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Hygrothermal behavior of Clay – Sunflower (*Helianthus annuus*) and Rape Straw (*Brassica napus*) Plaster Bio-Composites for building insulation

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Abstract.

The aim of this experimental investigation is to study and compare hygrothermal properties of different materials based on vegetable aggregates and clay in order to prove the performance of the developed composite in the framework of BIOCOMP project. Crude earth is known to have very high capacities to regulate indoor humidity. Vegetable aggregate is characterized by high porosity leading also to very effective natural indoor climate regulators. Then, we expect the mix of these two material will to produce an insulation bio-composite with enhanced thermal and hygroscopic performance. Sunflower stem, rape straw and clay used in this investigation are considered as a very low carbon footprint because they are real local agricultural byproducts. Clay-sunflower and rape bio-composites seem to be appropriate and effective biobased insulating plasters, further investigations must be performed in order to characterize more accurately their interesting hygrothermal properties as long as the acoustical and mechanical aspects.

Samples have been manufactured with four different vegetable aggregates (rape straw, sunflower bark, sunflower pith and a mix of sunflower pith and bark) at a same binder/aggregate ratio. The same clay soil is used as a binder for the four bio-composites. The thermal properties for the different bio-composites were obtained from the thermal conductivity measurements after various relative humidity, in order to evaluate the impact of the water content on the thermal parameters. For the hydraulic properties, the sorption-desorption isotherms for the four composites are obtained.

The results show a significant effect of the different aggregates on the hygrothermal behavior of the bio-composite based on clay.

Introduction

The twenty-first session of the Conference of the Parties (COP) that took place in Paris about climate change leads to an agreement [1] between 174 countries. By signing this agreement, all those policy makers were aware about the emergency to reduce drastically the emission of CO₂. The building sector (both for the construction and operation) widely contributes in the total emission of CO₂ on the planet, due to the energy consumption (heat, air ventilation and cooling) and materials production (mostly Portland cement used in concrete, and mineral and petroleum derivative to produce insulating materials). Furthermore, natural resources depletion becomes a critical issue, the use of renewable and recyclable resources, with low embodied energy is therefore relevant. The use of agricultural byproducts as lightweight aggregates for thermal insulation purpose is particularly judicious. They have low embodied energy and can be considered as wastes with a positive carbon assessment as they stock CO₂. Replacing Portland cement by local crude earth enhance positively the material carbon footprint. Its lower mechanical properties are balanced with its vapor regulation capacities when used as insulating, non-structural material. The mix of crude earth with lightweight's vegetable aggregates enhance the material's insulating and hygrothermal properties preserving its low carbon footprint. Those materials can additionally be very effective both thermally and for their vapor regulation properties.

Usually, when we talk about vegetable concretes, we refer to lime-hemp concretes [2], here the aggregates (sunflower stem, rape-straw) are other byproducts of largely cultivated crops in France [3]. Some studies have been made with lime-rape straw composites [4] showing interesting thermal and hygrothermal properties same as for sunflower stem with pozzolanic binders [5,6]. On the other side lightweight crude-earth [7] materials start to be studied. For the first time, we investigate here the thermal properties of mixes of crude earth with rape-straw and sunflower stem (bark and pith). To be considered as building thermal insulator, a material should have a thermal conductivity lower than 0,065W.m⁻¹.K⁻¹. Lime/vegetable or clay/vegetable plasters can hardly reach those performances, but one of their major interest is their capacities to regulate indoor climate as well as being compatible with vernacular buildings made with crude earth or limestone.

In this paper, we present some results about different mixes of clay/vegetable aggregates (rape straw, sunflower stem and sunflower bark) at the same binder/aggregates (w) ratio with and different densities. We measured the thermal conductivity at four relative humidity and so, were able to link the thermal conductivity with relative humidity, water content, types of aggregates and densities. Additionally, we determined the sorption curves at a given fresh density for all four clay-vegetable aggregates mixes.

Materials and method

Aggregates. The vegetable aggregates used in this investigation are rape-straw and sunflower stem. This stem is composed by a rigid external part (bark) and a lightweight inner part with honeycomb structure (pith). We were able to separate two different aggregates: pure bark and a mix of bark and pith with the same density as the used rape-straw. Those vegetables have been grinded with a Retsch SM100 knife mill using a 15mm grid. We had to remove the dust off the bark aggregates using a 2mm sieve.

Binder.

The crude earth is excavated from a local quarry then crushed and passed into a 2mm sieve into the same site to obtain the following grain size: 35% clay (kaolinite, montmorillonite, nontronite), 40% silt and 25% sand.

