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Thermal Insulation Blocks Made of Sunflower Pith Particles and Polysaccharide-Based Binders: Influence of Binder Type and Content on their Characteristics

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Abstract. Co-product of sunflower cultivation, pith of stem has a little exploited insulating potential. Blocks in which pith particles are glued together using a starch-based binder have already been obtained. However, they are highly water-sensitive. Replacing this binder with others has been considered here.

Polysaccharide-based binders were tested, chosen for their more hydrophobic character: sodium alginate, chitosan, *Citrus* pectin, and a modified starch. Like starch, these binders are physically binding. They are first solubilised in water (except chitosan, dissolved in 2% acetic acid). The solution is then mixed with pith particles before cold compression molding for 90 s.

A 10% binder content was initially considered. The blocks were all cohesive with a dry density from 36 to 42 kg/m^3). Their performances were assessed through water absorption capacity and resistance *via* capillary absorption tests on wet sponges, mechanical test and thermal conductivity.

Chitosan and pectin-based blocks show the best properties, particularly concerning water resistance and mechanical properties. The pectin-based block has improved its elastic modulus by 40% compared to a starch-based block. The pectin-based block in its case absorbs 2.7 times less water than starch. Finally, thermal conductivities of pectin and chitosan-based pith blocks are in the same order of magnitude as for starch (39.8-40.1 mW/m.K), and close to values from commercial materials (*e.g.*, polystyrene).

Pectin and chitosan were also tested at three rates (5%, 10% and 15%). A significant improvement in the blocks' compressive strength was observed with the increase in binder rate, while thermal conductivities varied little.

1 Introduction

The use of biobased particles as raw materials in building materials is expanding, mainly due to their good environmental performances. New biobased insulating materials can be considered in a logic of sustainable construction, as an alternative to conventional insulating materials derived from non-renewable resources (*e.g.*, petrochemicals) or having an important embodied energy.

Some recent works highlighted the interest of using stems, and more particularly the pith located in their middle, of sunflower or corn, in order to develop insulating materials with interesting hygric properties (Bacoup et al. 2019, Palumbo et al. 2016, Mati-Baouche 2015). Use properties of these insulating pith panels seem very promising, with a very low thermal conductivity, good moisture buffering capacity or high water vapour permeability (Sabathier et al. 2017, Palumbo et al. 2016). Various binders were used to glue the pith particles together, such as starch (Sabathier et al. 2017, Verdier et al. 2019), alginate (Palumbo et al. 2016), arabic gum, natural latex or even sodium caseinate (Bacoup et al. 2019). Preliminary results were obtained in (Verdier et al. 2019) concerning the durability from the point of view of the microbial and fire resistances. However, there is still very little scientific data about the durability of this type of biobased materials, particularly concerning their water resistance, which might be problematic in case of the use of a water-soluble binder.

The objective of this study is to test different biobased binders to find a viable alternative to starch allowing the insulating blocks to be more water-resistant. It is indeed important to ensure a good water durability in case of an accidental exposure to liquid water. The capillary test allows to assess this performance, but it is also essential to check their minimal use properties, *i.e.*, thermal and mechanical characteristics, so that they are not degraded compared to the pith panel glued with a starch-based binder.

2 Materials and Methods

2.1 Raw materials

Sunflower pith particles

The sunflower pith comes from a local agricultural cooperative (Ovalie Innovation) located in the South-West of France. Sunflower stems were crushed and the pith was then separated from the bark. A mechanical sieving was realized for 10 min to keep the particles greater than 4 mm. These coarser pith particles represented 26.3% (w/w) of the pith mass. The eventual remaining bark was manually removed. The loose density of the sieved particles was around 21 kg/m^3. This density was determined following the Rilem protocol (Amziane et al. 2017).

