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 Auto-coherent homogenization applied to the assessment of thermal conductivity: case of sugar cane bagasse fibers and moisture content effect

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16 Abstract

17 The purpose of this study is to evaluate the thermal conductivity of sugar cane bagasse 18 fibers when they reinforce vegetable fibers/cement composites using auto-coherent 19 homogenization. The moisture content effect on the thermal conductivity of composites 20 is also studied. When the fiber content increases, porosity increases according to a linear rule and bulk density and thermal conductivity of composites decrease. When the 21 22 moisture content grows, the thermal conductivity of composites increases. When 23 applying auto-coherent homogenization, in dry state, results show a gap of less than 24 10% between experimental and modeled values of composites thermal conductivity and a mean sugar cane bagasse fibers thermal conductivity of 0.110 W/m.K. In wet state 25 26 (56% RH), there is an approximate agreement of 1,4% between estimated and 27 experimental values of thermal conductivity of composites up to 4 wt% of fibers 28 content.

29

Keywords : Natural fibers ; Thermal properties ; Cement composites ; Auto - coherent
 homogenization; Moisture content.

- 33 1. Introduction
- 34

Guadeloupe (French West Indies) is a Caribbean island with a tropical climate [1, 2] were various vegetable fibers, including agro – industrial wastes, are low-cost and/or abundantly available [3-4]. This archipelago has a high seismicity and is prone to violent hurricanes [5]. Therefore, the main construction material used in the building sector is conventional cement and its derivatives (concrete blocks, bricks, ...) [6-7].

40 French thermal and acoustic regulations advocate the use of 50% renewable power in 41 energy consumption by 2020 [7]. One way to reduce the energy consumption, as 42 consequence to increase the renewable energy ratio, is to use less air-conditioning in 43 infrastructures [7]. To achieve this goal, the partial replacement of cement by vegetable fibers as reinforcement of cementitious matrix is considered [1, 3, 6, 7]. Indeed, 44 45 vegetable fibers are known to have insulating properties that is to say a low thermal 46 conductivity and appear to be a good alternative to synthetic fibers such as glass fibers or asbestos fibers, commonly used for building insulation [8-10]. As observed by 47 48 Onésippe et al. in 2010 [3] and even now, few works have demonstrated the low thermal

49 conductivity of vegetable fibers cement-based composites as well as the thermal

- 50 conductivity of vegetable fibers (when they are incorporated in the composites) and the
- 51 influence of moisture on thermal conductivity of these composites. The presence of 52 wood fibers reduces the density of the material [11] and decreases its thermal 53 conductivity [3, 6, 10].
- 54 Vegetable fibers are vulnerable to alkalinity of matrix thus causing debonding of the
- 55 fiber / matrix interface and thus a decrease in the mechanical performance of the
- 56 composites [3, 9]. To limit vulnerability of vegetable fibers in the matrix, various
- 57 treatments of fibers have been considered such as chemical (acid or alkaline), physical-
- 58 chemical, thermal and mechanical treatments [9, 11]. These various treatments lead to a 59 strengthening of fiber/matrix interface [12] because they induce, on the one hand, the
- 60 modification of the morphology of the fibers and, on the other hand, the reduction of the
- 61 contents of hemicellulose and extractible materials which are inhibitors of the hydration
- 62 of the binder [12].
- 63 Moreover, vegetable fibers are hydrophilic and their high affinity for water may thwart
- 64 the hydration of matrix [13]. To overcome this competition, some authors propose the
- 65 pyrolysis [12, 14] or the pre-wetting [15-18] of vegetable particles to retain their
- 66 original volume (porosity) and so, play a role of water tank during the setting phase of
- 67 the binder. Collet [13] recommends a pre-wetting of the binder with water prior to its
- 68 addition to vegetable particles in order to ensure a good hydration of binder and to
- 69 avoid competition between cement and fibers hydration [19].
- 70

The purposes of this study are to (1) estimate the modeled value of thermal conductivity of vegetable fibers when they reinforce cement matrix and (2) measure the effect of

