogen-based products) (Laoutid et al., 2009). In this case, not only the char will be formed, but it will also expand, isolating the inner layers even more.

When manufacturing vegetal wool panels for building applications, it is essential to identify a treatment that can be applied at an industrial scale. That also implies on finding an adequate application method to limit the amount of flame retardant in relation to the quantity of fibers treated, while improving the reaction to fire from them. When treating with phosphorus-based products, frequently, studies reduce the flammability of natural fibers by grafting through irradiation or by impregnating the fibers in solution and grafting by heating (Sonnier et al., 2015; Hajj et al., 2018; Moussa et al., 2020; Hajj et al., 2020; Antoun et al., 2022; Taibi et al., 2022). Despite being two effective methods, some limitations may hinder the use of these techniques on a large scale. Even if the irradiation method seems effective, it requires sophisticated equipment that does not currently appear suitable for use on an industrial level. On the other hand, the impregnation in solution allows the treatment of fibers in a large scale, but through the use of large quantities of solution and the need to dry treated fibers in an oven.

Another point to be considered when using the impregnation method is how aggressive is the treatment. Studies show that low temperatures can provide enough energy to graft the phosphors on aromatic hydroxyl groups which are found in the lignin. However, in order to graft a higher amount of phosphorus and obtain natural fibers with lower flammability, a higher temperature is needed to graft the phosphorus in the cellulose (Hajj et al., 2018; Moussa et al., 2020; Antoun et al., 2022).

There is, however, a lack of information related to the impact that the fireproof treatment could cause in performance of the vegetal wools, as an acoustic material. It is known that the treatment will modify the chemical composition, but it is uncertain of its impact on the microstructure of the fibers, which are strongly linked to the acoustic performance of a material (Chabriac et al., 2016; Piégay et al., 2018, 2021).

Therefore, based on these considerations, this paper aims to evaluate two environmentally friendly fireproof treatment methods, and their impacts on the acoustic performance of hemp fibers. One method follows the “traditional” chemical grafting method, commonly found in the literature, and the second one is a less common method that aims to treat the hemp fibers with less amount of flame retardant.

II. MATERIALS AND METHODS

Hemp fibers were fireproof treated with phytic acid, a bio-based source of phosphorus commonly found in seeds and grains, and urea, as a source of nitrogen, through two different methods and two different conditions. The first method is impregnation, which consisted on the immersion of 300g of fibers in a solution containing 10% of phytic acid and 30% of urea for 1 hour, with a solution to fiber mass ratio of 10, followed by a heating step in oven at 80°C (mild condition) or 120°C (intermediate condition) for 2 hours. This treatment condition was chosen and adapted from the methodology and the results of (Hajj et al., 2018, 2020; Moussa et al., 2020; Antoun et al., 2022). After that, the fibers were washed three times with distilled water, by immerging them, manually mixing them for 30 seconds and then pouring the liquid. After each washing, the color of the water became less brown and more transparent. Three washing steps were considered enough to significantly remove the flame retardant molecules that were not grafted, which is the same number used by (Hajj et al., 2018).

The second application method tested was through pulverization, which consisted on spraying the same solution over 300g of fibers and heating them at 80°C or 120°C, depending on the condition tested. In this method, the fibers were not washed at the end of the treatment. For this method, the solution to fiber mass ratio was 1. Both treatments methods, as well as the solution used, are presented on Figure 1. The Table 1 presents the nomenclature used for the samples presented in this study.

TABLE 1. Sample's nomenclature

Nomenclature	Treatment	Condition
REF	-	-
IMP_MILD	Impregnation	Mild
IMP_INT	Impregnation	Intermediate
PUL_MILD	Pulverization	Mild
PUL_INT	Pulverization	Intermediate

Treated and non-treated fibers had their true density measured by helium gas pycnometer, in dried conditions. They had their mass stabilized in oven at 60°C, until three consecutive measurements, in three days, had a variation lower than 0.5%. Their thermal degradation was measured by thermogravimetric analysis (TGA) in air atmosphere until 800°C, using a temperature increase rate of 10°C/min and samples of 17.5±2.5mg.