Formulation and manufacturing. Prior to this experiment, we realized clay:vegetable aggregates mixes using volume ratios. These first results revealed different thermal behaviors for a similar density regarding the used aggregates and the compactness. Following those primarily results, we decided to investigate the hydro-thermal properties of those mixes at the same 2.4:1 binder:aggregates weight ratio despite the significant differences in aggregates' densities. The differences in aggregates' densities imply that a same used weight ratio of 2.4:1 correspond to 7 volumes of rape-straw or bark-sunflower aggregates and only 4.7 volumes of sunflower bark for 1 volume of clay. Different fresh mortar densities obtained by modifying the compaction strength have also been studied.

In addition to the measurement of the thermal conductivity at different densities, we measured for a same ρ_1 fresh density, the thermal conductivity at different humidity rates, so we did for a second density ρ_2 for the clay/sunflower bark sample. For those same mixtures, we investigated their hydric behaviors by drawing the sorption/desorption curves.

Materials have been weighted with a Denver Instrument SI-4002 (0.01g precision scale). Crude earth and water have then been first mixed together at a 1:1 ratio. We waited 10 min before adding the aggregates in order to let the clay being sufficiently hydrated. We then mixed the small amounts of lightweight earth concretes until we reach sufficient homogeneity. Two molds have been used, one 5x5x4cm for sorption/desorption purpose see Fig 1 and another 11x5x4cm for thermal conductivity measurements see Fig 2. In order to have the same fresh density of the materials, we weighted scrupulously the same quantity for each mix for a given density. By modifying the compaction strength we were able to reach different fresh densities, the samples have been compacted by hand. The samples have then been demolded the same day of the elaboration and let air-dried in a 23°C 50%HR regulated room.

Experimental methods.

Thermal conductivity. In order to characterize the thermal conductivity at different moisture content, the samples have been exposed to different humidity atmospheres into desiccators using different salt-solutions. The following humidity rates: 66%, 86% and 98% have been used. In addition we measured the thermal conductivity at dry state (the samples have been oven dried at 60°C for 24h). The measurement have been realized by using the hot-wire technic following the ASTM D5930-97 standard by using a NEOTIM FP2C conductivity meter equipped with hot wire sensor placed between 2 samples of similar geometry.

Sorption/desorption isotherm. To control the relative humidity of the environment, the samples were stored into desiccators containing different mineral salts. Relative humidity rates used for this study are 12, 33, 44, 55, 66, 76, 86, 93 and 98 %. In order to determine the sorption curve, the different samples were first oven-dried (60°C for 24h) before being placed in the corresponding desiccators. In parallel, the other half of the samples were water-saturated by pouring water on them using a wash bottle (this method have been employed because the samples tends to disintegrate when immersed in water) and stored in other desiccators at the same relative humidity.



Fig 1. Samples for sorption/desorption measurements, from left to right: clay-sunflower pith, clay-sunflower bark, clay-sunflower bark and pith and clay rape-straw



Fig 1. Samples for thermal conductivity measurements, from left to right: clay-sunflower pith, clay-sunflower bark, clay-sunflower bark and pith and clay rape-straw

Results and discussions.

Effect of the aggregate types on thermal conductivity. Aggregates of two different densities have been used here: on the one hand, rape straw and sunflower bark-pith at $80,5\text{g}\cdot\text{dm}^{-3}$ (we adjusted the bark-pith quantities putting more bark in order to obtain the same volume as rape-straw for the same weight) and on the other hand, sunflower bark at $126,4\text{g}\cdot\text{dm}^{-3}$. The sunflower bark is significantly denser than the other aggregates that implies that for a given volume, we will have more binder for the clay/sunflower bark sample than for clay/rape-straw or clay/sunflower bark and pith. As the binder has a much higher thermal conductivity than the vegetable aggregates, we logically measured higher thermal conductivity values for the clay/sunflower bark samples than for clay/rape-straw or clay/sunflower bark and pith ones for a given density.

Effect of dry density on thermal conductivity. For the same binder:aggregates ratio, we were able to obtain various densities by modifying the compaction strength. The corresponding dry thermal conductivities have then been measured and compared.

