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1 **Hygric properties of wheat straw biocomposite containing natural additives**
2 **intended for thermal insulation of buildings**

3
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19 **Highlights**

- 20 • The hygroscopic properties of wheat straw external insulation materials are assessed.
- 21 • The isothermal water vapour diffusivity was estimated from the sorption curve and the
22 water vapor permeability.
- 23 • The impact of material's thickness on moisture buffer value was studied

24 **Abstract**

25 Straw concrete is a biobased composite developed for thermal insulation of new buildings
26 and thermal rehabilitation of old buildings. It is made of the combination of wheat straw, lime

27 or plaster binder and natural additives (hemoglobin, casein, gelatin). The natural additives are
28 used to improve the thermal and mechanical properties of the material. The characterization
29 of these composites has shown an attractive thermal conductivity and an acceptable
30 mechanical strength. The present paper focuses on the hygric characterization of these
31 materials which have different constituents (straw aggregates, binder and additives). Three
32 properties were experimentally determined, namely, sorption-desorption curves, water vapor
33 permeability and moisture buffer value (MBV). These hygroscopic properties are of great
34 importance when it comes to the comfort of the building. The isothermal water vapour
35 diffusivity was also estimated from the sorption curve and the water vapor permeability. The
36 impact of the sample thickness on the moisture buffer value (MBV) was also highlighted. The
37 results showed that the studied composites have excellent hygric properties which can
38 contribute significantly to ensure hygrothermal comfort in buildings. These hygric
39 performances are competitive with those of other insulating bio-based materials studied in the
40 literature and better than those of conventional materials used in construction.

41 **Keywords:** Wheat straw, lime, gypsume, sorption-desorption, water vapor permeability,
42 moisture buffer value.

43 **1 Introduction**

44 The use of porous biobased insulators in the construction of new buildings or the
45 rehabilitation of old ones offers many advantages such as biodegradability, renewability, local
46 abundance, and carbon dioxide storage, which contribute significantly to improving the
47 energy efficiency of the building and compliance with environmental requirements [1–3].

48 The hygric behavior of porous materials is generally studied through three properties:
49 sorption-desorption isotherms, permeability to water vapor and the moisture buffer value [4–
50 6]. The sorption-desorption curves are essential to evaluate the modifications of the properties

51 caused by the hygroscopic behavior and to deduce the water vapor diffusion properties. In
52 general, due to their high hygroscopicity as most of the porous materials, the sorption
53 isotherms of the materials based on vegetable aggregates present a sigmoidal form of type II
54 or III according to the IUPAC classification [7]. Biobased composites have a high moisture
55 storage capability compared to other building materials. Samri 2008 [8] compared the
56 adsorption isotherms of three materials, namely aerated concrete clay and hemp concrete. He
57 noticed that the moisture storage capacity of the hemp concrete is higher when the relative
58 humidity is between 40% and 90% unlike the other two materials. According to the author,
59 this property (moisture storage capacity) has a direct consequence on the capacity of the
60 materials to fluctuations in relative humidity, present the room (human activity, kitchen,
61 bathroom). The different vegetal concretes studied in the literature show a similar behaviour.
62 For the adsorption isotherm, the mass water content of the materials increases progressively
63 as a function of relative humidity up to 80% RH depending on the constituents of each
64 composite. Beyond RH=80% the water content increases rapidly to reach values between 10
65 and 26% depending on the material [8–11]. Conversely, in desorption, the water content
66 decreases rapidly from RH=98% to RH=80% and then evolves progressively between
67 RH=80% and low humidity [8–11]. From this very sensitive behavior to humidity, an
68 important hysteresis between the adsorption and the desorption isotherm is present and has
69 been highlighted by several authors for the different composites of plant-based concretes [8–
70 11]. It has been noted that the water content of the material observed in adsorption for a given
71 relative humidity is lower than that observed in desorption. This difference is explained by the
72 existence of pores in the shape of ink bottle.

73 The effect of the type of constituents (aggregates, binders) and their fractions on the
74 adsorption isotherm has also been discussed, Mazhoud 2017 [12] noted that composites with a
75 lime-based matrix possess a higher water content than composites with a clay-based matrix.

76 However, Chamoin 2013 [13] showed that the binder type slightly impacts the adsorption-
77 desorption isotherm of hemp concrete. Indeed, the substitution of the lime-based binder with a
78 calcium sulfate-based binder leads to a similar water content value, except at high relative
79 humidity (above 80% RH), where capillary condensation occurs. Belayachi et al 2015 [14],
80 found no influence of aggregate type on the adsorption-desorption isotherms of straw
81 concrete. Barley straw-based composites have similar curves to that of wheat straw-based
82 composites. Brouard et al 2018 [15] did not observe a significant difference between the
83 behavior of rapeseed-based formulations and those with sunflower bark. On the other hand,
84 other authors have observed that an increase in the aggregate/binder ratio leads to higher
85 moisture content values, especially at high relative humidities. This was attributed, on the one
86 hand, to a higher polymolecular adsorption and on the other hand, to capillary condensation
87 [9,12,16]

88 The water vapor permeability represents the capacity of a material to transfer moisture.
89 The determination of this parameter is essential in the hygric characterization of materials and
90 their use in the design of effective thermal insulation of buildings. The most used method in
91 the literature to measure this coefficient is the cup method, described in the standard EN ISO
92 12572 [17].

93 Overall, due to their high open porosity, the water vapor permeability of plant-based materials
94 is high. Several authors in the literature have studied the water vapor permeability of plant-
95 based materials using the cup method [17]. Collet 2004 [18], Evard 2008 [19], Chamoin 2013
96 [13], Walker et al 2014 [20], Bennai 2017 [11] determined the water vapor permeability of the
97 various hemp concrete mixtures. They found values between $1.6 \cdot 10^{-11}$ and $3 \cdot 10^{-11}$
98 (kg/m.s.Pa) depending on the constituents of each composite. Rahim et al 2015 [21], Rahim
99 et al 2016 [22], obtained a value of $2 \cdot 10^{-11}$ and $4.5 \cdot 10^{-11}$ (kg/m.s.Pa) for rape straw concrete
100 and a value of $2.34 \cdot 10^{-11}$ (kg/m.s.Pa) for flax concrete. These experimental values proposed

101 by various authors confirm the same order of magnitude for the different biobased materials
102 without much effect of aggregate and binder type.

103 The relative humidity is a factor that strongly influences the water vapor permeability.
104 Kumaran et al 2002 [23], Collet et al 2013 [16] determined the water vapor permeability of
105 biobased composites as a function of the humidity. They indicated that the permeability
106 evolves according to an exponential or power law with increasing humidity.

107 The measurement of water vapor permeability and the determination of adsorption-
108 desorption isotherms allow to deduce the isothermal water vapour diffusivity. This coefficient
109 characterizes the change rate of the water content of a material under transient changes in
110 humidity. Collet et al 2012 [24], Rahim et al 2015 [21] and Rahim et al 2016 [22] estimated
111 the water vapour diffusivity coefficient for hemp, rapeseed and flax concrete, they found
112 values between 10^{-10} and 10^{-7} m²/s.

113 Among the hygric parameters of porous materials of interest for the design of a
114 comfortable indoor environment, the Moisture Buffer Value (MBV). This property indicates
115 the ability of the material to regulate the moisture variations in the ambient air. The MBV is
116 measured according to the approach defined in the Nordtest Project Rode et al. 2005 [25].
117 According to several works in the literature [10,21,22,24,26–29] plant-based concretes have
118 shown a MBV higher than 2g/(m².%RH), which allowed them to be classified as an excellent
119 water regulators according to the classification of Rode et al 2005 [25].

120 The present paper aims to study the hygric behavior of three biobased materials,
121 developed in the framework of the research program PEPITE with a great potential of thermal
122 insulation of buildings by interior or exterior [30]. These new materials are based on cereal
123 straw, lime or gypsum binder and additives from a renewable and biodegradable source to
124 optimize their performances for the hygrothermal comfort of buildings. Three hygric

125 properties were experimentally determined, namely sorption-desorption curves, water vapor
126 permeability and moisture buffer value. The objective is to compare the hygric performance
127 of the three optimized composites which have different constituents such as the straw fraction,
128 the type of binder and the type of additives. The isothermal water vapour diffusivity was
129 estimated from the sorption-desorption curves and the water buffer value. The impact of the
130 thickness of the material on the moisture buffer value was also investigated. The influence of
131 additives and thickness on hygric properties is analyzed based on the comparison between our
132 research outcomes and literature findings.