Polysaccharide-based binders

Five polysaccharide-based binders were studied: a starch (used as the reference), a sodium alginate (a linear biopolymer made up of carbohydrate units of alginic acid type and extracted from brown algae), a chitosan (a polyoside composed of randomly distributed D-glucosamine and N-acetyl-D-glucosamine units and produced from chitin, the component of the exoskeleton of crustaceans), a *Citrus* pectin (a polyoside made up mainly of galacturonic acid units and with high methylation degree, which is the co-product of the orange juice industry), and a chemically modified starch (sodium octenyl succinate starch). All these binders are commonly available in the market, and at reasonable cost (with starch glue being the least expensive, and sodium alginate and chitosan being the most expensive). They were diluted in distilled water with a dry binder to solvent ratio (by weight) of 8.8%, except for chitosan, which was diluted in a 2% acetic acid solution (as in Mati-Baouche 2015) with a dilution ratio of 4.4%. The dilution rate of modified starch has been increased to 17.6% and 100% because of the poor quality of specimens with only 8.8% modified starch as binder. Concerning the mixing of the binder with the solvent, starch, alginate and chitosan were manually stirred at 20°C with a 45-minute rest. Pectin and modified starch, for their part, were mechanically stirred at 60°C for 30 min.

2.2 Manufacturing process

In order to manufacture the insulating panel samples, the sunflower pith was manually mixed with the diluted biosourced binder. The particleboard thickness targeted was 5 cm. To obtain the panel as light as possible and with a sufficient mechanical handling, the bulk pith quantity to insert in the mold was 7.5 cm high, as it was determined in a previous study (Sabathier et al. 2017). The mold, lined with greaseproof paper on each surface to avoid the particles to stuck to the mold during demolding, was filled in three layers with manual pre-compaction during 5 sec per layer. Then, a compression stress was applied during 90 sec at around 20 bars. The specimens were directly demolded after compaction.

Three blocks of 15 × 15 × 5 cm^3 dimensions per formulation were manufactured for this study, which were cut with a band saw for the different tests. Density was measured after drying at 60°C and sawing to obtain smooth surface and thus, more precise values.

Different formulations were manufactured in order to compare the influence of the nature of the binder and then of the binder rate, on the properties. After preliminary tests, the binder ratio used was

10% (dry mass of the binder over dry mass of the pith), except for the modified starch, where 30% was also tested as the specimen made with only 10% modified starch had a lot of loss of matter when cutting. Two binders presenting high potential after the first experiments were selected in order to test 5% and 15% binder ratios: chitosan and pectin.

Tab. 1 summarizes the various formulations manufactured, with their dilution rate, their binder ratio and their dry density.

Tab. 1: Mix proportions and dry density of the insulating blocks

Reference	**Binder**	**Dilution** $W_{dry\ binder}/W_{solvent}$ **(%)**	**Binder ratio** $W_{dry\ binder}/W_{pith}$ **(%)**	**Dry density (kg/m^3)**
S10	Starch	8.8	10	39.1 ± 0.1
A10	Alginate	8.8	10	40.0 ± 0.1
C10	Chitosan	4.4	10	39.7 ± 0.5
P10	Pectin	8.8	10	41.5 ± 0.4
MS10	Modified starch	17.6	10	36.2 ± 0.8
MS30	Modified starch	100	30	48.5 ± 0.2
C5	Chitosan	2.2	5	37.8 ± 0.7
C15	Chitosan	6.6	15	43.6 ± 2.1
P5	Pectin	8.8	5	38.8 ±0.2
P15	Pectin	13.2	15	45.9 ± 0.8

2.3 Capillary absorption and water resistance

Three specimens with dimensions of 40 × 40 × 30 mm^3 and at equilibrium with environment (25°C and 60% RH) were put on sponges placed in a water container, with water level around half of the sponge height (Fig. 1). The mass absorption was measured at defined times: 15 s, 30 s, 1 min, 1.5 min, 2 min, 5 min, 10 min, 20 min, 30 min, 45 min, 1 h, 1.25 h, 1.5 h, 2 h, etc. Visual damage was also assessed from no damage to full loss of cohesion between the pith particles. The measurement of the mass absorption was stopped when there was a loss of several pith particles, as the measures were not representative anymore.

Fig. 1: Capillary absorption test

2.4 Thermal conductivity

Thermal conductivity was measured with the hot wire method with a 0.5 mW/m.K precision on a pair of specimens with 13 × 7 × 4 cm^3 dimensions from the same block, with a weight on the top to ensure a good contact between the two specimens. Thermal conductivity was assessed on both dry specimens and specimens at equilibrium with environment (25°C and 60% RH).