- 72 of vegetable fibers when they remote cement matrix and (2) measure the effect of 73 moisture content on vegetable fibers/cement composites thermal properties. To reach
- 75 Indisture content on vegetable inters/cement composites thermal properties. To reach
- these goals, cement composites reinforced by various amounts of sugar cane bagasse
  fibers ranging from 2 to 8 wt % were considered. Sugar cane bagasse fibers are widely
  available in Guadeloupe : according to Food Directorate of Agriculture and Forestry
- available in Guadeloupe : according to Food Directorate of Agriculture and Forestry
  from Guadeloupe, the island produces 680000 tons/year of sugar cane mainly used for
  the sugar and rum industries [20]. Bagasse is the solid lignocellulosic leftover after
  extraction of juice from the sugar cane stalk and is cheap compared to synthetic fibers
- 80 [21].
- 81 In order to achieve the modeling of thermal conductivity of fibers, the first part is 82 dedicated to the study of bagasse fibers / cement composites : measurements of thermal 83 conductivity and physical-chemical characteristics (density, apparent volume and 84 porosity). The thermal conductivity is studied at three different moisture contents. In 85 the second part, a model of thermal conductivity of fibers and composites, in dry and 86 wet states, is applied. The purposes of this work is (1) to propose a way to assess the 87 thermal conductivity of bagasse fibers incorporated in this kind of composites, as very few data's of thermal conductivity of vegetable fibers are available and (2) to compare 88 89 the (experimental and numerical) behaviors of composites when they are placed in 90 different relative humidity (RH) to establish, if possible, a relation between thermal 91 conductivity of composites and RH. Generally, in the literature, many models make it 92 possible to estimate the thermal conductivity of dry concrete and dry composite 93 materials on the basis of knowing the conductivity of each component and its 94 concentration [22-27]. Several of them are based on the auto-coherent homogenization (HAC) model [25-28]. This model uses the statements of self-consistent field concept 95 96 and of spherical geometry of inclusions in composites; for examples:
- 97 autoclaved aerated concrete for Boutin [29],

- 98 synthetic foams in continuous medium for Felske [30],
- 99 wood shaving and concrete for Bederina et al. [25],
- 100 cement composites containing rubber waste particles by Benazzouk et al.
  101 [26],
- 102 hemp concretes by Collet and Pretot [28],
- 103 insulating building materials made from date palm fibers mesh by
  104 Boukhattem et al. [26].

105 Among them, Felske [30], established equations allowing the estimation of the thermal 106 conductivity of regular hollow spheres particles (synthetic foam), while the other 107 previous authors [25-29] focused on a distribution of spherical particles with different 108 diameter. Although the geometries involved are the same as in our materials, it appears 109 that Felske's special case and critical values are not suitable in this study because Felske 110 assumed that the particles are uniform. Bagasse fibers, being vegetable matter, are not 111 uniform [31] and more, there is no perfect thermal contact between the vegetable 112 matter and the cementitious matrix. Finally, the auto-coherent homogenization 113 equations, as described by [25-29], were chosen because they are well suited to 114 materials with very different pores sizes (such as concrete) [29] and they are currently 115 employed when studying thermal conductivity of concrete.

- 116 Thus, in this work on vegetable fibers/cement composite materials,
- a focus has been made on the model already confirmed and largely used in the
   field of cementitious materials containing vegetable matter that is to say the
   HAC model,
- this HAC model as described by [22-29] has been applied to assess the numerical value of thermal conductivity of bagasse fibers/cement composites in wet and dry states and it was possible for us to deduce the thermal conductivity of sugar cane bagasse fibers in the cement composites.
- To our knowledge, very few models of thermal conductivity of vegetable fibers / cement composites and even less models of the thermal conductivity of vegetable fibers alone are proposed in the literature [3, 8].

127

- 128 2. Materials
- 129 2.1 Sugar cane bagasse fibers
- 130 Sugar cane bagasse fibers are named NBF and were collected from Montebello distillery
- 131 (Petit-Bourg, Guadeloupe, FWI). Fibers were crushed with a knife mill (Restch, France)
- and sieved in the laboratory to obtain final length varying from 1 to 10 mm and width
- 133 between 0.4 and 1 mm. The sugar cane bagasse fibers chemical composition has already
- 134 been determined [32] and is summarized in Table 1.
- 135 136 2.2 Binder
- 137 The binder was a white Portland cement CEM I 52.5 N manufactured by Axton society.
- 138 This Portland cement CEM I 52.5N complies with European standard EN 197-1 [33]. Its
- 139 chemical composition and some of its physical characteristics are presented in Table 2.
- 140
- 141 2.3 Composites formulations and preparation