133 **2 Materials and methods**

134 **2.1 Materials**

135 *2.1.1 Raw materials*

136

- 137 • *Wheat straw*: the straw used in this study to prepare the biocomposites was harvested
138 in the Region Centre Val de Loire in France. It was manually cut in the laboratory
139 only in the longitudinal direction in order to maintain its natural tubular shape. The
140 size obtained after cutting is the one that was intended for mixing with 90 % straw of
141 3.5 cm.

142 Two mineral binders were used in the present study to prepare the biocomposites:

- 143 • *Tradical PF70*: is a pre-formulated binder manufactured in accordance with the
144 requirements of NF 459. It is based on aerial lime (75%), hydraulic binders (15%) and
145 pozzolan (10%).
- 146 • A commercial plaster produced by Diall, wich is characterized by its quick setting
147 time (10-15 minutes).

148 Three natural additives was adopted to optimize the thermal and mechanical properties
 149 of biocomposites :

- 150 • *Hemoglobin*: this is a proteinaceous substance from pork blood obtained by
 151 fractionation and spray drying. Vepro 95 PHF is the commercial name of the product
 152 used in this study and is manufactured by the Vapran Group in powder form.
- 153 • *Casein*: this is a protein extracted from milk, produced by EPI Ingredients Group in
 154 the form of a fine white powder (<400µm). It is used as a natural fixative for lime
 155 coating.
- 156 • *Gelatin*: this is a technical gelatin 200 bloom produced by the Weishardt Group in
 157 powder form with a particle size less than 20 mesh (841 µm).

158 2.1.2 Biocomposites

159
 160 Three straw composites were selected from previous work [30] for this investigation, the first
 161 one is based on lime, hemoglobin, and casein (Fig.1-a). These additives were used to increase
 162 the porosity of the matrix and improve the fiber-matrix interface. The second one is based on
 163 lime and gelatin (Fig.1-b). The last one is based on gypsum plaster and gelatin. The use of this
 164 biopolymer allows increasing of straw fraction (Fig.1-c). The manufacturing procedure of the
 165 developed biocomposite is detailed in the previous work of Ismail et al. 2020 [30]. The
 166 composites are denoted respectively LHC, LGB and GGB, as indicated in Tab. 1.
 167 The porosity of composites was determined by the pycnometer method, the values obtained
 168 for the three materials are remarkably close.

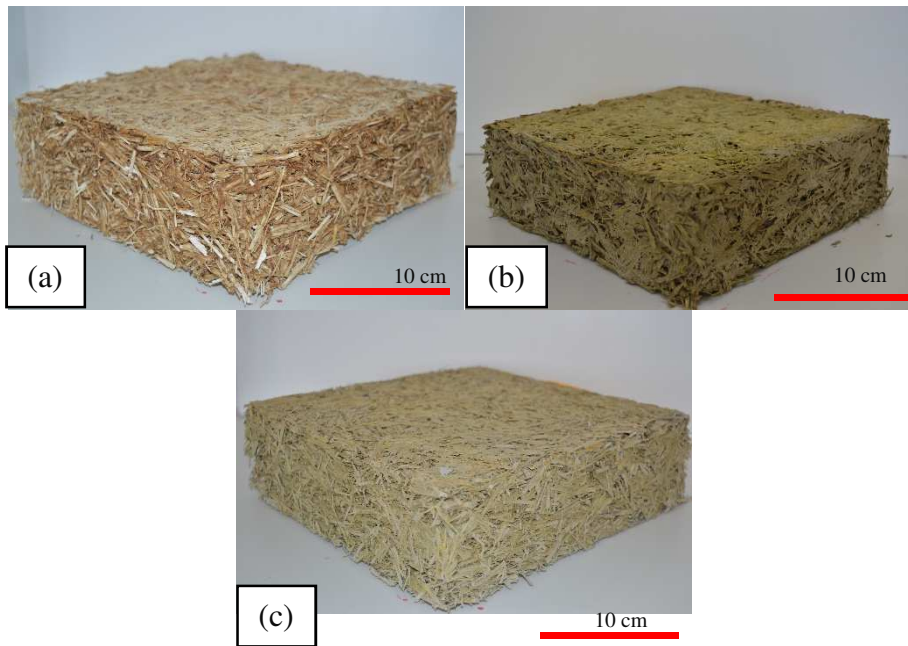
Mixture-ID		Mass ratio (kg/kg)				Density (Kg/m ³)	Total porosity (%)
Short Ref	Long Ref	S/B	H/B	C/B	GB/B		
LHC	Lime+5% H+2.5% C	0.3	0.05	0.03	-	308	85.47
LGB	Lime+20%GB	0.4	-	-	0.2	288	85.37
GGB	Gepsume+10%GB	0.4	-	-	0.1	277	87.14

169 S/B: Straw/Binder H/B: Hemoglobine/Binder C/B: Caseine/Binder
 170 GB/B: GelatinBinder/Binder.

171

Tab. 1. Composites designation and detailed mixture proportions

172



173

174

Fig. 1. Appearance of composites (a) LHC (b) LGB and (c) GGB

175 2.2 Methods

176 In order to evaluate the hygric behavior of the selected biocomposites, three types of
177 experimental tests were carried out, the sorption-desorption, the water vapor permeability and
178 the moisture buffer value

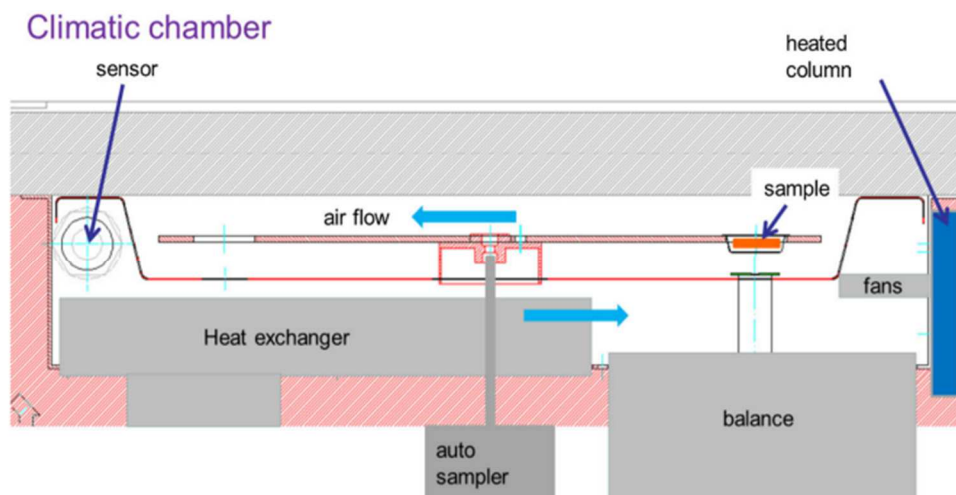
179 2.2.1 Sorption-desorption isotherms

180

181 Sorption-desorption isotherms are often determined by gravimetric measurement, where
182 material samples are placed in different relative humidities at constant temperature. Mass
183 tracking is used to determine the equilibrium point and to calculate the water content for a
184 given humidity. The most commonly used method is the saline solution method, which allows
185 the use of nine humidity levels between 10% and 98%.

186 In this work, the sorption-desorption curves of the three composites were determined
187 automatically using Vsorp equipment from ProUmid GmbH, (Fig. 2). The device is equipped
188 with a sample tray and a balance with a resolution of 100 μg . The sample tray is located in a

189 climatic chamber with controlled temperature and humidity. Materials samples of
190 approximately $5 \times 5 \times 5 \text{ cm}^3$ were prepared for each composite and dried at 60°C until
191 constant mass was reached. Then, the material samples were placed in aluminium pans of
192 sample tray. Nine humidity levels (varying from 10% to 90% RH with a 10% step) were set at
193 a constant temperature of 25°C . The weighing of the samples was performed automatically
194 every 15 min. Equilibrium is assumed to be reached when the mass variation does not exceed
195 0.01% within 24 h.



196

Fig. 2. Sorption-desorption test (Vsorp equipment)

197 2.2.2 Water vapour permeability

198 The Vsorp equipment was also used here to measure the water vapor permeability of the
199 composites based on the cup method according to the European Standard EN ISO 12572 [10].