2.5 Mechanical strength

Mechanical strength was assessed by compression of four specimens stabilized at 60% RH and 25°C per formulation, with dimensions of 40 × 40 × 30 mm^3. A hydraulic press with 5 kN sensor and a rate of 3 mm/min was used for this test.

The compressive strength was considered to be the limit between the elastic and elasto-plastic behaviors on the axial stress – axial strain curve, which happened at around 6% strain in all cases. The apparent global stiffness modulus (E) was estimated as the slope of the linear regression of the curve corresponding to the elastic behavior, lower than 4% strain.

3 Results

3.1 Capillary absorption and water resistance

Fig. 2 represents the water absorption of the various formulations, expressed in weight percentage per unit area (wt%/cm^2). This unit allows the comparison between the specimens, even if the area differs slightly. The influence of the binder nature on the amount of absorbed water by the material can be observed in this figure.

This graph includes only the water absorption of the specimens before losing pith particles, which explains the drop of some curves (S10, P10 and MS30) when one of the three tests was stopped, which may have distinct water absorption values than the two other specimens.

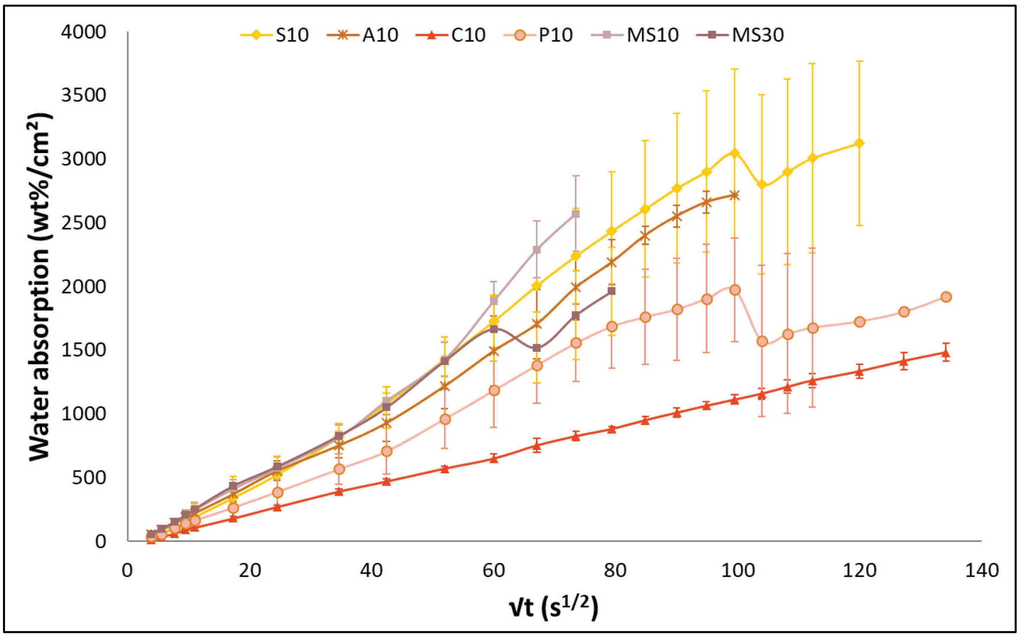

Fig. 2: Water absorption by capillarity of the various formulations

It can be observed in Fig. 2 that S10 specimens show the highest water absorption along with A10, MS10 and MS30, which are within the standard deviations. MS10 and MS30 curves stop before the curves of the other formulations as these specimens fell apart quickly, after around 1.25 h of testing. Thus, the replacement of the starch binder by alginate or modified starch does not reduce the water sensitivity of the insulating panel.

However, chitosan and pectin seem to improve the water behavior of the pith specimens. After 2.5 h of capillary test, S10 absorbed a water quantity of 2900 wt%/cm^2, whereas C10 and P10 absorbed only 1062 wt%/cm^2 and 1903 wt%/cm^2, respectively. Thus, the replacement of starch by chitosan and pectin led to a decrease of 63% and 34%, respectively, of the water absorption value.

In addition to the water absorption measurements, the water resistance of the specimens was also assessed. Fig. 3 represents the duration of the capillary test before the specimens lose several pith particles. The formulations MS10, MS30 and A10 present the lower test duration, between 1 h and

5 min for MS10 and 1 h and 50 min for A10. Formulations made with pectin and chitosan, for their part, present similar test duration than S10, considering their standard deviations. In some cases of these three formulations (P10, C10 and S10), the test durations even exceeded 4 h.