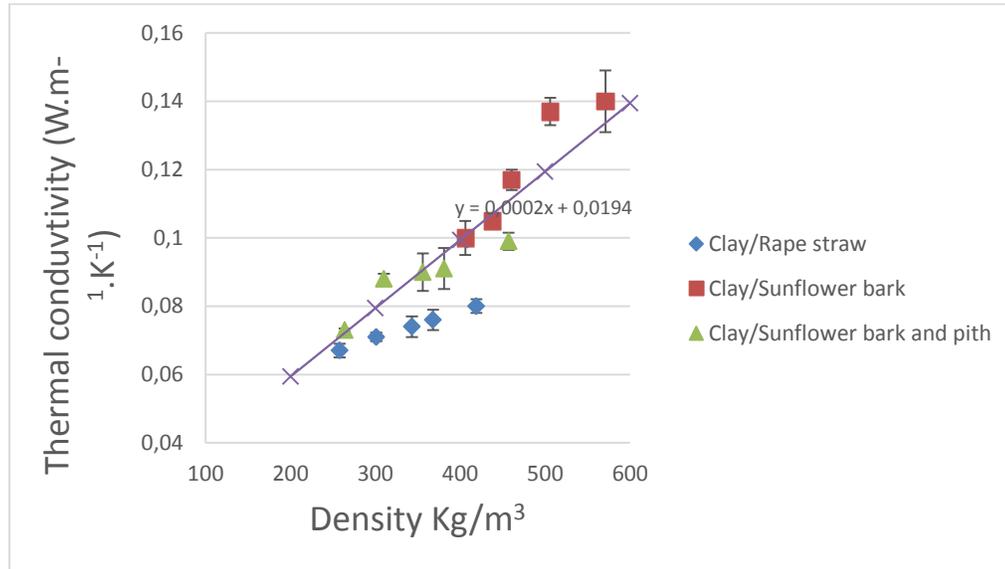


Fig 3. Thermal conductivity versus density of several clay/vegetable aggregates

As expected, the thermal conductivity increased with density. For densities ranging from 258 to 571 kg m^{-3} , the thermal conductivity increase from 0,067 to 0,140 $\text{W.m}^{-1}.\text{K}^{-1}$. The different mixes show different behaviors in their thermal conductivity when their density increases as seen on Fig. 3. If we plot the empirical relationship established by Cerezo [8] for hemp concretes (Eq. 1) on the same graphic, we notice wider deviation than for straw-clay mixes and other earth-based materials [9].

$$\lambda = 0.0002 \cdot \rho + 0.0194. \quad (1)$$

Clay/rape straw mixes thermal conductivities are less affected by their compaction than the ones for clay/sunflower bark mixes for similar binder/aggregates weight ratios. As explained above, clay/sunflower bark mixes contains more volume ratio of binder, with higher compaction, the inner binder porosities are reduced first in respect to the intra aggregate porosities which affect drastically the conductivity of the material. At the opposite, clay/rape straw and clay/sunflower bark and pith shows a higher content of aggregates so that the diminution of the inner binder porosities will less affect the thermal conductivity of the sample.

Effect of moisture on thermal conductivity. For this experiment, five set of samples of the same mix have been put at different moisture contents: oven-dry, 66%, 86%, 98% and water-saturated. The samples have been weighted until stability is reached, which could take several weeks. Once no more weight variation is detected, the thermal conductivity is measured and plotted against relative humidity or specimen's water content (Fig 4 and 5).

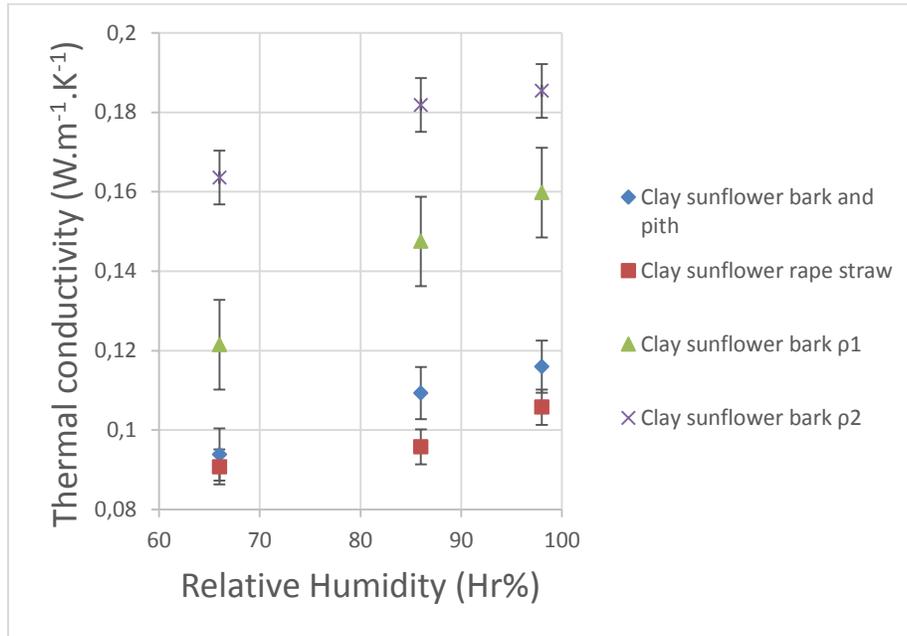


Fig 4. Influence of relative humidity on thermal conductivity for different clay/aggregates' samples