(a) (b) (c)

FIGURE 1. **(a) Phytic acid and urea solution; (b) Impregnation method; (c) Pulverization method**

Characterization of sound absorption (noted α) was carried out using an impedance tube in accordance with standard ISO 10534-2 (NF EN ISO 10534-2, 2003) at normal incidence. The diameter of the impedance tube, which is shown in Figure 2a, (Akustik Forschung - AcoustiTube AFD 1000/1200) used in this study is 10 cm. Prior to the test, the fibers were kept in the measurement room for three days, in order to stabilize their temperature and humidity. The room conditions were 23.5±0.5°C and 40±5% of relative humidity. The impedance tube test was performed using the three positions method without cavity. This method was firstly developed in (Iwase et al. 1998) and has been adapted for microphones positioned outside the tube in (Salissou & Panneton 2010). Two cylindrical specimens for each of treated and non-treated hemp fibers, with 100mm of diameter and 100mm of height were prepared, by simultaneous addition and compaction of the fibers, until the totality of the fibers, weighted for the desired density of 60kg/m^3, were added to the sample holder. Due to the direction of the compaction, the fibers orientation is favored to be horizontal. A specimen of the reference fibers can be observed inside and outside of the sample holder in the Figures 2b and 2c. The test was performed on the both sides of the specimens, and the sound absorption curves show the average of the four measurements.

(a) (b) (c)

FIGURE 2. **(a) Impedance tube; (b) Specimen of hemp fibers; (c) Specimen inside of sample holder**

III. RESULTS AND DISCUSSION

The water content and the density results for the dried fibers are presented on the Table 2. The water content results shows that the treated fibers stabilize with a lower amount of water content, when in comparison with the reference fibers. This indicates that the interaction with the water vapor is changed, possibly due to modification on its pores and microstructure. The density of the dried fibers can be separated in two groups: one with the reference and the fibers treated in mild condition, having a density near 1.50g/cm^3; and the second one with the fibers treated in the intermediate condition with 1.48 and 1.49g/cm^3 for the impregnated and pulverized samples, respectively. The reference hemp fibers' density are consistent to values found for non-treated fibers in the literature (Pereira et al., 2015; Piegay, 2019). It is worth noting that the condition IMP_INT was the one that showed the lowest water content and fiber density. The same behavior was not observed by the PUL_INT sample.

TABLE 2. **Water content and density results**

Sample	Water content (%)	Density dried fibers (g/cm^3)
REF	6.25%	1.503±0.004
IMP_MILD	4.75%	1.504±0.002
IMP_INT	4.55%	1.479±0.003
PUL_MILD	4.65%	1.496±0.003
PUL_INT	4.95%	1.489±0.002

The evolution of the weight (TGA) of the fibers as a function of the temperature, as well as its derivate (DTG), are presented on Figure 3a, with straight or dashed lines, respectively. The results for reference fibers (black) shows that the thermal degradation occurs in three major steps. The first one consists on the loss of humidity until 150°C. The second peak, at about 230°C, corresponds to the degradation of hemicelluloses, followed by the degradation of the cellulose, represented by the larger peak in the DTG curve. Afterwards, a third mass loss is observed, which is a further degradation of the products of the combustion, known as humins, which are a product of the pyrolysis and the acid-catalyzed degradation of furanoid and benzoid moieties (Bausch et al., 2021). This step occurs between 380°C and 520°C for the reference fibers.