- A reference sample (i.e. without any fibers) has been prepared. It is named L.
  Composites were made by mixing each type of binder with various amounts of sugar
  cane bagasse fibers. Formulations of composites, named LFN, are presented in Table 3.
- All composites were prepared according to the mixing sequence indicated in Table 4 and tap water was used.
- 147 The obtained pastes were casted in normalized molds of dimensions 40\*40\*160 mm<sup>3</sup>.
- According to EN 196-1 standard [34], a compaction (10 strokes by step 2 steps) with a
- shaking table was applied to each sample. Then the isotropic samples, with a random
- 150 repartition of fibers in the binder, were removed from the mold and left for curing
- 151 during 28 days in a climatic chamber (25°C, RH = 70%).
- 152
- 153 3. Methods
- 154
- 155 3.1 Bulk dry density, apparent volume and porosity measurements of composites
- 156 The bulk dry density  $\rho_{\text{A}}$  was deduced from the dry mass and apparent volume of
- 157 samples. To determine the dry mass, samples were placed in an oven at 105°C (+/- 1°C).
- 158 The dry state was reached once the mass of each sample was stabilized.
- The apparent volume of each sample was measured by using a sliding caliper (accuracy
  of +/- 0.01 mm). Each mean dimension is calculated from an average of three
  measurements.
- 162 Composites true density  $\rho_V$  has been measured using helium gas intrusion under helium
- 163 gas flow with a "Pycnomatic" Thermo Electron Corporation equipment (France) 164 pycnometer. Five measurements were conducted for each composite at 298 K, relative
- pycnometer. Five measurements were conducted for each composite at 298 K, relative
   humidity of 70–80%. The open porosity n is deduced from bulk dry and true densities
- 166 using equation (1):
- 167  $\eta = 1 \frac{\rho_A}{\rho_V}$  equation 1
- 168 where  $\rho_A$  is bulk dry density,  $\rho_V$  is true density of samples and  $\eta$  is open porosity.
- 169

170 3.2 Thermal conductivity testing

- 171 The tests were performed with a C-therm TCi unit (Setaram, France). The C-Therm TCi 172 employs the Modified Transient Plane Source (MTPS) technique. The one-sided, 173 interfacial heat reflectance sensor applies a momentary constant heat source to the 174 sample. A known current is applied to the sensor's spiral heating element, providing a 175 small amount of heat. The sensor's guard ring is fired simultaneously supporting a one-176 dimensional heat exchange between the primary sensor coil and the sample. The 177 current applied to the coil results in a rise in temperature at the interface between the 178 sensor and sample, which induces a change in the voltage drop of the sensor element. 179 The increase in temperature is monitored with the sensor's voltage and is used to 180 determine the thermo-physical properties of the sample. The thermal conductivity is inversely proportional to the rate of increase in the sensor voltage (or temperature 181 182 increase). Thermal conductivity is measured directly [35].
- For precise measurement of thermal conductivity, samples are first polished using a rotate polishing machine with sandpaper (Dremel, USA). Three levels of moisture are evaluated: 0%, 56% and saturated state 100% of relative humidity. We choose 56% RH as an intermediate value in relative humidity because, in real conditions, cement composites do not reach dry-oven or saturated conditions. The dry state (0% RH) is obtained by drying the material in an oven at 105°C (+/- 1°C) and then placing it in a

- desiccator until the time of testing. To measure the thermal conductivity at 56 % RH,samples are placed in a desiccator whose humidity is controlled using saturated saline
- 191 solutions of sodium bicarbonate. Local measurements are made on the lateral and
- 192 transverse sides of samples and an average value is calculated. To ensure that the
- 193 moisture content has not changed, samples are weighed before and after measurement.
- 194
- 195 3.3 Modeling of thermal conductivity of composites by auto-coherent homogenization
- 196 The auto-coherent homogenization is used in order to model the thermal conductivity of 197 bagasse fibers / cement composites. This method was initially developed for the 198 mechanical characterization of composites materials [25-26] and was then extended to 199 electrostatic, magnetostatic, electric conduction and thermal properties, which are 200 mathematically analogous [18, 28]. The principle of the method is that the heterogeneous material is assimilated to an equivalent homogeneous material, which 201 202 must be characterized (knowledge of the conductivity of each component and its 203 concentration). Thus, a transition of micro-scale (components) to macroscopic scale 204 (material) allows to express overall thermal conductivity in terms of characteristics of 205 each component (conductivity, volume concentration).
- 206
- 207 3.3.1 Dry state

The auto-coherent homogenization is well described by Collet and Pretot [28]. Briefly, the dried material is considered as an assembly of spherical inclusions of various sizes. In the case of material with two-components, it is meant to be a sphere of radius  $R_a$  and thermal conductivity  $\lambda_a$  (component "a" is the air contained within the vegetable fibers) surrounded by a concentric shell of component "s" (s is the fiber block) of thermal conductivity  $\lambda_s$  and radius  $R_s$ .  $\lambda_d$  is the thermal conductivity of the equivalent homogeneous material also called effective conductivity (Figure 1).