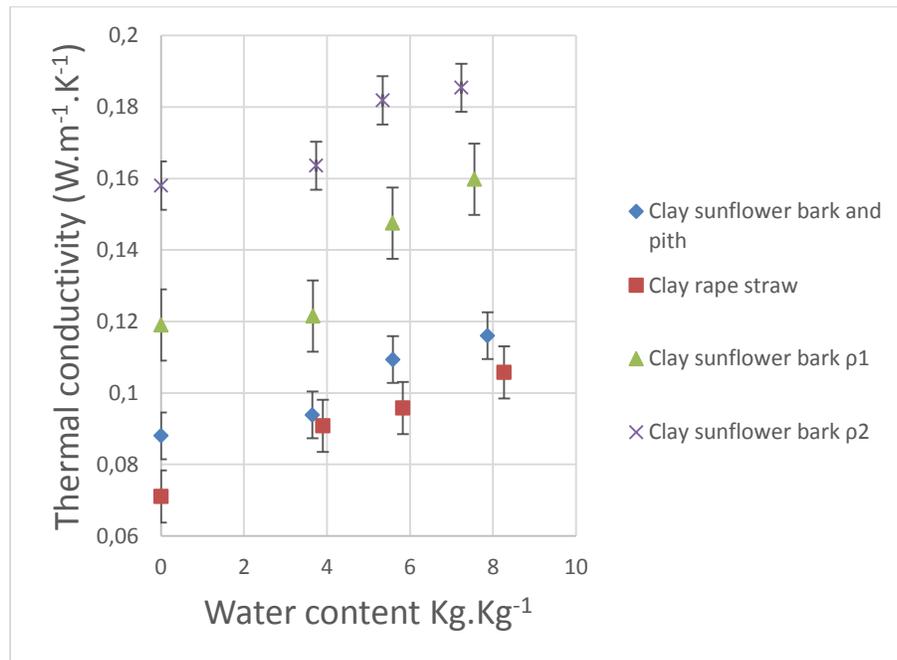


Fig 5. Influence of water content on thermal conductivity for different clay/aggregates' samples

The clay/rape straw mix shows the highest increase of thermal conductivity (48.5%) between dry state and 98%RH, the clay/sunflower bark at ρ_1 shows a 34,5% increase, the clay/sunflower bark and pith shows a 32% increase and the clay/sunflower bark at ρ_2 shows a 17.5% increase. This increase is correlated with the water content of this different samples as shown on Fig 5. As explained by El Fgaier et al [10] an increase of the water content leads to higher amount of water in the sample's porosities, replacing air. As a consequence, as water shows a higher thermal conductivity than air, the thermal conductivity of the sample is increasing. Furthermore at certain relative humidity rates, the water vapor contained in pores induces menisci generating thermal bridges [10].

Sorption/desorption hysteresis.

Clay/rape-straw and clay/sunflower bark and pith mixes show similar sorption-desorption behavior, their sorption curve is also similar to that of clay/sunflower bark at ρ_1 density. The influence of the vegetable aggregates on the sorption/desorption properties is not clearly significant as shown by Belayachi et al [11] for different straws in straw concretes. Clay/sunflower bark mix at higher density ρ_2 shows slightly different behavior with a higher hysteresis and reach higher water saturation for high relative humidity. This hysteretic behavior is usually explained in concretes by a combination of phenomenon: capillary condensation hysteresis, contact angle hysteresis and ink-bottle effect [12]. El Fgaier et al study [10] showed that mass moisture content of unfired clay bricks could reached 3.5% at 95%HR. Here, from dry state to 98%HR the mass moisture content reached 17.2% for clay/rape straw sample, 16.9% for clay/sunflower bark and pith, 15.5% for clay/sunflower bark at ρ_1 and 14.6% for clay/sunflower bark at ρ_2 .

