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### ► To cite this version:

Jonathan Page, Sofiane Amziane, Moussa Gomina, Chafika Djelal, Fabrice Audonnet. Using linseed oil as flax fibre coating for fibre-reinforced cementitious composite. *Industrial Crops and Products*, 2021, 161, pp.113168. 10.1016/j.indcrop.2020.113168 . hal-03242731

**HAL Id: hal-03242731**

**<https://hal.science/hal-03242731>**

Submitted on 16 Dec 2022

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# 1 Using linseed oil as flax fibre coating for fibre- 2 reinforced cementitious composite

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## 10 ARTICLE INFO

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11 *Keywords:*

12 Flax fibre

13 Cementitious composite

14 Surface treatment

15 Water absorption

16 Workability

17 Mechanical properties

18

19 *Highlights:*

- 20 • Linseed oil coating of flax fibres improves the workability of cementitious composite.
- 21 • The linseed oil coating treatment significantly improves the flexural strength of composites.
- 22 • The thermal insulation properties of the bio-composite are also enhanced by linseed oil
- 23 coating.

## 24 ABSTRACT

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25 In recent years, many studies have focused on the mix-design and characterization of  
26 cementitious composites reinforced with plant fibres. It appears that in the fresh state, the  
27 morphology and the hydrophilic nature of these fibres significantly affect the rheology of the  
28 material. In the hardened state, the mechanical performances of the composites are lower than that  
29 of composites reinforced with standard fibres (glass, synthetic, etc.). These poor mechanical  
30 performances are mainly due to weak neo-formed fibre/matrix interface. Indeed, the high-water  
31 absorption capacity causes swelling/shrinkage of the fibre within the matrix, which subsequently  
32 generates an unbounded interface. In order to address these issues, numerous studies have been  
33 carried out over the past few years on compatibilisation treatments applied to plant fibres. In most  
34 cases, these are chemical treatments to leach the fibre, which often have a significant environmental  
35 impact. This work examines the influence of a coating treatment with linseed oil applied to flax fibres  
36 on the properties of cementitious composites. The first part of this study deals with the optimisation  
37 of this treatment. The second part focuses on the effect of this treatment on the properties of the  
38 composite in the fresh and hardened states. The results showed a noticeable improvement in the  
39 workability of the treated fibre composites. The linseed oil coating treatment can also significantly  
40 improve the flexural strength of the composite, depending on the mix-design. Flax fibres and linseed  
41 oil can also slightly enhance the thermal insulation properties of the material.

## 42 **1. Introduction**

43 For decades, fibres have been used in cementitious materials to improve the tensile strength and  
44 post-cracking behaviour [1–3]. The most commonly used in cementitious composites are steel fibres,  
45 glass fibres and polypropylene fibres. However, all these reinforcement materials have the  
46 disadvantage of being derived from non-renewable resources. Economic issues related to the rising  
47 costs of fossil resources, their increasing scarcity, and environmental impacts inherent to their  
48 production therefore lead to explore other raw materials. This is how plant fibres are viewed as a  
49 solution for the future of the construction industry [4–7]. Among the potential plant fibres, flax  
50 stands out because of its excellent mechanical properties and low density [8,9].

51 Many research projects dealt with the incorporation of bio-based fibres into cementitious  
52 materials. They highlighted that in the fresh state, flax fibres affect the rheology of the mixture [10–  
53 13]. These rheological disturbances are mainly attributed to the high demand for water due to the  
54 high-water absorption capacity of flax fibres (up to 150 % of dry fibre mass) and their high specific  
55 surface area [14]. Numerous studies have shown that plant fibres have a considerable influence on  
56 the rheological behaviour of mortars and concretes [10,14]. The use of a superplasticizer and a  
57 viscosity-modifying admixture significantly reduces these disturbances [15,16]. In the hardened  
58 state, composites reinforced with plant fibres show an improvement of the flexural toughness or  
59 strength and increased ductility [14,17,18]. The transition from a brittle matrix to a ductile fibre-  
60 reinforced composite having controlled post-peak behaviour is noted by several authors [14,19,20].  
61 However, this change in behaviour is not always accompanied by an improvement in flexural  
62 strength [21].