The expression of effective conductivity  $\lambda_d$  (equation 2) is obtained by assuming that the energy contained in the heterogeneous medium is equivalent to that of the homogeneous medium under the same boundary conditions [26, 28].

218

219 
$$\lambda_{d} = \lambda_{s} = \left[1 + \frac{\varepsilon}{\frac{1-\varepsilon}{3} + \frac{1}{\lambda_{a}/\lambda_{s} - 1}}\right]; \quad \varepsilon = \left(\frac{R_{a}}{R_{s}}\right)^{3}$$
 equation 2

220

where  $\epsilon$  is the volume concentration of air phase: it is assumed that the concentration of

unconnected phase is equal to the ratio of external and internal pores [29].

According to Boutin [29], this assumption is only satisfied if the material consists of an assembly of composites spheres of variable sizes. Collet and Pretot [28] made this same assumption in case of hemp concrete composites, which are close to bagasse cementitious composites of this study.

226

We assume that the fibers / dry binder composite is a three components material. In that case, the tri-composite inclusion method is used for the modeling of thermal conductivity assuming that a spherical air bubble "a" is surrounded by a concentric vegetable particles shell "f" itself surrounded by a binder shell "s" (Figure 2).

- 231 This type of wildcard inclusion is based on three assumptions:
- the binder consists of cement and microscopic air bubbles trapped in closed-pores;
- 234 the vegetable particles consist of plant part and intra-particle air;
- 235 the air bubble is the microscopic and macroscopic air contained in open pores of236 material.
- The expression of thermal conductivity of vegetable fibers / cement composites istherefore given by equation 3.

$$\lambda = \lambda_{s} \left[ 1 + \frac{\theta}{\frac{1 - \theta}{3} + \frac{1 + \frac{\delta}{3} \left( \frac{\lambda_{a}}{\lambda_{f}} - 1 \right)}{\frac{\lambda_{a}}{\lambda_{s}} - 1 - \frac{\delta}{3} \left( \frac{\lambda_{a}}{\lambda_{f}} - 1 \right) \left( \frac{2 \lambda_{f}}{\lambda_{s}} + 1 \right)} \right]$$

240

241 where 
$$\theta = 1 - \frac{1}{k+1} \left( \frac{\rho}{\rho_s} \right)$$
,  $\delta = \frac{\rho}{\rho_f} \frac{k}{k+1} \frac{1}{1 - \frac{\rho}{\rho_s} \frac{1}{k+1}}$  and  $k = m_f / m_s$ 

242  $\theta$  and  $\delta$  are concentrations directly calculated from the mass m of each component (of 243 known density  $\rho$ ). This definition of k is based on the assumption that the change of 244 properties between powdered cement and dry hydrated cement (i.e. hydrated cement at 245 0%RH) does not cause a significant variation in the thermal conductivity of composite 246 [15]. For each formulation, the characteristic parameter k is calculated as the ratio 247 between the mass of bagasse fibers and the mass of powdered cement [25].

equation 3

248 249 3.3.2 Wet state

250 The modeling process of wet composite involves two steps:

- the first step allows creating homogeneous medium "sf" of vegetable particles "f" and hydrated binder "s" at 0% RH,
- in the second step, the homogeneous medium "sf" is included in a tri-composite
  (air, water, "sf") model to obtain the final homogenized wet composite. The
  vegetable fibers "f" and dry binder "s" are therefore considered as a
  homogeneous medium "sf" and not as two separated phases (Figure 3).

257 Equation 4 gives the expression of equivalent conductivity of "sf" composite as a 258 function of conductivity of each component (fibers and hydrated binder at 0% RH). The 259 parameter  $\varepsilon$  is used to evaluate the volume concentration of fibers in the binder.