200 The test procedure consists of applying a pressure difference between the upstream and
201 downstream sides of the specimen and then calculate the amount of water vapor flow through
202 the material (Fig. 4). The density of water vapor flow rate, g , is given by the following
203 equation:

$$g = \frac{G}{A} \quad (1)$$

204 With :

205 A : is the exposed area of the test specimen (m^2)

206 G : represents the mass change rate, calculated from the mass versus time curve. According to
207 the standard EN ISO 12572 [10], the value of G is obtained when each of the last five
208 successive determinations of Δm_{12} is within $\pm 5\%$ of G (the mean of five successive
209 determinations).

210 Where :

$$\Delta_{12} = \frac{m_{i+1} - m_i}{t_{i+1} - t_i} \quad (2)$$

211 With : m_i is the mass at time t_i ; m_{i+1} is the mass at time t_{i+1}

212 The moisture transfer under a vapour pressure gradient can be expressed as:

$$g = -\delta \nabla p_v \quad (3)$$

213 So, the water vapour permeability, δ ($kg/(m.s.Pa)$), is given by the equation 3 :

$$\delta = g \frac{e}{\Delta p_v} \quad (4)$$

214 Where :

215 e : is the thickness of the test specimen in m

216 Δp_v : is the water vapour pressure difference Pa

217 The vapour pressure on either side of the specimen was calculated from the temperature (T)
218 and relative humidity (HR) according to the formula proposed by the standard [10]:

$$p_v = \text{HR} \cdot 610.5 e^{\frac{17.269 T}{237.3 + T}} \quad (5)$$

219 In order to take into account, the resistances of the layer of air in the test cup between the base
220 of the specimen and the desiccant or saturated salt solution, and on both internal and external
221 sides of the specimen. The standard EN ISO 12572 [10] and other works [20-21] of the
222 literature proposes to correct the water vapour permeability calculated in formula 2 by
223 formula:

$$\delta = \frac{e}{\frac{\Delta p_v}{g} - (Z_{p,air} + Z_{p,in} + Z_{p,out})} \quad (6)$$

224 With :

225 $Z_{p,air}$: is the resistances of the layer of air in the test cup between the base of the specimen
226 and the desiccant or saturated salt solution is given by:

$$Z_{p,air} = \frac{d_a}{\delta_a} \quad (7)$$

227 Where d_a (1.5 cm) is the thickness of the air layer and δ_a is the water vapour permeability of
228 air.

229 $Z_{p,in}$ and $Z_{p,out}$ ((Pa.m² s)/kg) are the resistance of the boundary layers which can be estimated
230 from the convective mass transfer coefficients (ρ) [20-21].

$$Z_p = \frac{1}{\beta_p} \quad (8)$$

231 β_p is estimated using the following equation:

$$\frac{1}{\beta_p} = \frac{R_v \cdot T \cdot (\rho \cdot C_p)_{air}}{h_c} Le^{3/4} \quad (9)$$

232 where q is the air density (125 kg/m³), C_p : heat capacity of air (1005 J/(kgK)), R_v : gas
 233 constant for vapor (461.5 J/(kgK)), T : temperature (20°C), Le is the Lewis number (ratio
 234 between the thermal diffusivity to vapor diffusivity of air and the volumetric heat capacity of
 235 air), and h_c : the convective heat transfer coefficient (W/(m²K)), is estimated from the air
 236 velocity (m/s) by the following relation [20-21]:

$$h_c = 5.83 + 3.96 \cdot v \quad (10)$$

237 The $h_{c,out}$ is calculated from the air velocity above the specimen (0.25 m/s) and $h_{c,in}$ is
 238 calculated from the air velocity below the specimen (0 m/s).

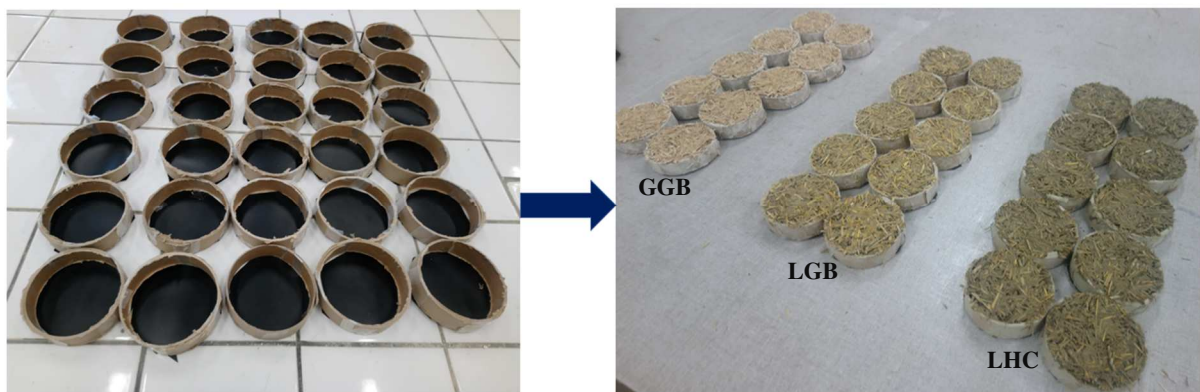
239 Several works in the literature also use the vapor resistance factor (μ) instead of the water
 240 vapor permeability:

$$\mu = \frac{\delta_a}{\delta} \quad (11)$$

241 where δ_a is the water vapor permeability of air (2.10⁻¹⁰kg/(m.s.Pa)).

242 In this work, the water vapor permeability of composites was evaluated under dry cup
 243 (3%/50% RH) and wet cup (93%/50% RH) conditions. The tests were performed at 23°C, the
 244 50% RH was provided with a climatic chamber of Vsorp equipment and the 3% RH or 93%
 245 RH was maintained by the molecular sieve (spherical metal aluminium silicates with sphere

246 diameters between 1.6 and 2.5 mm) and the saturated salt solutions of KNO_3 , respectively.
247 Circular samples with a diameter of 8.5 cm and a thickness of about 1 cm, were prepared in
248 cylindrical cardboard moulds for each material (Fig. 3). After a curing period of 28 days, the
249 specimens were conditioned at 50% RH and 20°C until mass stabilization. To avoid errors
250 due to leakage, a mixture of 60% of microcrystalline wax with 40%
251 refined crystalline paraffin was used to ensure a seal between the sample and the cup. The
252 weighing of the cups is programmed every 10 minutes.



253

254 **Fig. 3** : Preparation of samples for water vapor permeability test (a)

255 cylindrical cardboard moulds (b) samples of composites

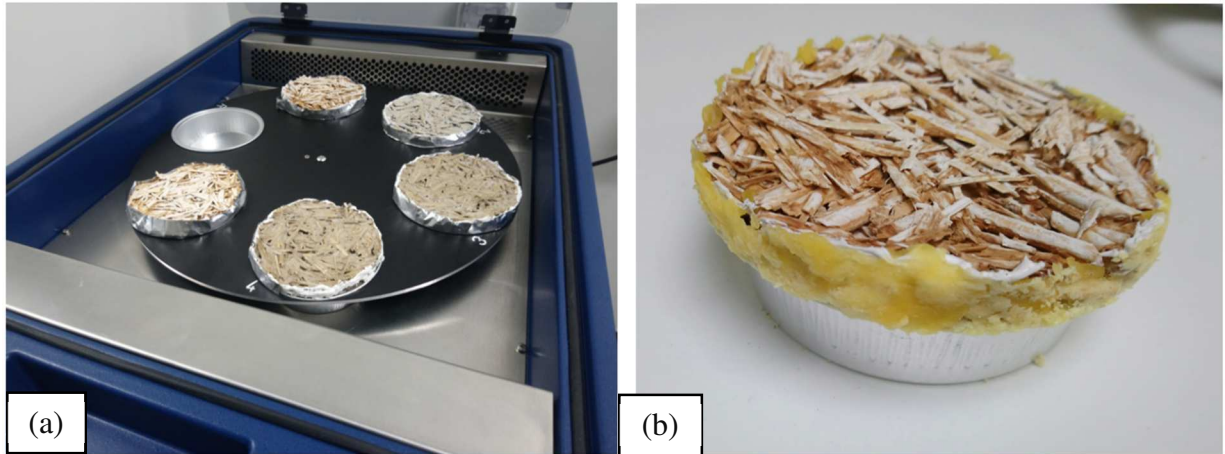
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261

262 **Fig. 4.** Water vapor permeability test (a) experimental device (Vsorp) (b) and (c) sample+cup
 263 used for measuring water vapor permeability according to standard cup-method

264 *2.2.3 Isothermal water vapour diffusivity*

265 The combination of the results obtained for the adsorption isotherms and the water vapor
 266 permeability allows the calculation of the isothermal water vapour diffusivity D_w . Indeed,
 267 under isothermal conditions, the vapour flux density is given by the following equation:

$$g = -D_w \nabla \omega \quad (12)$$

268 With :

269 D_w : water vapour diffusivity in m^2/s

270 ω : water content in kg/m^3

271 For a one-dimensional transfer the equation can be written as a function of the relative
 272 humidity as follows :

$$g = -D_w p_{sat} \frac{\partial HR}{\partial x} \quad (13)$$

273

274 HR : relative humidity

275 p_{sat} : saturated vapor pressure in Pa

276 So, the isothermal water vapour diffusivity is calculated from sorption and water vapour
 277 permeability according to the following equation:

$$D_w = \frac{p_{sat} \cdot \delta}{\frac{\partial \omega}{\partial HR}} \quad (14)$$

278 $\frac{\partial \omega}{\partial HR}$ represents the derivative of the adsorption isotherm which reflects the moisture storage
 279 capacity of the material.