63 Despite the advantageous physical properties obtained for cementitious composites reinforced  
64 with plant fibres, many studies highlighted problems at the fibre-matrix interface, sometimes  
65 manifested by a gap between these two components [22–24]. This gap is often attributed to swelling  
66 of the fibres during the concrete mixing process (in the fresh state) and consecutive shrinkage during  
67 the curing of the material (in the hardened state). Moreover, these studies have shown that the  
68 hydrophilicity of plant fibres is responsible for these volume variations, which could cause this poor  
69 fibre/matrix interface [25].

70 Water absorption by plant fibres is therefore an important issue limiting their use in composite  
71 materials. Surface treatment may be a solution to limit or eliminate water absorption by these fibres  
72 [26,27]. Given the problems encountered when using plant particles with a cementitious matrix,  
73 many studies have been conducted to modify some of their characteristics. Most of the treatments  
74 tested had the objective of limiting the hydrophilic behaviour of the fibres while others aimed to  
75 modify their surface composition. Chemical treatments are the most common experienced. They  
76 consist of prolonged immersion of the fibres in water-based solution [28–31] or in an alkaline  
77 solution (sodium or calcium hydroxide, etc.) [27,31–33] or esterification [34,35]. Chemical  
78 treatments generally dissolve potentially troublesome plant extractibles and modify the chemical  
79 composition of the fibre surface. In addition, these treatments have significant environmental  
80 impacts (use of strongly acidic or basic chemicals, water consumption, need for temperature cure,  
81 etc.) [36]. Thermal treatments are regularly applied on wood; they have therefore been experienced  
82 on plant fibres [37,38]. In much the same way as chemical treatments, thermal treatments eliminate  
83 hemicelluloses and other extractibles, and limit the volume variations of the fibre. However, this  
84 treatment remains difficult to master and requires heating for a long period. A mineral coating can be  
85 applied to the lignocellulosic fibres or aggregates using a hydraulic binder (cement, lime, etc.)  
86 [14,39–41]. Mineral coatings seem to reduce the hydrophilicity of the fibres, they chemically isolate  
87 the fibre from the matrix and limit the volume variations. However, they cause a considerable mass  
88 increase. Moreover, the long-term mechanical performances are degraded by the mineralisation of  
89 the fibres in contact with the binder used for the coating [33,42]. Still with the aim of limiting the  
90 plant fibre or aggregate's water absorption, another solution is to apply an organic coating  
91 (polymer): paraffin wax [43], acrylic polymer [28,44,45], or linseed oil [27,35,40]. These treatments  
92 significantly reduce the water absorption of the aggregates and improve the composite properties.  
93 Nevertheless, the surface created could modify the adhesion with the matrix. But, depending on the  
94 nature of the polymer, the environmental impact may also be significant. From this point of view,  
95 linseed oil appears as the most eco-friendly polymer.

96 Linseed oil (LO) is a drying oil susceptible to auto-oxidation and polymerization resulting in tough  
97 elastic films upon exposure to air [46]. Linseed oil is composed of three fatty acids with long

98 unsaturated aliphatic chains (oleic, linoleic and linolenic acids) and saturated chains (stearic and  
99 palmitic acids). The polymerization reaction is a three-step radical chain reaction (induction,  
100 propagation and termination). Both linoleic and linolenic acids rapidly oxidize, since their  
101 unsaturated bonds are close to one another. The reaction rate is improved with an increase in  
102 temperature and depends on the linoleic fatty acid to linolenic fatty acid ratio [47]. Moreover, the  
103 heat liberated during the oxidation–polymerization steps improves even more the reaction kinetics,  
104 eventually leading to spontaneous combustion of the linseed oil [48]. For this reason, it is  
105 recommended not to use linseed oil at a temperature above 60°C.

106 From the literature review, linseed oil stands out because of its bio-based origin and its efficiency  
107 in reducing the water absorption of plant aggregates. Thus, this work investigates a way to improve  
108 the properties of flax-fibre-reinforced mortar using linseed oil as coating treatment applied to flax  
109 fibres. The first part of this study concerns the optimisation of the linseed oil-to-fibre ratio in order to  
110 avoid agglomeration of the fibres or overconsumption of oil. The second part focuses on the effects of  
111 this treatment on the bio-composite properties in the fresh and hardened states.