260 
$$\lambda_{sf} = \lambda_s \left[ 1 + \frac{\varepsilon'}{\frac{1-\varepsilon'}{3} + \frac{1}{\lambda_f/\lambda_s - 1}} \right] ; \quad \varepsilon' = \left( R_f/R_s \right)^3 = \frac{1}{1+k\frac{\rho_f}{\rho_s}}$$
 equation 4

where  $\lambda$  is for the thermal conductivity, k is the ratio of mass (as defined in equation 3) and  $\rho$  is the density.

In the second step, the tri-composite inclusions model allows expressing the conductivity of composite in wet states (equation 5).

265 
$$\lambda_{H} = \lambda_{sf} \left[ 1 + \frac{\theta}{1 + \frac{\delta_{sf}}{3}} + \frac{1 + \frac{\delta}{3} \left( \frac{\lambda_{a}}{\lambda_{w}} - 1 \right)}{\frac{\lambda_{a}}{\lambda_{w}} - 1 - \frac{\delta}{3} \left( \frac{\lambda_{a}}{\lambda_{w}} - 1 \right) \left( 2 \frac{\lambda_{w}}{\lambda_{sf}} + 1 \right)} \right]$$
equation 5  
266 
$$k_{1} = -\frac{m_{w}}{3}, \quad \theta = \left( \frac{R_{w}}{2} \right)^{3} = 1 - \frac{1}{1 + \frac{1}{2}} \frac{\rho_{h}}{\rho_{h}} \text{ and } \delta = k_{1} \frac{\rho_{sf}}{\rho_{sf}} \left[ \frac{1}{1 + \frac{1}{2}} - 1 \right]$$

266 
$$k_1 = \frac{w}{m_s + m_f}, \quad \theta = \left(\frac{w}{R_{sf}}\right) = 1 - \frac{1}{1 + k_1} \frac{\gamma_n}{\rho_{sf}} \text{ and } \delta = k_1 \frac{\gamma_{sf}}{\rho_w} \left(\frac{1}{1 - \frac{\rho_H}{\rho_{sf}} \frac{1}{k_1 + 1}} - 1\right)$$

 $\lambda$  is for the thermal conductivity, k is the ratio of mass of water (w) related mass of cement (s) + mass of fibers (f).

269  $\theta$  and  $\delta$  are concentrations directly calculated from the mass m of each component (of 270 known density  $\rho$ )

271

272 4. Results and discussion

In order to estimate thermal conductivity of vegetable fibers, some properties of composites are required: physical properties (bulk dry density, open porosity) and thermal conductivity.

276

4.1 Bulk density and open porosity of composites at 0% RH

Figure 4 reports the variation of bulk density as a function of the fiber contents for LFN composites in the dry state. As expected, the more the fiber content, the lower the bulk density is [36]. The first significant decrease is obvious: it corresponds to the addition of 2% by weight of bagasse fibers, which are a lightweight material. Between 2 and 4% by weight of fibers, there is a light decrease. After 4 wt%, bulk density decreased at a relatively constant rate with increased fiber content as observed by [37].

Figure 5 presents the evolution of LFN composites porosity according to their fiber content in the dry state.

Porosity of material is gradually increasing with the amount of bagasse fibers. This increase with addition of fibers is explained by the formation of air voids in the

288 microstructure of paste (due to the presence of fibers which are porous) and the voids

- content becomes high as fiber volume fraction increases [36]. Moreover, short fibers, as
- used in this study, are considered to be more difficult to align and pack densely. The

- 291 packing of short fibers in cement paste leads to increase the amount of voids [10] and
- therefore porosity.
- 293
- 294 4.2 Thermal conductivity of composites
- 295 4.2.1 Experimental thermal conductivity
- 296 4.2.1.1 Dry state

Figure 6 illustrates the evolution of thermal conductivity of LFN composites according to fiber content at 0% of relative humidity. According to Demirboga [38], thermal conductivity of Portland cement type I is (1.230 +/- 0.050) W/m.K.