280 In order to calculate $\frac{\partial \omega}{\partial HR}$, a modeling of the adsorption isotherm is necessary. For that, several
 281 models were used in the literature, such as the model of Langmuir 1918 [31], the model of
 282 Brunauer-Emmet-Teller (BET) [32], the model of Guggenheim-Anderson- DeBoer (GAB) or
 283 the model of Generalised-D'Arcy-Watt (GDW). These last two are derivatives of the
 284 Langmuir model and are the most widely used in the literature.

285 In this study, the GAB model was used to smooth the adsorption curve of the three
 286 composites. The expression of this model is given by the following equation:

$$287 \quad \omega = \frac{\omega_m C_2 C_3 HR}{(1 - C_3 HR)(1 - C_3 HR + C_2 C_3 HR)}$$

$$288 \quad C_2 = \exp\left(\frac{E_1 - E_m}{RT}\right), C_3 = \exp\left(\frac{E_L - E_m}{RT}\right)$$

289 With :

290 w : water content by mass of the material,

291 w_m : water content of the material when the walls are completely covered,

292 E_1 : molar heat of adsorption in J/mol,

293 E_L : latent molar heat of vaporization in J/mol,

294 T : temperature in K,

295 R : perfect gas constant.

296 The constants ω_m , C_2 and C_3 are determined by fitting the GAB model to the experimental
297 measurements using the least squares method.

298 2.2.4 Moisture buffer value (MBV)

299 The moisture buffer value is measured according to the protocol developed by Rode et al
300 2005 [25]. The test was performed using a Binder climatic chamber. The samples were
301 subjected to a cycle of 24 hours in which the relative humidity was fixed at 75% for 8 hours
302 and then at 33% for 16 hours (Fig. 5). An adjustment of the chamber fan speed was necessary
303 to obtain the value recommended by the protocol (0.1 ± 0.05 m/s) in the vicinity of the
304 samples. The samples were weighed five times in the adsorption phase and twice in the
305 desorption phase using a balance with a resolution of 0.01 g. The test was stopped when the
306 difference between the mass variations recorded became less than 5% over the last three
307 cycles. The moisture buffer value was calculated according to the following equation:

$$MBV = \frac{\Delta m}{A \cdot (HR_{High} - HR_{Low})} \quad (15)$$

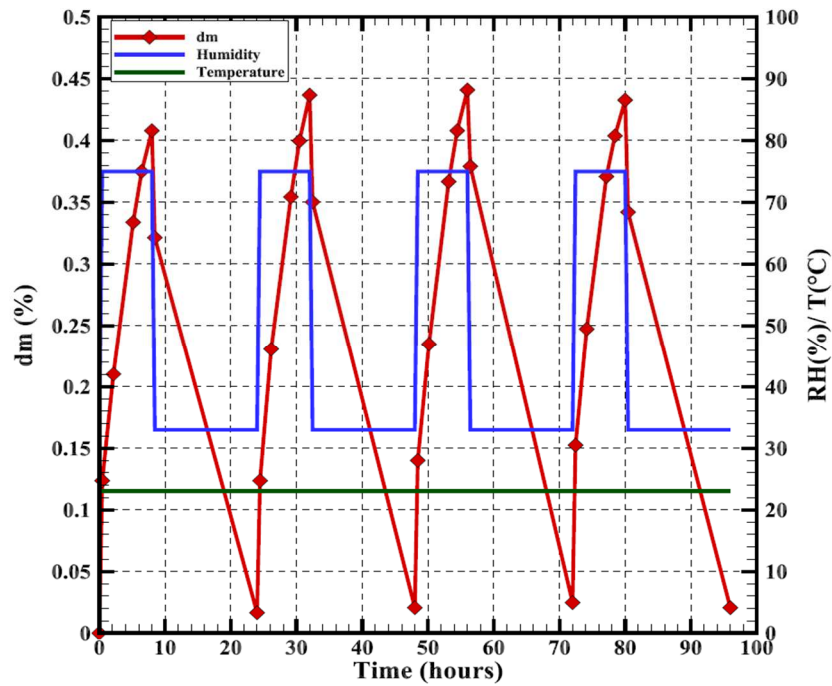
308 Where :

309 MBV: is the moisture buffer value ($\text{g/m}^2 \cdot \text{RH} (\%)$),

310 Δm : is the mass variation during the sorption or desorption phase (g),

311 A : is the exchange surface (m^2),

312 HR_{High} , HR_{Low} : are (75 %) for the high relative humidity and and (33 %) for the lower one.



313

314

Fig. 5. Example of a moisture uptake and release cycles for a sample

315 In order to evaluate the influence of material thickness on MBV, four thicknesses were

316 studied for each composite: 6 cm, 8 cm, 10 cm and 12 cm. Three samples with an area of $10 \times$

317 10 cm^2 were prepared for each composite and each thickness (Fig. 6). All sample surfaces are

318 protected with adhesive aluminium tape to leave a single exchange surface as shown in Fig. 7.



319

320

Fig. 6 : Experimental setup and placement of samples

321 **3 Results and discussion**

322

323 *3.1.1 Sorption-desorption isotherms*

324

325 Fig. 7 shows the mass variation of the three composites as a function of time for the different
326 humidity levels imposed at constant temperature. It can see that for the humidity levels below
327 80% RH, the mass stability was reached in a few days. However, for the 90% humidity value,
328 the kinetics of mass gain becomes very slow, especially for lime-based composites. The
329 achievement of equilibrium required two months respectively for LGB and LHC compared to
330 4 days for the gypsum plaster-based composite (GGB). This shows that the binder type
331 influences the moisture adsorption kinetics of the composites. It was observed that during the
332 90% humidity level, the water content of the plaster composite (GGB) was decreased slightly
333 (by 1%) between 60 days and 110 days. This reduction may be due to the formation of mold
334 in this composite which leads to the modification of its water content. Indeed, it was observed
335 at the end of the test an appearance of mold inside and on the surface of the samples of this
336 material. The curves also show that the desorption kinetics is very fast compared to the
337 adsorption kinetics. The desorption path time is approximately 18 days against 4 months for
338 adsorption path. This shows the very long time necessary for the hygric characterization of
339 porous materials, and the biobased materials particularly.