## 112 **2. Experimental program**

### 113 **2.1. Materials**

114 The hydraulic binder was developed from two commercial powders: CEM I cement (67% in  
115 mass), metakaolin (33% in mass). The cement used is a CEM I 52.5 R ordinary Portland cement  
116 (OPC) in accordance with EN 197-1 standard. The compressive strength on standardized mortar at  
117 28 days is 72.0 MPa, the Blaine fineness is 4200 cm<sup>2</sup>.g<sup>-1</sup>, and the density about 3.13 g.cm<sup>-3</sup>. Metakaolin  
118 was also used as a mineral addition of Portland cement to develop the binder. The metakaolin used is  
119 in accordance with NF P18-513 standard. This powder has a Blaine fineness of 4270 cm<sup>2</sup>.g<sup>-1</sup>, and the  
120 density about 2.57 g.cm<sup>-3</sup>. In addition to cement and metakaolin, a highly pure limestone was used  
121 (total carbonates equal to 98.7%), in accordance with the NF P 18-508 standard. The Blaine fineness  
122 of the limestone filler is 5440 cm<sup>2</sup>.g<sup>-1</sup>, and its density is 2.70 g.cm<sup>-3</sup>. These mineral additions were

123 used to reduce the negative effect of the alkaline medium produced by OPC, and the crystallization of  
124 portlandite into the fibres [49–51].

125 In addition, chemical admixtures were used for mortars reinforced with flax fibres. A high range  
126 water reducer superplasticizer based on polycarboxylates was added with a ratio of 0.30% relative  
127 to the mass of binder (dry extract) to improve workability in the fresh state. In addition, a viscosity-  
128 modifying agent (VMA) based on a bio-polymer of high-molecular weight was added to the mortars  
129 with a ratio of 0.02% relative to the mass of binder (dry extract) to prevent segregation between the  
130 paste and the aggregates.

131 An alluvial sand with a grain size of 0/2 mm was used. This sand has a specific gravity of 2.66  
132 g.cm<sup>-3</sup>, and an absorption coefficient of 0.15%. The flax fibres used were harvested in Normandy  
133 (France) in 2014, cut at a constant length of 12 mm and provided by Vandecandelaere Company  
134 (Depestele Group, Caen – France), which is specialized in flax farming and scutching. These raw flax  
135 fibres have a real density of 1.52 g.cm<sup>-3</sup>, and a mean diameter of (14.7±3.0) μm. The water absorption  
136 of the flax fibres after immersion for 24 h is (132±5) %. This value was used to correct the effective  
137 water content for the mix design of the mortars. A flax fiber can be described as a 10–20 μm wide  
138 cylindrical composite with concentric cell walls which differ in terms of chemical composition and  
139 morphology. The polymers involved in the composition of fibers are cellulose (64-74 %),  
140 hemicelluloses (11-21 %), lignin (2-3 %) and pectin (1-3 %) [52–55]. The tensile strength of flax  
141 fibres is (1254 ± 456) MPa. The mechanical tests were performed on 50 samples of elementary  
142 fibers, using the protocol described in the French standard NF T25-501-2.

## 143 **2.2. Linseed oil treatment**

144 Linseed oil (LO) is a drying oil susceptible to auto-oxidation and polymerization resulting in tough  
145 elastic films upon exposure to air [46]. Linseed oil is composed of three fatty acids with long  
146 unsaturated aliphatic chains (oleic, linoleic and linolenic acids) and saturated chains (stearic and  
147 palmitic acids). The polymerization reaction is a three-step radical chain reaction (induction,  
148 propagation and termination). Both linoleate and linolenate acids rapidly oxidize, since their

149 unsaturated bonds are close to one another. The reaction rate is improved with an increase in  
150 temperature and depends on the linoleic fatty acid to linolenic fatty acid ratio [47]. Moreover, the  
151 heat liberated during the oxidation–polymerization steps improves even more the reaction kinetics,  
152 eventually leading to spontaneous combustion of the linseed oil [48]. For this reason, it is  
153 recommended not to use linseed oil at a temperature above 60°C.

154 The linseed oil used in this work is a boiled linseed oil with an accelerated oxidation, supplied by  
155 Natura® laboratories. It was extracted from farmed flax, harvested and produced in Normandy. It  
156 was obtained by cold extraction without any solvent, filtered at 1 µm, and then heated at 150 °C. This  
157 linseed oil has a density of 0.93.

158 For carrying out the treatment, the flax fibres were previously dried at 50±1°C and then  
159 introduced into a planetary mixer. The mixer is then started at a speed of 30 rpm and after 30 s the  
160 linseed oil is introduced for a period of 2 min 30 s. Different oil / fibre mass ratio were experienced:  
161 0.25, 0.5, 1.0, 1.5 and 2.0. Mixing was maintained for 2 min 30 s more to ensure a homogeneous fibre  
162 coating. The fibres thus treated are placed in a ventilated oven at 50±0.1°C for 14 days.