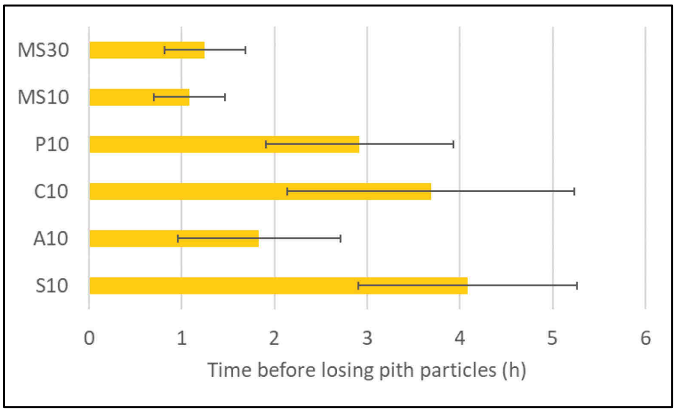

Fig. 3: Duration of capillary test before the specimens lose pith particles

Thus, it is reasonable to assume that pectin and chitosan allow to reduce the water absorption of the pith block while maintaining a good water resistance. These two binders thus show a potential to replace the starchy one in order to improve the composite water properties. As complementary study, an optimisation of the ratio of these two binders has been carried out in a second time to assess its influence on the properties. The water absorption values of formulations made of 5%, 10% and 15% of pectin and chitosan are presented in Fig. 4.

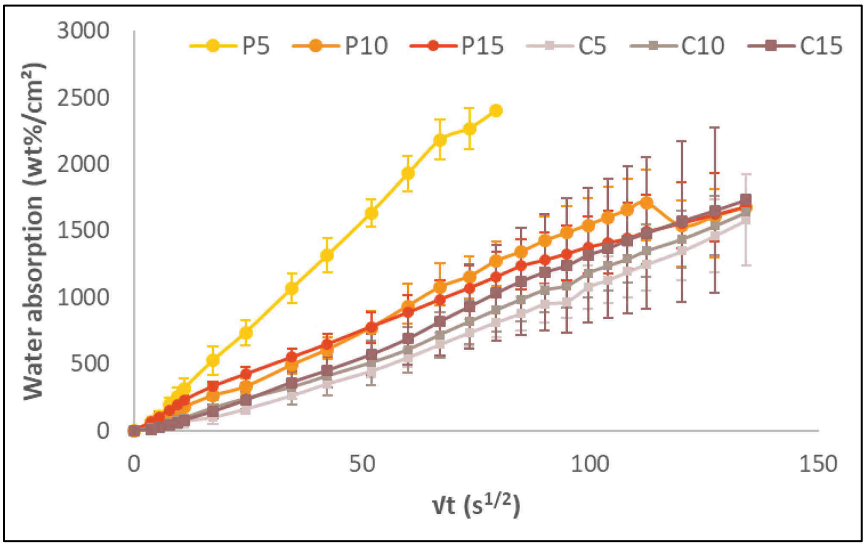

Fig. 4: Water absorption by capillarity of pectin and chitosan's formulations

A strong difference in water absorption can be noticed between the specimens made with 5% of pectin (P5) and those made with 10% and 15% (P10 and P15), which present very similar values considering the standard deviations (Fig. 4). Indeed, the water absorption of P5 is around 2500 wt%/cm², very close to S10 results (Fig. 2), whereas water absorption of P10 and P15 specimens is around 1600 wt%/cm². Moreover, a lower rate of pectin decreases the water handling: there is a loss of cohesion of P5 specimens after around 1.75 h of capillary test, instead of around 4 to 5 h for P10 and P15 specimens. No significant improvement can be observed by adding 15% of pectin instead of 10%. The hydrophobic nature of the *Citrus* pectin, caused by its high methylation degree, is highlighted between P5 and P10 specimens. However, the increase of pectin from 10% to 15% does not counter, which seems to be a minimal water absorption threshold due to the high water absorption capacity of the sunflower pith.