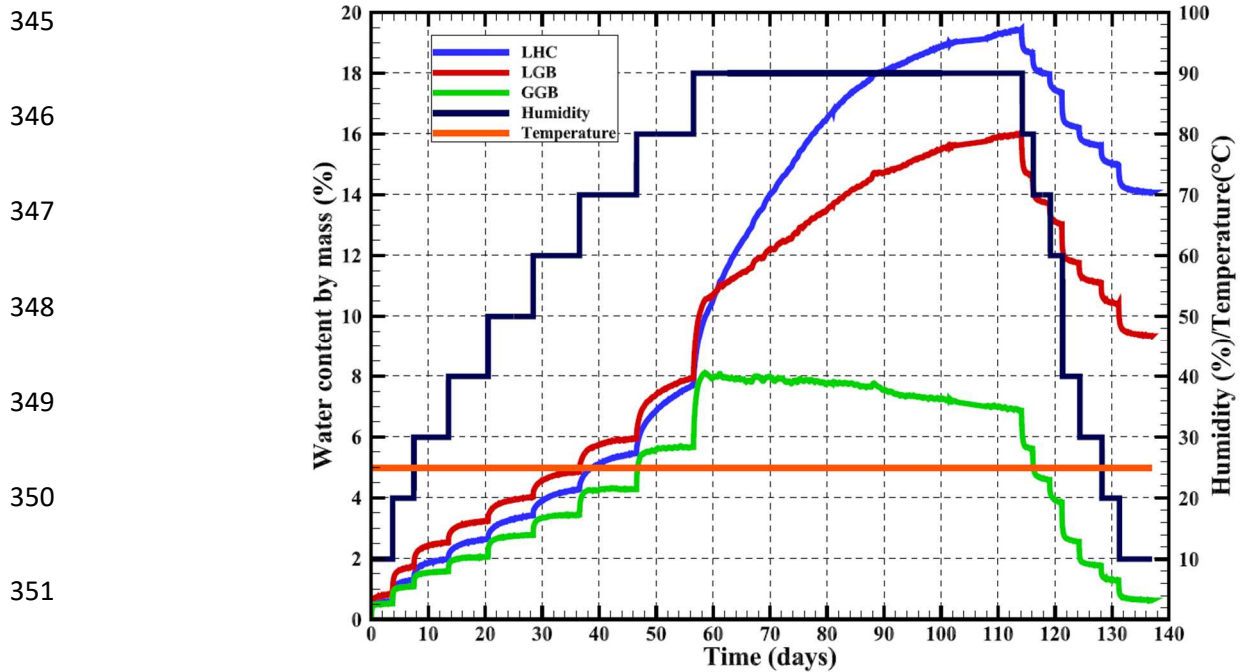
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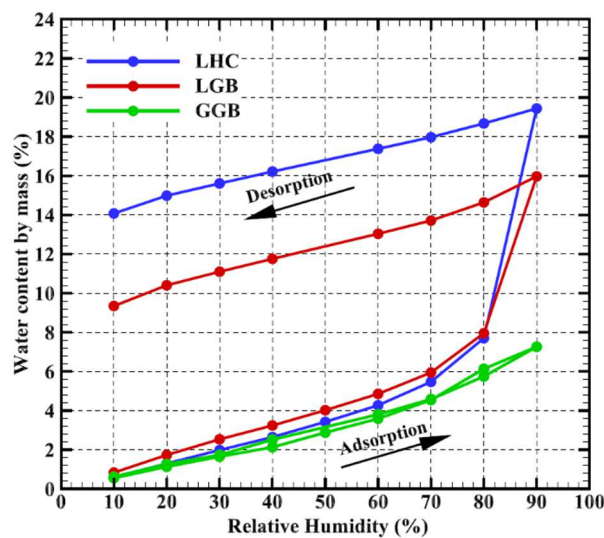
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353 **Fig. 7.** Mass variation as a function of time at different humidity levels

354 Fig. 8 shows the sorption-desorption isotherms for the three studied composites. The shape of
 355 the curves is similar to that found in the literature for other plant-based materials. For the
 356 LHC and LGB lime-based composites, the water content in adsorption increases progressively
 357 and almost linearly with the increase in humidity up to a RH=80%. Beyond this last value, the
 358 water content increases rapidly to reach a value of 20% and 16% respectively for LHC and
 359 LGB. In desorption, the water content decreases progressively and linearly over the whole
 360 moisture range until reaching a value of 14% and 9.8% for a humidity of about 10 %. This
 361 was reflected in a large hysteresis that was observed over all humidity levels for lime based
 362 biocomposite. This hysteresis can be explained by the existence of the « ink bottle » shaped
 363 pores but also by the consumption of H₂O during the carbonation reaction. Indeed, as it has
 364 been noted above, the samples tested had a cure time of 28 days, whereas the complete
 365 carbonation of lime can last several months [33].

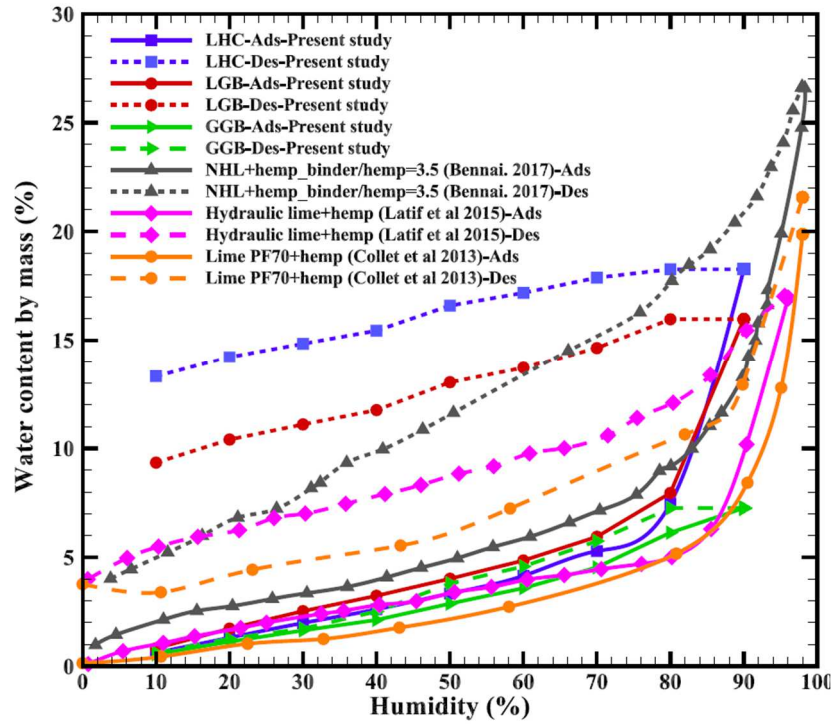
366 For the gypsum-based composite (GGB), the increase in water content was almost linear over
367 the whole study humidity range, a small hysteresis was observed between RH=40% and
368 RH=70% but with a negligible area compared to that observed in the case of lime-based
369 composites.

370 Fig. 9 shows a comparison of the adsorption-desorption curves of the studied composites with
371 those of other biobased materials from the literature. The evolution of the water content in
372 adsorption and desorption is almost similar between the different materials. However, it is
373 clear that the hysteresis between the adsorption and desorption curve of LHC and LGB
374 composites is more significant than that of other materials in the literature. The hysteresis
375 observed in the case of plaster-based composite (GGB) is less important than that of hemp
376 concrete. These results can be explained by the variation of the constituents such as the binder
377 type, the binder content, and the nature of the plant aggregates which results in a different
378 porosity network and consequently a different hygric behaviour of the materials.



379

380 **Fig. 8.** Adsorption-desorption isotherms of the three composites

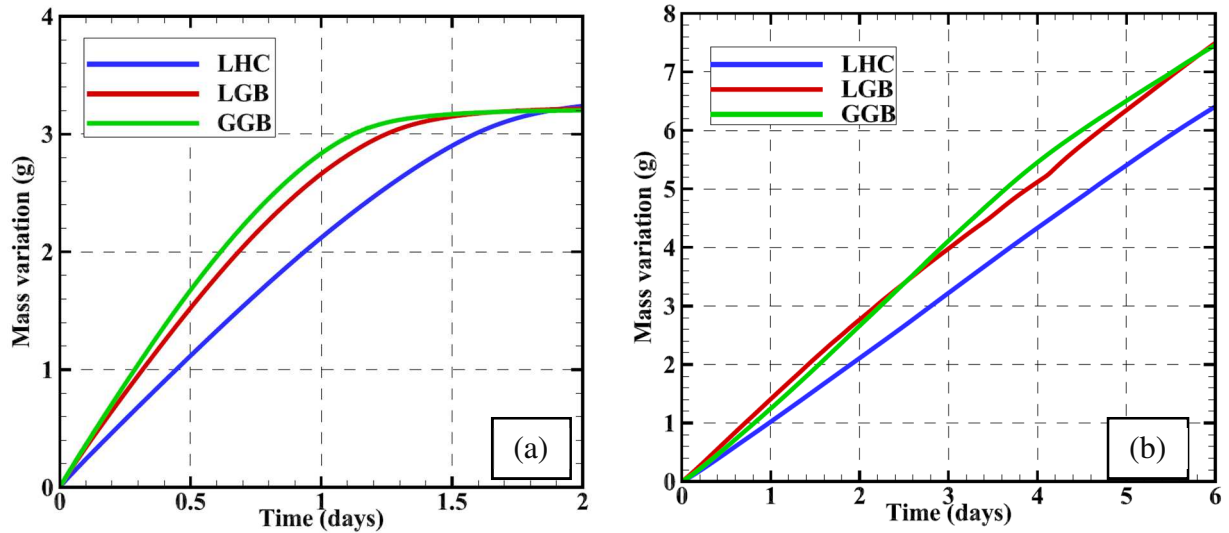


381

382 **Fig. 9.** Adsorption-desorption isotherms of the studied composites with other biobased
 383 materials in the literature