### 163 ***2.3. Preparation of cement composite***

164 The respective proportions of the mortar's components are detailed in Table 1. For all  
165 compositions, the effective water on binder (CEM I cement + Metakaolin) ratio was set at 0.50 and  
166 the sand on powders (binder + limestone) ratio was set at 1.0.

167 Flax fibres were added with different proportions: 1.0, 2.0 and 3.0 %vol. to the total batch volume.  
168 An additional amount of water was added to take into account the water absorption and flax fibres.  
169 These quantities are given in Table 1.

170 After mixing, the mortars were cast in two layers into prismatic moulds (40x40x160 mm). To  
171 expel the air from the mixture, the moulds were subjected to 60 hits on a vibrating table. The test  
172 pieces were demoulded 24 h after manufacturing, and then placed in a climatic chamber at (20±1) °C  
173 and (65±5) %RH.

174 **Table 1.** Composition of cement composites

Mortars	Ref. Mortar	Raw Fibres Mortars				Linseed Oil (ratio = 0.25) treated Fibres Mortars			Linseed Oil (ratio = 0.5) treated Fibres Mortars		
		Fibre content (%)	0 %	1 %	2 %	3 %	1 %	2 %	3 %	1 %	2 %
Sand 0/2 (g)	800.0	800.0	800.0	800.0	800.0	800.0	800.0	800.0	800.0	800.0	800.0
Binder (g)	600.0	600.0	600.0	600.0	600.0	600.0	600.0	600.0	600.0	600.0	600.0
Limestone (g)	200.0	200.0	200.0	200.0	200.0	200.0	200.0	200.0	200.0	200.0	200.0
Eff. Water (g)	300.0	300.0	300.0	300.0	300.0	300.0	300.0	300.0	300.0	300.0	300.0
Flax fibres (g)	0.0	13.4	26.8	40.2	12.3	24.6	36.9	12.0	24.0	36.0	
Plasticizer (g)	0.0	1.8	1.8	1.8	1.8	1.8	1.8	1.8	1.8	1.8	
VMA (g)	0.0	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	
Add. water (g)	0.0	17.4	34.8	52.3	10.3	20.7	31.0	8.8	17.6	26.4	
Density (g.cm <sup>-3</sup> )	2.158	2.130	2.103	2.077	2.137	2.117	2.098	2.139	2.120	2.103	

175

176 **2.4. Testing methods for flax fibre**

177 *2.4.1. Real density*

178 The real density of raw and treated flax fibres was measured using a helium pycnometer, on an  
 179 oven-dried fibres sample of about 1.00 gram; six samples were tested. Experiments were performed  
 180 on a Micromeritics AccuPyc II 1340.

181 *2.4.2. Water absorption*

182 Due to their nature and chemical composition, flax fibres are very susceptible to water absorption  
 183 [56]. The flax fibres were heated to (50±0.1) °C in a ventilated oven for 48 h prior to the test. The  
 184 water absorption kinetic was obtained by placing 3.00 g of flax fibres in a microperforated bag, then  
 185 immersing the bag in water for different times (from 1 min to 72 h). The experiment was repeated  
 186 six times for each duration of soaking. After immersion, the bags were placed in a centrifuge  
 187 operating at 500 rpm for 30 s to remove the excess water on the surface of the fibres.

188 The water absorption capacity of these fibres was determined gravimetrically using Eq. 1. This  
189 testing method is derived from the recommendations of the RILEM Technical Committee 236 [57].  
190 The experiment was repeated six times. The result is expressed by:

$$WA(t) = \frac{m(t) - m_0}{m_0} \times 100 \quad \text{Eq. 1}$$

191 with  $WA(t)$  being the water absorption ratio at time  $t$ ,  $m(t)$  the soaked fibres' mass at time  $t$ , and  $m_0$   
192 the initial dry mass.

### 193 2.4.3. ATR-FTIR

194 To obtain information on the surface chemistry of flax fibres, attenuated total reflectance (ATR)  
195 was used in conjunction with infrared spectroscopy (FTIR). This technique enables direct analysis of  
196 particles to be carried out. It gives information on chemical groups and bonding by studying  
197 vibratory frequencies in the mid-infrared domain (4000–400  $\text{cm}^{-1}$ ). The experiments were  
198 performed on a Nicolet Magna IR 600. All background and sample acquisitions were acquired in the  
199 transmission mode at 4  $\text{cm}^{-1}$  resolution, with accumulation of 50 scans, at room temperature.