On the other hand, no real influence can be observed with the different chitosan ratios on the water absorption or on the water resistance. All the chitosan specimens kept cohesion after 4 to 5 h testing.

As a first conclusion, the formulation of a pith block formulated with 5% chitosan seems to be interesting in regard with the water absorption and resistance of the material and the low binder content.

3.2 Thermal conductivity

Fig. 5 presents the results of thermal conductivity of the developed materials at dry state and at equilibrium with the environment (25°C and 60% RH) as a function of their density. Commercial values of dry thermal conductivity of some conventional insulating materials in the market (*i.e.*, expanded polystyrene (EPS), glass wool, rockwool and cellulose wadding) were also added as a comparison.

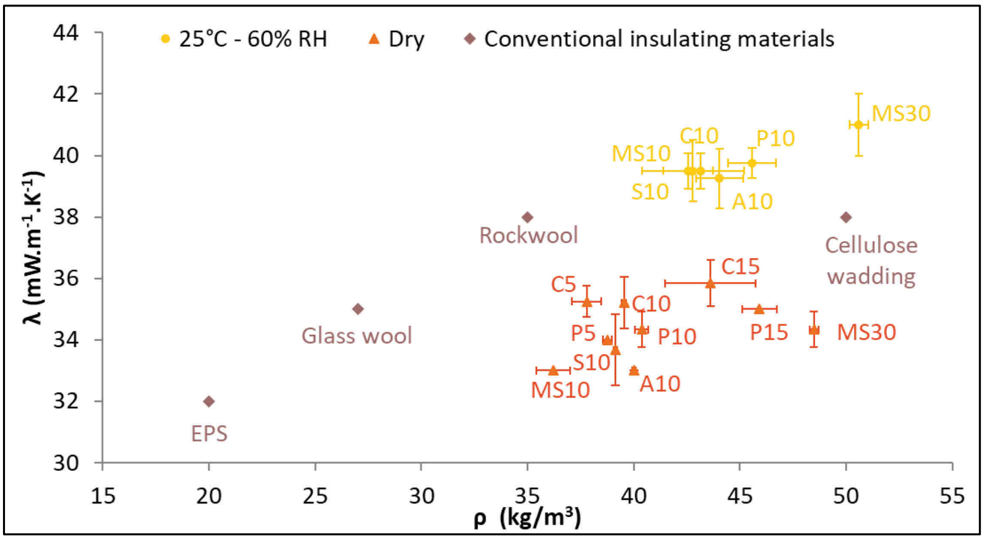

Fig. 5: Thermal conductivity (λ) as a function of the density (ρ) of the studied materials and conventional materials

Different observations can be drawn from Fig. 5. The first and expected result is that the thermal conductivity tends to increase with the material density. Another parameter influencing the thermal conductivity values is the water content of the specimens. Indeed, the values measured on specimens at equilibrium with the environment (25°C and 60% RH) are 16 to 20% higher in average than the ones measured at dry state. This phenomenon is due to the ability of the sunflower pith to adsorb moisture. The pores of the particles contain some water instead of the air, which is less thermal insulating. Thermal conductivity is thus increased.

At similar binder ratio (10%), no real difference between the various binders can be made considering the standard deviations. At equilibrium with the environment, all formulations with 10% binder (S10, A10, C10, P10, MS10) have thermal conductivity between 39.3 and 39.8 mW/m.K. These specimens have moreover very similar density values. The binder ratio, however, has a slight influence on thermal conductivity, which is mainly due to the increase in density. For example, the increase in pectin content at dry state increases the thermal conductivity with 34.0, 34.3 and 35.0 mW/m.K values, respectively, for P5, P10 and P15.

Conductivity values of some conventional materials in the market have been added to the graph as a comparison. The thermal conductivity at ambient conditions of the formulations developed in the present study is very close to rockwool and cellulose wadding (38 mW/m.K), which makes these pith insulating panels competitive in terms of thermal properties. On the opposite, EPS, with a thermal conductivity of around 32 mW/m.K, is the most insulating material. Nonetheless, to obtain the same thermal resistance than an EPS panel of 10 cm thickness, the sunflower pith panel should be 12 cm thickness, which is only 20% thicker.