300 The purpose of including vegetable fibers in cement is to develop a more insulating 301 material than cement in order to use it as an interior partition or an interior insulating 302 coating in the housing, for examples. It would help to prohibit the leaking of heat on both sides of the partitions [36]. First, as expected, the thermal conductivity of Portland 303 304 cement is strongly decreased (by a factor of around 2) by introducing 2 wt % of 305 vegetable fibers. Then, thermal conductivity of cement samples decreases slightly when 306 increasing fiber content [10, 27, 36]. This decrease follows a logarithmic evolution. 307 According to [3], thermal conductivity of treated bagasse fibers is lower than that of 308 cement paste, so we assume it's the same for raw bagasse fibers: its thermal 309 conductivity is lower than that of cement. Consequently, this decrease is expected with 310 law of mixture. Moreover, thermal conductivity is inversely proportional to the voids in 311 composites [10, 39] and as shown by Figure 5, porosity of samples increases with the 312 fiber content. Based on these results, the linear relation between the thermal 313 conductivity k of bagasse composites and bulk density p is:

314 
$$k = 0.0005\rho - 0.0924(R^2 = 0.78267)$$
 equation 6

- 315 where k is thermal conductivity and  $\rho$  bulk density.
- 316 Equation 6 is consistent with classical equations applied to evaluate thermal
- 317 conductivity of insulating materials used in the field of construction [39].
- 318
- 319 4.2.1.2 Sensitivity of thermal conductivity to moisture content
- Thermal conductivity was also studied for the same composites at 56% and 100% of relative humidity (saturated state).

Fibrous media are known to often have a geometric anisotropy linked (1) to the anisotropy of the fibers themselves and (2) to their orientation within the material. In the case of vegetable fibers, an anisotropy of the thermal properties appears locally, as the tensor of conductivity of fibers is generally orthotropic [38]. We assume that the thermal conductivity of studied composites is isotropic because fibers are randomly

oriented and properties of short fibers (in this study, length varying from 1 to 10 mmand width between 0.4 and 1 mm) composites are isotropic [41].

- Figure 7 presents the thermal conductivity of samples according to their bulk density fordifferent moisture contents.
- 331 The more the percentage of pores, the lighter the specimen are and the lower their
- thermal conductivities are as observed by others [10]. That is to say that the lighter a
- 333 material is and the better is its insulating power. The thermal conductivity of the
- composites in saturated conditions is greater than in the dry state [27]. This fact is explained by the thermal conductivity of water, which is 25 times higher than air [42]:

- 336 the presence of air will alter the overall thermal conductivity of the material by
- decreasing it. The composite subjected to a relative humidity of 56% exhibited a mass
- 338 gain of 4 to 8% (in comparison with dry state), which involves the increase from 15 to
- 50% of the thermal conductivity of materials. For moisture contents close to saturation,
  the thermal conductivity of the composite increases by around 100 % of its value in the
- 341 dry state. These results are in accordance with those exhibited by Asadi et al. [39].
- 342
- 343 4.2.2 Numerical thermal conductivity
- 344 4.2.2.1 Dry state
- Firstly, by knowing the thermal conductivity of pure hardened cement paste,  $\lambda_d$  and its porosity measured with helium pycnometer, we can calculate the conductivity of solid particles  $\lambda_s$  using equation 2. The value of  $\lambda_s$  is 1.125 W/m.K.
- To estimate thermal conductivity of fibers, the auto-coherent method applied to three phases medium (equation 3) is used in combination with least squares method of minimization. The mean square deviation between experimental and numerical thermal conductivities of dry composites is minimized and bagasse fibers thermal conductivity is estimated. Its average value is about 0.110 W/m.K. This value is consistent with the values estimated by Onésippe et al. [3] for treated bagasse fibers.
- Figure 8 shows that the theoretical values calculated using equation 3 are consistent with the experimental measurements in the case of dry composites, whatever the bulk
- density. A gap of less than 10% is obtained.
- 357
- 358 4.2.2.2. Wet state (100% RH)
- 359 As with the previous model, a comparison of the results is made in the case of wet 360 composites (56% RH, Figure 9 and 100% RH, Figure 10). For 56% RH, there is a good agreement between numerical and experimental values in low fibers contents (approx. 361 362 less than 4 wt%), which correspond to bulk densities greater than 1360 kg/m<sup>3</sup>. Below 363 this value, it seems that the model underestimates thermal conductivity of composites 364 by about 10 to 20%. This underestimation reaches nearly 30% at saturated state (100% RH), whatever the bulk density, as shown by Figure 10. The model used for numerical 365 366 calculation considered the shape of the aggregates to be spherical. This underestimation may be due to the geometrical distribution of void phase that is to say distribution of 367 pore structure. This difference can also be explained by conduction phenomenon due to 368 free water or entrapped water that implies the use of a more complex modeling to 369 370 explain the behavior. Moreover, the experimental conductivity is the average of local 371 measurements that depend on surface conditions, orientation of fibers and compacting 372 direction. In addition, polishing of samples with high fiber content is difficult and can 373 cause local differences in thickness.
- The thermal conductivity calculated by the model seems to be little sensitive to changes induced by the generic water cell. This little sensitivity is explained by low volume of water adsorbed in this range of humidity.
- 370 W
- 378 5. Perspectives
- Generally, in Guadeloupe, the relative humidity of the air varies between 70% and 80%
- 380 [42], that is to say that the thermal conductivity of composites would be greater than in
- the dry state. The composites of this study cannot be considered as insulating materials
- 382 for our climatic conditions. In order to decrease their thermal conductivity, we