## 200 2.5. Testing methods for mortar properties

### 201 2.5.1. Workability

202 The workability of the different mortars was assessed with the flow table, in accordance with EN  
203 1015-3. The flow value is measured by the mean diameter (in two perpendicular directions) of a  
204 sample of the fresh mortar which was placed on the flow table tamper by means of a truncated  
205 conical mould, and given 15 vertical hits by raising the flow table and allowing it to fall freely from a  
206 height of 10 mm.

### 207 2.5.2. Air content

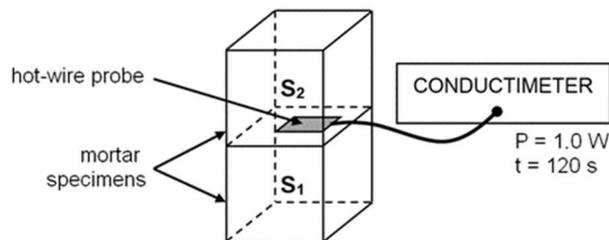
208 The air content of the fresh mixes was evaluated with a 1 L pressure gauge according to the EN  
209 1015-7 standard.

210 *2.5.3. Compressive and 3-point bending tests*

211 The mechanical properties of the mortars were determined by implementing compressive and  
212 three-point bending tests in accordance with the EN 196-1 standard after 28 days of curing. For each  
213 mix, three specimens were tested. The compression test was implemented at a loading rate of  $2.4 \text{ kN.s}^{-1}$   
214 and the bending test at  $50 \text{ N.s}^{-1}$ .

215 *2.5.4. Thermal conductivity*

216 The thermal conductivity of the mortar specimens was measured with a conductimeter using the  
217 transient hot-wire method (EN ISO 8894). A heat flow by Joule effect is generated, and the  
218 temperature variation over time was measured using a thermocouple. The heating element was  
219 associated with the thermocouple in the hot-wire probe. The rise in temperature measured by the  
220 sensor is limited to  $20^\circ\text{C}$  and the measuring time is dependent on the material, here fixed at 120 s.  
221 Measurements were taken by positioning the thermal probe between two specimens of the same mix  
222 (Fig. 1) and were conducted three times after 28 days of curing. The specimens were oven-dried  
223 before thermal measurements.



224

225 **Fig. 1.** Thermal conductivity measurement with the hot-wire probe.

226 *2.5.5. Water accessible porosity*

227 The water accessible porosity of the hardened mortar specimens was calculated through vacuum  
228 saturation and with a hydrostatic balance according to the NF P18-459 standard at 28 days curing.  
229 For each mix, three specimens were tested. The saturated surface dry state of the specimens was  
230 obtained through vacuum saturation. Initially, the specimens were placed in a vacuum chamber in  
231 the presence of silica gel in order to decrease the contact angle between the water and the pores and

232 dry the specimens. After 4 h, the water was introduced into the chamber to immerse the specimens,  
233 and the vacuum was maintained for an additional 44 h. The porosity was obtained from the mass of  
234 the saturated surface dry specimens measured in air ( $m_{air}$ ) and in water ( $m_{water}$ ), and oven-dried at  
235 50°C ( $m_{dry}$ ), using Eq. 2.

$$Porosity = \frac{m_{air} - m_{dry}}{m_{air} - m_{water}} \times 100 \quad \text{Eq. 2}$$

### 236 2.5.6. Water absorption by capillary action

237 The porosity and permeability of materials induce water to rise through capillary networks. This  
238 capillary rise leads to the deposition of hygroscopic salts in the material pores and causes the  
239 proliferation of fungi and molds. The capillary water absorption of a material defines the water  
240 quantity that can be absorbed by the specimen when one of its sides is in prolonged contact with  
241 water. This coefficient of capillary water absorption  $C_w$  ( $\text{kg}\cdot\text{m}^{-2}\cdot\text{min}^{-0.5}$ ) was determined as described  
242 in the EN 1015-18 standard. The procedure given in this standard for determining this coefficient  
243 involves several stages. First, samples were oven-dried until a constant dry mass was obtained. The  
244 samples were then immersed in water to a depth varying from 5 to 10 mm. Two masses  $M_{10}$  and  $M_{90}$   
245 were then recorded, respectively, at 10 and 90 min after water immersion. The capillary coefficient  
246 was then calculated by means of Eq. 3. Three measurements were carried out for each mix, after 28  
247 days of curing.