3.3 Mechanical strength

Fig. 6 brings out the effect of the binder nature on elastic modulus in comparison with the 10% starch formulation used here as the reference insulating block (E/E_{S10}). The elastic modulus is increased by the replacement of starch by alginate, chitosan and pectin, whereas it is strongly decreased in the case of the modified starch, whichever binder rate (10% or 30%). The increase in elastic modulus reaches 17% in the case of C10, and up to 40% in the case of P10.

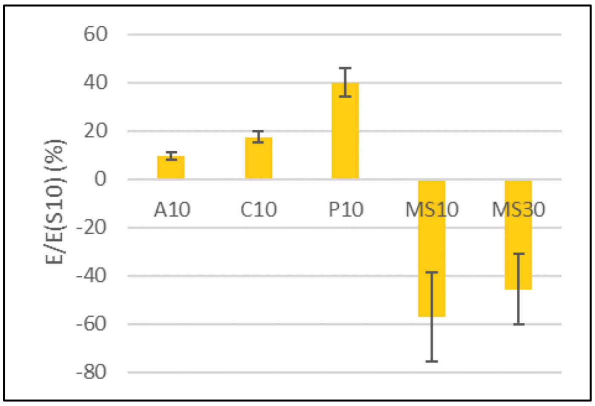

Fig. 6: Effect of the binder nature on the elastic modulus in comparison with 10% starch (S10)

Fig. 7 presents the elastic modulus of the chitosan and pectin formulations, for the three different binder rates tested: 5%, 10% and 15%. A clear increase in the elastic modulus with the increase in binder content is highlighted. The elastic modulus of C15 specimens is increased by 46% compared to C10. In the same way, the elastic modulus of P15 specimens is increased by 22% compared to P10.

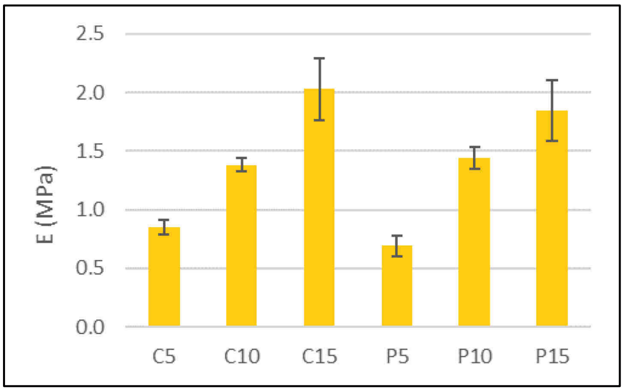

Fig. 7: Elastic modulus of chitosan and pectin formulations

4 Conclusion

The present study focuses on the potential replacement of starch in sunflower pith blocks by other polysaccharide-based binders, namely alginate, chitosan, *Citrus* pectin and a chemically modified starch, in order to improve the water behavior of the insulating material. Water absorption by capillary test, thermal conductivity and mechanical performances were evaluated.

Chitosan and pectin appear to be the most interesting binders considering the three properties studied. Indeed, the formulation with 10% chitosan and the one with 10% pectin reduce the water absorption of the material by 63% and 34%, respectively, compared to the formulation with 10% starch. The water resistance with these two binders, which was assessed considering the time before the specimen lose pith particles during the capillary test, was of the same order of magnitude than the specimens with starch. Moreover, the mechanical performances were also improved with the addition of these two binders. The increase in elastic modulus reaches 40% in the case of pectin and 17% in the case of chitosan instead of starch. The binder nature did not influence the thermal conductivity of the insulating material, which thus stays very good compared to conventional products. Concerning the other binders studied, modified starch decreases mechanical performances and water resistance of the material. For its part, alginate decreases only the water resistance.

The influence of the binder content was also investigated for pectin and chitosan. A rate of 5% might be sufficient in the case of chitosan to reduce the water absorption. However, the mechanical performances decrease slightly. In the case of pectin, a rate of 10% seems necessary to reduce the water absorption in comparison to the starch composite. Contrary to chitosan, pectin is a cheaper agro-industrial co-product. It is also abundant.

In order to further develop these promising insulating panels made from sunflower pith, complementary studies to assess other durability properties, *e.g.*, fire resistance or microorganism resistance, will be necessary in the near future. The study of the evolution of the panel's properties over time (*i.e.*, ageing study) would also deserve to be conducted.

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