383 considered treatment of bagasse fibers before incorporating them into Portland cement. 384 We choose to analyze the effect of pyrolysis treatment on sorption/desorption behaviors of the bagasse fibers. The sorption isotherm of natural bagasse fibers NBF is 385 386 compared to that of pyrolyzed fibers, TBF which are prepared under controlled inert 387 atmosphere (N<sub>2</sub> flow, 2 L/h) during 2 h at 240°C [43]. It is obtained experimentally by assessing the moisture content of the product in equilibrium with different air relative 388 389 humidity at an average temperature of (22±3) °C. Relative humidity is controlled by 390 saturated saline solutions according to ISO-12571 norm [44]. The moisture content *w* is 391 deduced using the equation 7:

$$392 \qquad w = \frac{m - m_0}{m_0} \qquad \text{equation 7}$$

Where m and  $m_0$  are the mass of the sample in respectively steady state conditions and initial dry state.

395 In Figure 11, the isotherms present a S-shape corresponding to the type IV of the Rogers 396 classification. This behavior is frequently observed in cellulose-based materials [45]. We 397 can see that pyrolyzed bagasse fibers are less hygroscopic than natural bagasse fibers, 398 particularly at high relative humidity (decrease of 30% of its sorption capacity). Indeed, 399 hemicellulose degradation during pyrolysis (between 160 and 260°C) [32] makes the 400 TBF less sensitive to moisture; generally, they become more hydrophobic [3]. This last 401 point encourages us to develop pyrolyzed bagasse fiber / cement composites. As 402 mentioned by Collet and Pretot [28], thermal conductivity of hygroscopic materials 403 increases with moisture content. The pyrolyzed fibers being more hydrophobic [3] than the raw fibers, they would generate less moisture in the composites that is to say that 404 405 the ratio air/water will be increased. As thermal conductivity of air is lower than that of 406 water [35], the resulting composites would have lower thermal conductivities (if we 407 consider a simple law of mixture) than the LFN and would allow our materials to be 408 considered as insulating materials in the field of construction.

- 409
- 410 6. Conclusions

411 A theoretical modeling based on auto-coherent homogenization is proposed to estimate

the thermal conductivity of bagasse fibers, that is a novelty. The model is also used to
estimate the conductivity of fibers and cement and then the conductivity of composites
under both dry (good fitting with the model) and wet conditions.

The bagasse fibers / cement composites materials present low thermal conductivity. The study shows that the thermal conductivity of such composites depends both on

- fibers / cement ratio (bulk density) and moisture content. The best insulating properties
  are obtained with an untreated bagasse fiber content around 8 % wt.
- Further investigations on pores size and distribution are required in order to improvethe accuracy of the model in wet conditions.
- 421 We explore the isotherm sorption of pyrolyzed bagasse fibers. These data confirm that
- the pyrolysis allows obtaining more hydrophobic fibers, which once included in cement,
- 423 would provide more insulating materials for building applications.
- 424

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580

# 581 Figures caption582

Figure 1: Auto-coherent method applied to two-phase medium - geometry of an elementary inclusion composite

585 Where  $R_a$  and thermal conductivity  $\lambda_a$  (component "a" is the air contained within the 586 vegetable fibers) surrounded by a concentric shell of component "s" (s is the fiber block) 587 of thermal conductivity  $\lambda_s$  and radius  $R_s$ ,  $\lambda_d$  is the thermal conductivity of the equivalent 588 homogeneous material also called effective conductivity

589

Figure 2: Auto-coherent method applied to three-phase medium - geometry of anelementary inclusion composite