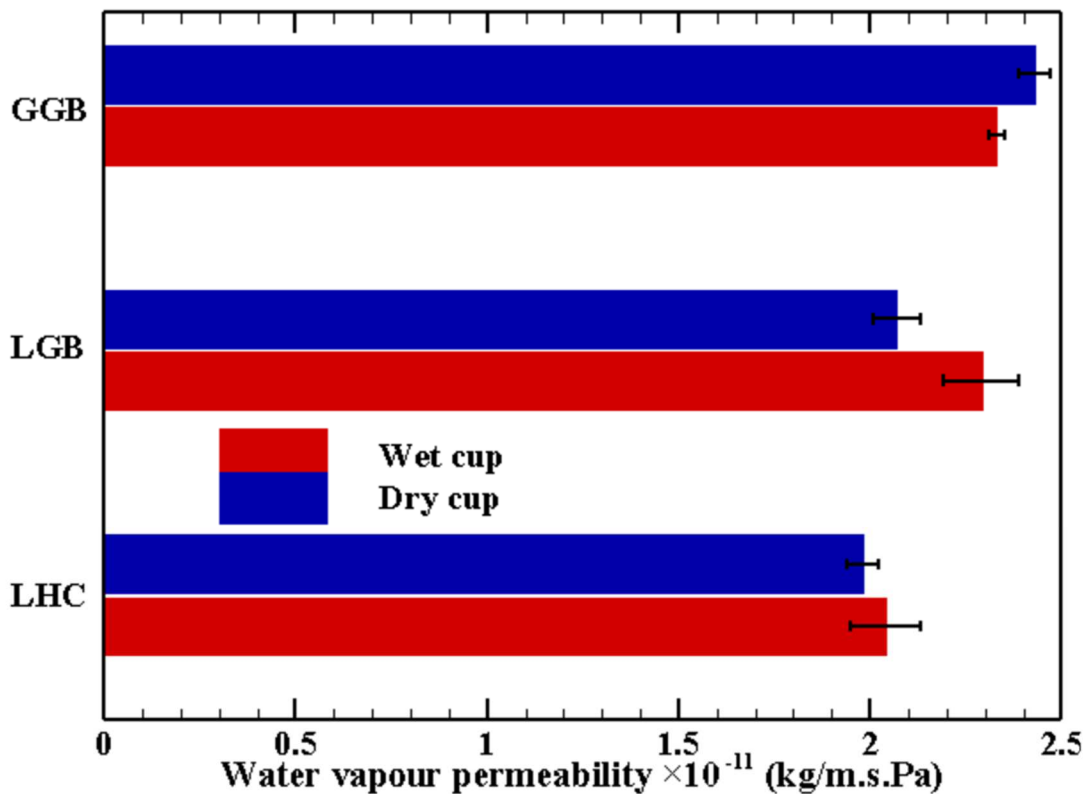
384 *3.1.2 Water vapor permeability*

385 The mass variation (cup and sample) is illustrated in Fig. 10 as a function of time, obtained
 386 for the dry and wet cup chosen here for the study. It can be seen that the saturation of
 387 molecular sieve was reached during 24 h, because of its ability to absorb moisture very
 388 quickly. These curves were used to calculate the flow density through the samples in order to
 389 deduce the water vapor permeability.



390 **Fig. 10.** Variation of mass as a function of time (a) dry cup (3%/50%) (b) wet cup (50% /
 391 93%)

392 We can notice that the deduced water vapor permeability of the three composites (Fig. 11) is
 393 in the same order of magnitude, both for the dry (3%/50% RH) and wet cup (50%/93% RH).
 394 The values obtained in both cases are close and the standard deviation remains in the same
 395 range for the three composites. This can be justified by the fact that the porosities of three
 396 composites are very close (Tab. 1). We also note that the binder type has no impact on the
 397 coefficient of permeability to water vapor, with values which remain quasi-similar for lime or
 398 gypsum plaster in the case of the wet and dry cup. $4.3 \cdot 10^{-11}$ kg/m.s.Pa. The same result was
 399 showed by Chamoin 2015 [13]. They noted that the water vapor permeability remains almost
 400 constant despite the substitution of a binder (Tradical PF70) by another (Microcem),
 401 regardless of the replacement rate. Evard 2008 [18] also found that the water vapor
 402 permeability of hemp concrete is about $4.3 \cdot 10^{-11}$ kg/m.s.Pa and $4.4 \cdot 10^{-11}$ kg/m.s.Pa
 403 respectively for wet and dry cup.



404

405 **Fig. 11.** Water vapor permeability of the three composites for the dry (3%/50% RH) and wet
 406 cup (50%/93% RH)

407 The obtained values for the three composites are in the same order of magnitude as those of
 408 other plant aggregate materials proposed by other authors in the literature as shown in Tab.2.
 409 We compare also the vapor resistance coefficient which represents the ratio of the diffusion
 410 coefficients of water vapour in air and in the building materials proposed by several studies.
 411 These values show that water vapour passes quickly through the biobased materials, thus
 412 avoiding the problem of condensation but it is necessary to remain vigilant in the case of
 413 rehabilitation of old building and to study the composition of the existing walls.

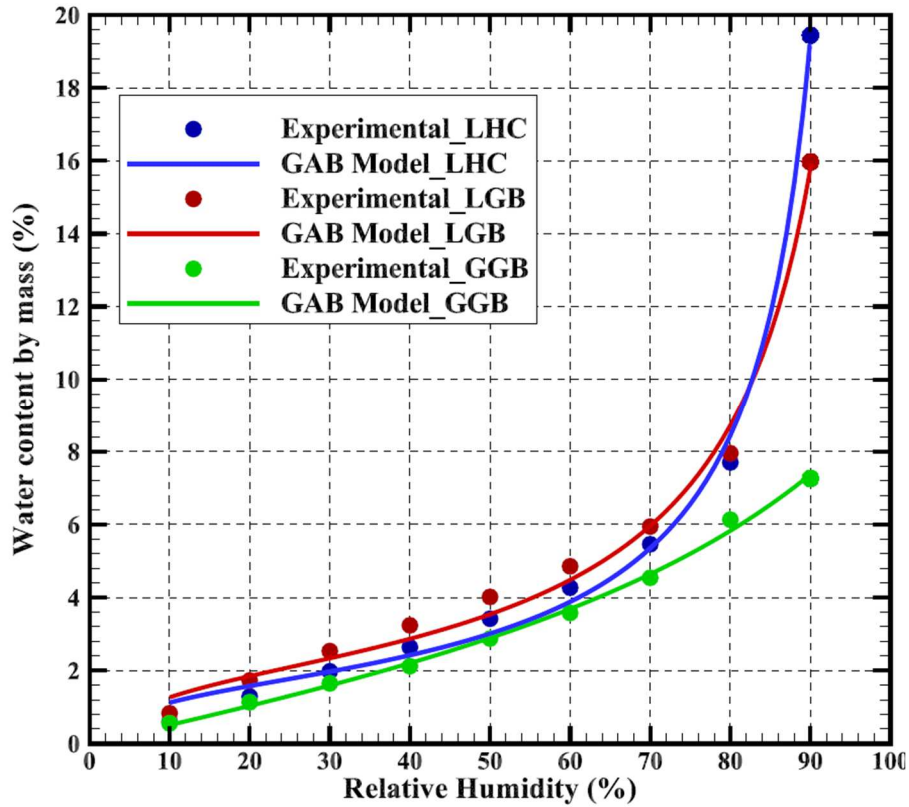
Material	$\rho_{app}[\text{kg/m}^3]$	$\mu[-]$	References
LHC	312	9.47	Present study
LGB	294	8.45	Present study
GGB	285	8.35	Present study
Lime + hemp	405 – 440	10 – 12	Collet 2004 [34]

Lime + hemp	400 – 466	5 – 7.5	Evard 2008 [19]
Lime + Microcem + hemp	406 – 475	6.46	Chamoin 2013 [13]
Lime + treated hemp	469	7.23	Chamoin 2013 [13]
Lime+colza	469	7.57	Rahim et al 2016 [21]
Lime+flax	469	8.29	Rahim et al 2015 [20]
Ordinary concrete	2300 – 2600	130	R-Thermique 2005 [35]
Natural pumice concrete	950 – 1150	50	R-Thermique 2005 [35]
Autoclaved aerated concrete	765 – 825	10	R-Thermique 2005 [35]
Wood concrete	450 – 650	15	R-Thermique 2005 [35]
Terracotta	2300 – 2400	16	R-Thermique 2005 [35]

414 **Tab. 2.** Vapor resistance coefficient of the studied materials in comparison with other
415 materials in the literature (dry cup)

416 3.1.3 Isothermal moisture diffusivity

417 As explained above, the calculation of moisture diffusivity requires the identification of the
418 parameters of GAB model selected for fitting of the adsorption curve as shown in Fig. 12 by
419 using the three curves for the studied biocomposites. Tab.3 gathers the estimated model
420 parameters. It is clear that the GAB model perfectly reproduces the experimentally obtained
421 adsorption isotherms for the three materials.