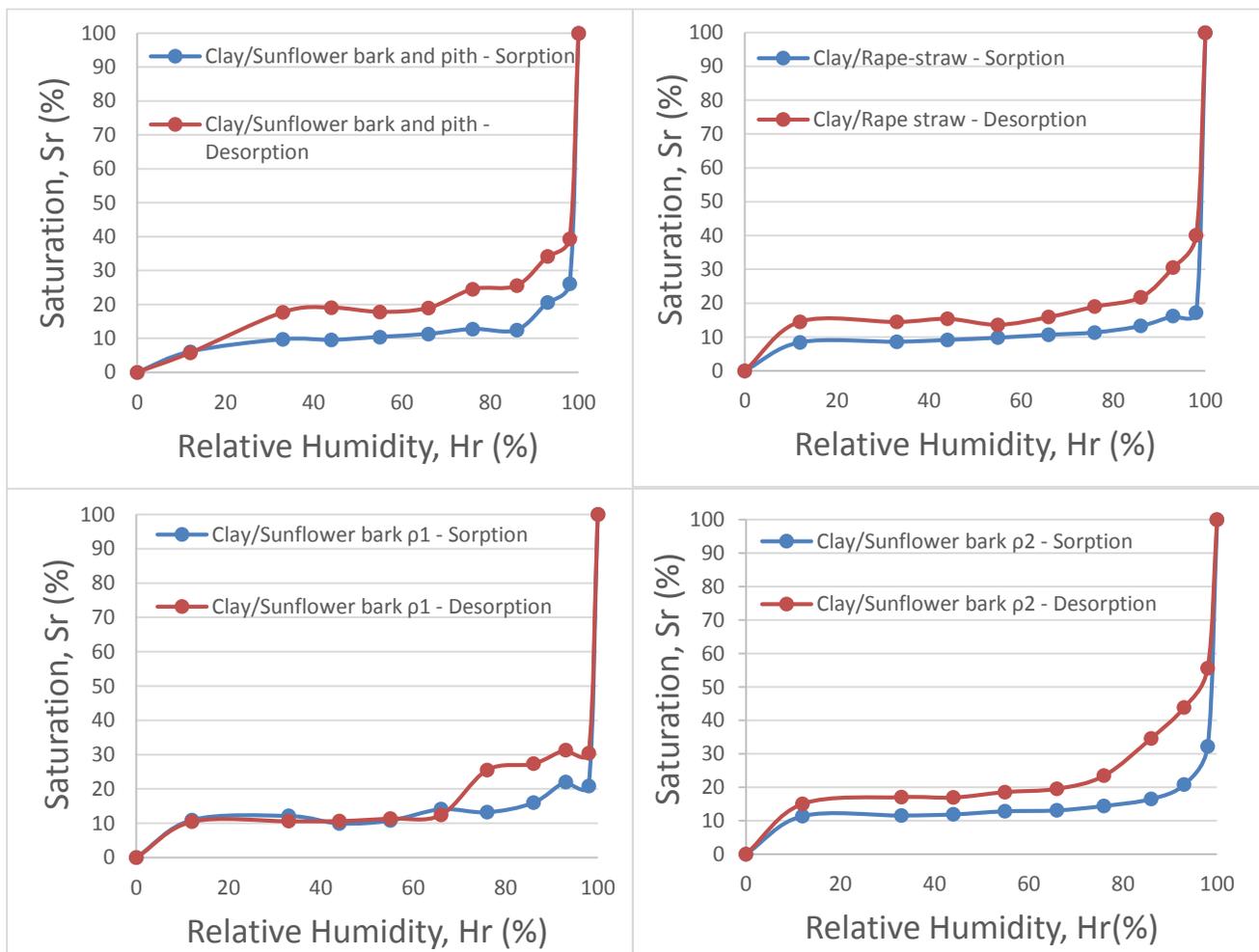


Fig 6. Isothermal sorption-desorption hysteresis of four different mixes of clay/vegetable aggregates' mixes.

Conclusion.

The results presented herein show that the thermal conductivity of lightweight crude earth depends both on its density, its water content and the type of aggregates. The thermal conductivity of those clay/vegetables aggregates plasters is more affected by the water content (increase up to more than 48%) than for lime hemp concretes (less than 20% increase) which is in accordance with previous studies

concerning earthen materials [5,7]. In fact, hemp concretes tend to be less affected by water content than density [13] contrary to the results we obtained for clay:vegetable aggregates.

The significantly higher amounts of water mass content obtained for clay:vegetable aggregates mixes than with unfired clay bricks while increasing the relative humidity lead us to assess that the addition of porous vegetable aggregates to crude earth substantially improve the capacities of raw earth to absorb water vapor.

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