592 Where a spherical air bubble "a" is surrounded by a concentric vegetable particles shell
593 "f " itself surrounded by a binder shell "s"
594

Figure 3 : Double homogenization of auto-coherent model (binder+ vegetable particles)
Where a is for air, w is for water and H for hydrated composite

Figure 4 : Bulk density of composites according to their fiber content in the dry state
(RH = 0%)

Figure 5 : Porosity of composites according to their fiber content in the dry state (RH =0%)

603

Figure 6 : Experimental thermal conductivity of LFN composites according to their fibercontent in the dry state (RH = 0%)

606

Figure 7 : Experimental thermal conductivity of LFN composites according to their bulk
density at different moisture contents

Figure 8 : Comparison between numerical calculations and experimental results of composites thermal conductivity in the dry state according to bulk density

612

Figure 9 : Thermal conductivity varying with bulk density: comparison between
numerical calculations and experimental results in the wet state (RH=56%)

Figure 10: Thermal conductivity varying with bulk density: comparison between
numerical calculations and experimental results in the wet state (RH=100%)

Figure 11: Isotherm sorption of natural (NBF) and pyrolyzed bagasse fibers (TBF) at
(22±3)°C. In red: sorption; in green: desorption.

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- 622

623 Table 1: Botanical composition of raw sugar cane bagasse fibers (NBF) [29].

Cellulose	Hemicellulose	Lignin	Extractives	Humidity	Sum	
wt %	wt %	wt %	wt %	wt %	(except humidity) wt %	
48.68	25.46	21.94	3.92	7.50	100	

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627 Table 2: Chemical composition and some physical characteristics of CEM I 52,5 N [5].

Content wt %							
SiO <sub>2</sub>	Fe <sub>2</sub> O <sub>3</sub>	Al <sub>2</sub> O <sub>3</sub>	CaO	Na <sub>2</sub> O	K <sub>2</sub> O	MgO	
21.45	0.22	4.08	65.05	0.06	0.20	0.49	
Loss on ignition (wt %)							
1.45							
Bulk density (g/cm <sup>3</sup> )							
3.08							
Median particle size (µm)							
15.70							
Specific area (cm <sup>2</sup> /g)							
4200							

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631 Table 3 : Formulations of LFN composites.

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Composites	Binder	NBF/binder ratio (by	Water/binder ratio (by	Mass (g) relative to 1000 g of cement paste			
-		mass)	mass)	NBF	CEM I	Water	
L	1	0	0.4	0	704	296	
(without fibers)							
LFN1	1	0.02	0.60	12.3	617.3	370.4	
LFN2	1	0.03	0.60	18.4	613.5	368.1	
LFN3	1	0.04	0.60	24.4	609.8	365.8	
LFN4	1	0.05	0.60	30.3	606.1	363.6	
LFN5	1	0.08	0.60	47.6	595.2	357.2	

- 635 Table 4 : Mixing sequence of elaboration of composites.

Mixing sequence	Time
Adding the pre-wetted binder in the mixing	0
container	
Mixing the binder at slow speed (140 rpm)	30 s
Adding the rest of the water	30 s
Mixing at high speed (285 rpm)	2 min
Progressive addition of vegetable fibers	2 min
Mixing at high speed (285 rpm)	5 min

*Figure 1 : Auto-coherent method applied to two-phase medium - geometry of an elementary inclusion composite* 



Figure 2 : Auto-coherent method applied to three-phase medium - geometry of an elementary inclusion composite



Figure 3 : Double homogenization of auto-coherent model (binder+ vegetable particles)

where a is for air, w is for water and H for hydrated composite



Figure 4 : Bulk density of composites according fiber content in the dry state (RH = 0%)



Figure 5 : Porosity of composites according to their fiber content in the dry state (RH = 0%)



### Figure 6 : Thermal conductivity of LFN composites according to fiber content



Figure 7: Thermal conductivity of LFN composites according to their bulk density at





Figure 8: Comparison between theoretical calculations and experimental results of thermal conductivity of dry composites according to bulk density



Figure 9: Thermal conductivity varying with bulk density: comparison between theoretical calculations and experimental results (RH=56%)



Figure 10: Thermal conductivity varying with bulk density: comparison between theoretical calculations and experimental results (RH=100%)



Figure 11 : Sorption isotherms of natural (NBF) and pyrolyzed (TBF) bagasse fibers at (22±3)°C. In red: sorption; in green: desorption.