422

423

Fig. 12. Fitting of absorption curves using the GAB model

Composite	ω_m (%)	C_2	C_3
LHC	1.56	16	1.02
LGB	1.96	12.88	0.97
GGB	3.89	1.91	0.67

424

Tab. 3 GAB model parameters

425 Finally, by introducing the storage capacity with the water vapor permeability in equation 4,

426 the evolution of the isothermal moisture diffusivity as a function of water content can be

427 deduced. The results obtained for the three composites are presented in Fig.13. We can

428 observe that the evolution of the water vapour diffusivity of two lime-based materials is

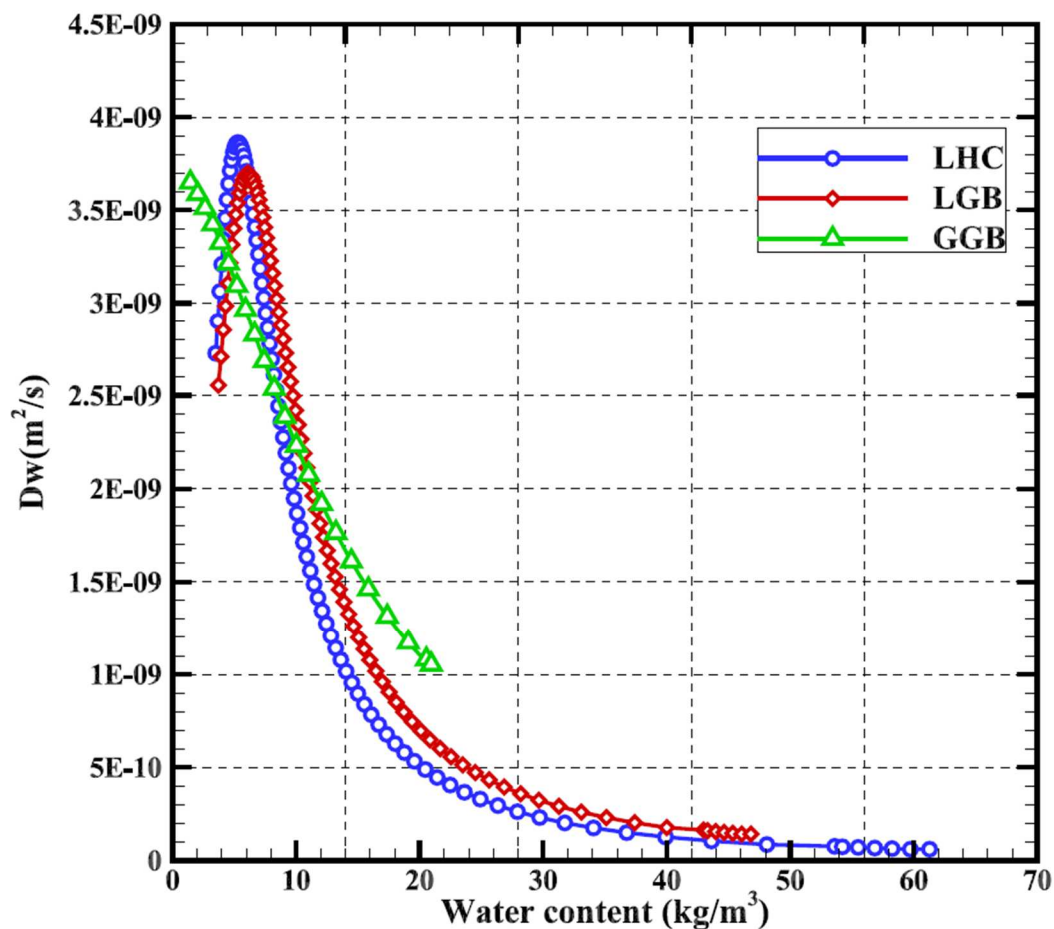
429 similar. Moreover, the shape of the three curves at low and medium water contents, coincides

430 with the one predicted by De Vries 1958 [36]. For the gypsum-based composite only the

431 decay is observed. This can be explained by the difference in the derivative of the sorption-

432 desorption isotherm curve calculated from the GAB model. On the other hand, the maximum

433 values of the water vapour diffusivity for the three composites are very close. They are equal
 434 to $3.9 \cdot 10^{-9}$, $3.7 \cdot 10^{-9}$ and $3.6 \cdot 10^{-9}$ respectively for LHC, LGB and GGB. These values, as well
 435 as the evolutions of the water vapour diffusivity, obtained for the three composites are in
 436 agreement with those found in the literature for other plant-based concretes with values in the
 437 same order of magnitude (10^{-9}) from the works of Collet et al 2012 [24], Rahim et al 2015
 438 [20] or Rahim et al 2016 [21].



439
 440 **Fig. 13.** Evolution of the isothermal water vapour diffusivity as a function of the mass water
 441 content for the three composites

442 *3.1.4 Moisture buffer value (MBV)*

443 Fig. 14 shows the obtained moisture buffer values for the three composites and for all selected
 444 material thickness. The values represent the average of the three measurements for the three

445 samples used. All the values are higher than 2 (g/m² %RH) which allowed to classify the three
 446 new materials in the range of excellent moisture regulators according to the classification of
 447 Nordtest project Rode et al 2005 [25]. The values obtained for the plaster-based composite are
 448 better than those for lime-based composites. As an example, for a thickness of 12 cm, the
 449 MBV of GGB reaches a value of 3.45±0.20 (g/m² %RH) against 2.53±0.18 (g/m² %RH) and
 450 2.66±0.12 (g/m² %RH) respectively for LGB and LHC composites. The standard deviation
 451 remains the same order of magnitude in all cases.

452 The evolution of the MBV with the thickness of the samples was not significant and no trend
 453 is noted. The thicknesses tested are greater than the water vapor penetration depths, which
 454 may be the reason for this result. Arfvidsson 1999 [37] defined the penetration depth (d_{p1%})
 455 of water vapor as the depth at which the amplitude of the variation of the water content is only
 456 1% of the variation of the material surface. For a sinusoidal variation of the moisture content
 457 on the surface of the material, the penetration depth is given by the equation 6

$$d_{p1\%} = 4,61 \sqrt{\frac{D_m t_p}{\pi}} \quad (16)$$

458 D_m : is the water vapour diffusivity in m²/s, for a relative humidity value of 54% RH (average
 459 between 33 %RH and 75% RH).