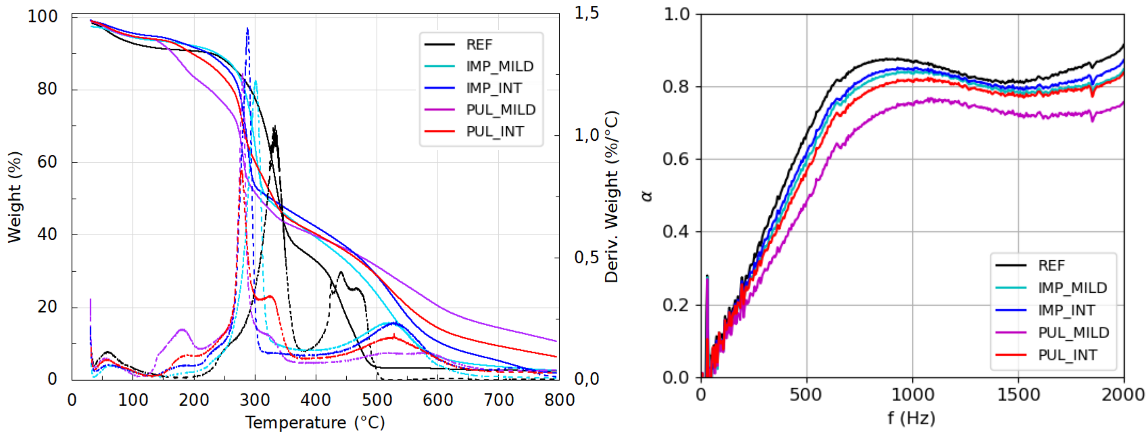
In comparison with the reference, it is possible to see an earlier thermal degradation for the treated fibers, which is due to the catalyst effect of the phosphorus on the degradation of the cellulose (Hajj et al., 2018; Antoun et al., 2022). The maximum of the mass loss derivative of the for the reference was at about 333°C, whereas for the treated fibers it ranges between 277°C and 301°C. The second and main effect of the phosphorus treatment is observed by comparing the residue values at 500°C. The reference fibers are almost entirely degraded, having about 3.6% of the initial mass remaining. The IMP_MILD, IMP_INT, PUL_MILD and PUL_INT presented residues of 23.7%, 28.8%, 31.4% and 28.6%, respectively. For the impregnated fibers, the residue increased when the temperature of the treatment increased. Similar results, with other phosphorus-based products and natural fibers, were previously reported in the literature (Gaan and Sun, 2007; Hajj et al., 2020; Moussa et al., 2020). The pulverized fibers presented a higher residue and the higher value was obtained for the fibers treated in mild condition.

Comparing both methods, the pulverization method presented a higher residue in mild condition, but at intermediate condition both methods presented the same values at 500°C. However, at higher temperatures, it is observed that the pulverization treatment induces a higher amount of residue than the impregnated ones.

The results on sound absorption of hemp fibers samples as a function of the frequency, obtained through the impedance tube test, are presented on Figure 3b. At low frequencies, especially lower than 300Hz, the fibers do not perform well, in terms of sound absorption, with a α value of 0.40 for the reference fibers, as expected for this type of material (Piégay et al., 2018). From 600 Hz and onwards, the reference presented a sound absorption higher than 0.80. Overall, the results showed that the reference had a better acoustic performance, followed by IMP_MILD, IMP_INT, PUL_INT and PUL_MILD, indicating that the treatment decreased the acoustic performance. At 750 Hz, for example, the reference fibers presented a α value of 0.85, whereas the PUL_MILD fibers presented a α value of 0.69.

As the acoustic performance of a material, is strongly associated with its microstructure (Piégay et al., 2021), it is hypothesized that the negative impact of the treatment is related to an increase on the representative fiber's radius, due to the grafting of phosphorus. In fact, airflow resistivity is directly correlated to fiber diameter (Tarnow, 1996). The more the diameter increases, the more the airflow resistivity decreases, as does sound absorption (Piégay et al., 2018).

Also, the treatment might also affect the porous structure present in the fibers, which can also be impactful for the acoustic performance of them. To confirm these hypotheses, studies on the air resistivity of the material, associated with images obtained from scanning electron microscopy will be performed.



(a) (b)

FIGURE 3. **(a) TGA and DTG results; (b) Sound absorption results**

IV. CONCLUSION AND PERSPECTIVES

This study presented the effects of two different fireproof treatment methods, with two different temperature conditions, of hemp fiber stacks. The treatment consisted on the association of a phosphorus-based product (phytic acid) and a nitrogen-based one (urea). The density, the thermal degradation, as well as the acoustic properties were evaluated. A small reduction of the density can occur, but no direct correlation with the treatment condition or method can be made. Further investigations with a scanning electron microscope will provide additional information of the microstructure of the fibers after the treatment to support the results obtained.

The phosphorus was successfully grafted in the hemp fibers, as observed through the TGA results. Moreover, the pulverized fibers achieved a higher residue amount, when compared to the impregnated samples, which indicates that the char layer formed was able to better protect the inner layers of the fiber. Reaction to fire tests will be made to complement this analysis. Lastly, the acoustic performance was measured by tube impedance, and it was observed that the treatment had a negative impact on the sound absorption of the hemp fibers, especially for the pulverized fibers. To explain the effects of the treatments, a more in-depth analysis of the characteristic parameters of the porous and fibrous microstructure of the samples is required, such as porosity, airflow resistivity, tortuosity, among others.

The characterization of microstructure changes due to fire treatment will enable an understanding of these results and the modification of treatment conditions to limit the impact on acoustic performance.

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