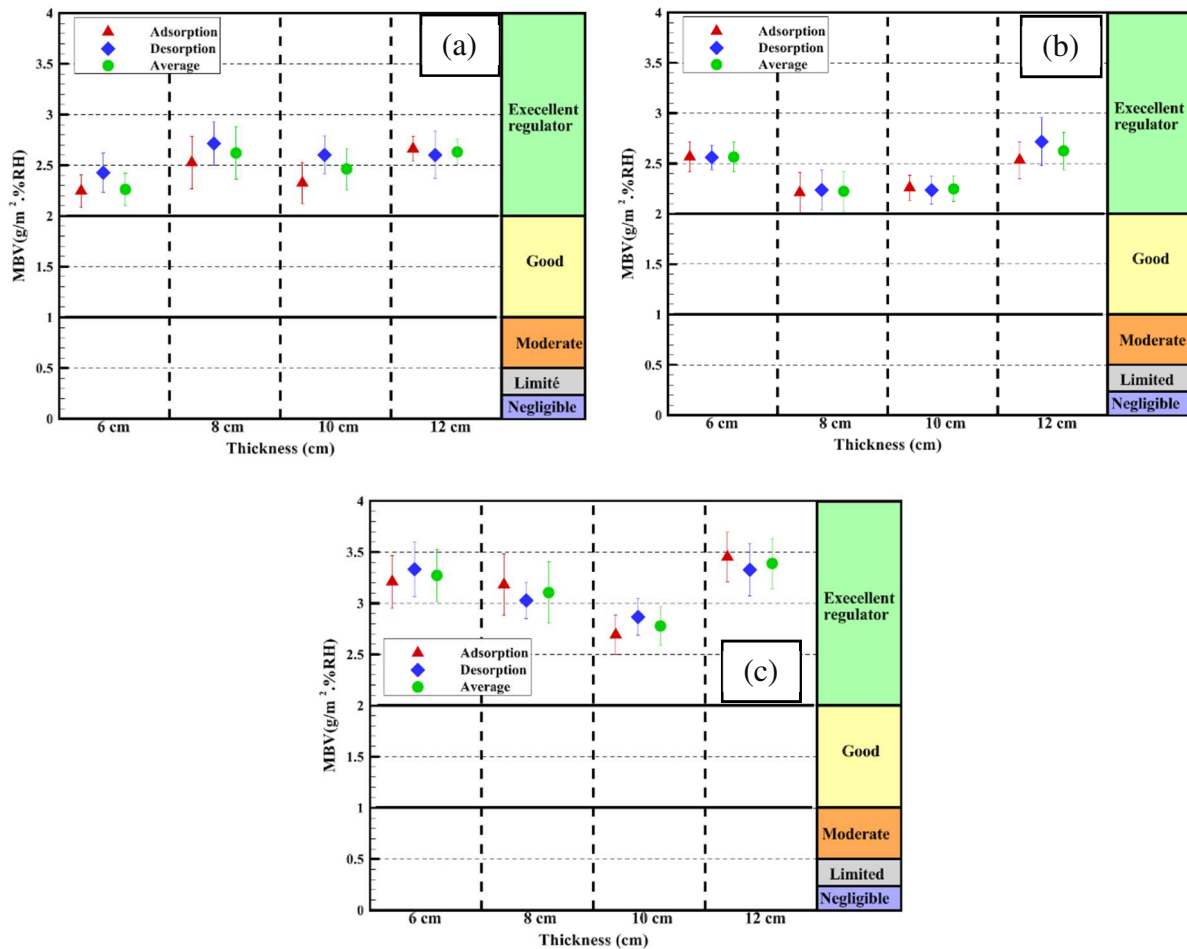
460 t_p : is the time period in (s) (24 h in this study).

461 The results presented in the Tab. 4 confirm that the d_{p1%} values calculated from equation 6 are
 462 lower than the studied thicknesses.

Composite	D _w (m ² /s)	d _{p1%} (cm)
LHC	1.78.10 ⁻⁹	3.24
LGB	1.69.10 ⁻⁹	3.15
GGB	2.38.10 ⁻⁹	3.74

463 **Tab. 4** Penetration depth (d_{p1%}) calculated for the three composites

464



465

466 **Fig. 14.** Moisture buffer values for different thickness of the samples (a) LHC (b) LGB and
 467 (c) GGB

468

469

470

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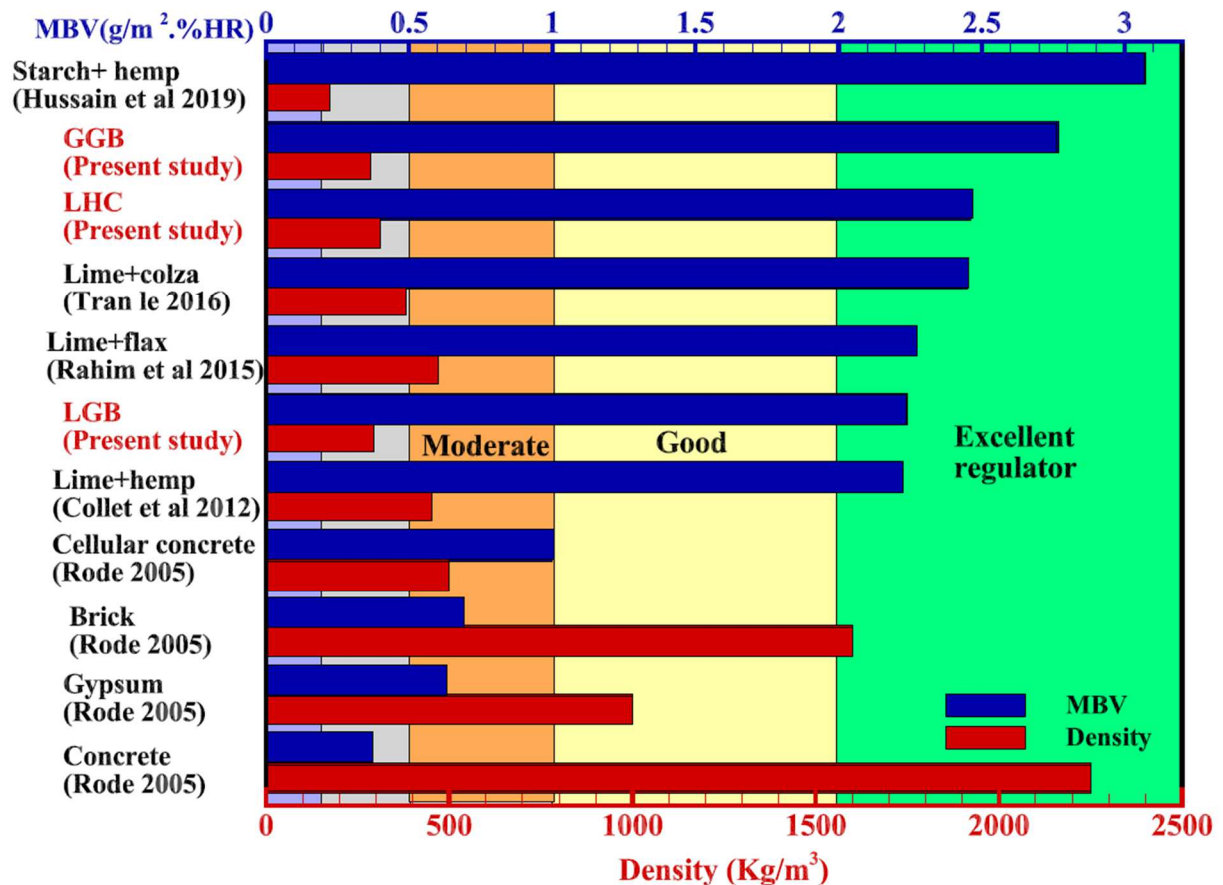
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In Fig. 15, a comparison between the MBV and the density values obtained for the studied biocomposites (with a thickness of 10 cm) and those found in the literature for other composites and other commonly used building materials. The MBV of straw biocomposite is in the same order of magnitude as that of the mentioned biobased materials and it is largely higher than that of conventional building materials as cellular concrete. It is also clear in the fig 15 that the smaller the density, the higher the MBV of the material. However, it should be noted that the most influencing factor on the MBV of the material is the pore distribution and its connectivity.



476

477 **Fig. 15.** MBV and density of the studied composites in comparison with other materials in the
 478 literature

479 **4 Conclusion**

480

481 The objective of this paper was to study the hygric properties of the three new straw biobased
 482 materials with different constituents. The sorption-desorption curves showed a similar pattern
 483 to other porous materials studied in the literature such as hemp concrete. A large hysteresis
 484 was observed in the case of the lime-based composites, explained by the existence of the ink-
 485 bottle shaped pores or the consumption of the water absorbed by the carbonation reaction. The
 486 gypsum-based composite has a different behavior than the lime-based composites, which
 487 confirms that the type of binder strongly influences the moisture storage of the composite
 488 material. All three composites showed high water vapor permeability (low resistance to water
 489 vapor diffusion) due to their high open porosity, which allows them to be qualified as highly

490 hygroscopic materials. In other words, they can contribute to naturally regulate the
491 hygrometry of the interior air of the buildings. The coupling of the results of sorption-
492 desorption and those of the permeability to the water vapor have allowed to deduce the
493 isothermal water vapour diffusivity of the materials. Finally, the moisture buffer value of
494 three composites is measured as a function of the thickness of the materials. The values found
495 made it possible to classify the three composites as excellent moisture regulators without any
496 effect of the thickness as the thicknesses tested were all greater than the penetration depth.
497 The results obtained showed the good hygric performance (permeable to water vapor and
498 moisture regulator) of the studied materials compared to the other classical materials often
499 used in the construction sector.

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501

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504 **6 References**

505

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