$$C_w = 0.1 \times (M_{90} - M_{10}) \quad \text{Eq. 3}$$

## 248 3. Results and discussion

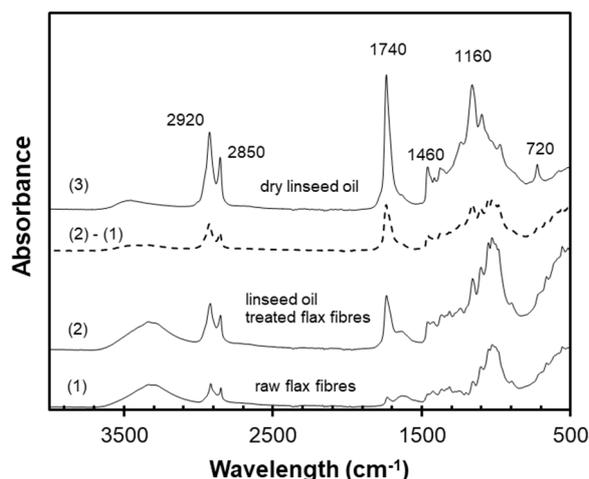
### 249 3.1. Optimisation of linseed oil treatment

250 Previous studies using linseed oil for the treatment of plant particles made use of variable  
251 oil/particle ratios. For example, Monreal *et al.* chose ratios of 0.33, 0.5 and 1 [40]. Nozahic *et al.*  
252 adopted an oil / sunflower aggregates ratio of 0.5 [27] while Khazma chose oil / flax shives ratios of  
253 1, 2 and 3 [35], and Sellami used an oil/fibre ratio of 0.78 [30]. A wide range of ratios has therefore

254 been explored, depending on the type of the plant particles. Thus, a wide range of ratios has been  
255 tested, depending on the type of the plant particles. Therefore, the literature review was not useful in  
256 determining an adequate ratio for the treatment of flax fibres. Hence, in order to optimize the mass  
257 quantity of linseed oil relative to the flax fibres, five different LO/fibre ratios were investigated: 0.25,  
258 0.5, 1, 1.5 and 2.

### 259 *3.1.1. Observation of the polymerized linseed oil treatment*

260 The absorbance of raw flax fibres, LO-treated flax fibres, and a thin film of polymerized linseed oil  
261 by means of ATR-FTIR are shown in Fig. 2. Table 2 summarizes the major bands observed in the  
262 FTIR spectrum of flax fibres and linseed oil, and their attachments to chemical group molecules. Flax  
263 fibre is rich in polysaccharides, with the -OH hydroxyl group stretching leading to a large peak  
264 between 3125 and 3600  $\text{cm}^{-1}$  [31,58]. Out-of-plane cellulose-water linkage between 700 and 650  $\text{cm}^{-1}$   
265 and cellulose  $\beta$ -links (900  $\text{cm}^{-1}$ ) are also noted for raw flax fibres [58,59]. They are characteristic of  
266 plant fibres. Hydroxyl groups are able to form hydrogen bonds with water, which is responsible for  
267 the significant water absorption capacity of plant fibres. Stretching vibrations of the  $\text{CH}_2$  methylene  
268 group (2920 and 2850  $\text{cm}^{-1}$ ) on the surface are also identified. This is characteristic of the presence  
269 of waxes and fats on the flax fibre surface. Peak characteristics from lignocellulosic compounds retain  
270 the same absorbance level as raw surfaces. The main difference is a major increase in  $\text{CH}_2$  methylene  
271 group stretching peaks. It indicates waxy or oily deposition on the surface by polymerized linseed oil.  
272 Linseed oil oxidation can be verified from the triacylglycerol ester-bonding peak at 1740  $\text{cm}^{-1}$  and  
273 from the triglycerides ester linkage peak at 1160  $\text{cm}^{-1}$  [60]. The dotted spectrum corresponds to the  
274 spectrum of fibres treated with linseed oil (2), subtracted from the spectrum of raw fibres (1). There  
275 is a very close correspondence with the spectrum of polymerized linseed oil (3). The FTIR spectrum  
276 information does not enables us to draw conclusions about the homogeneity of the surface coating.



277

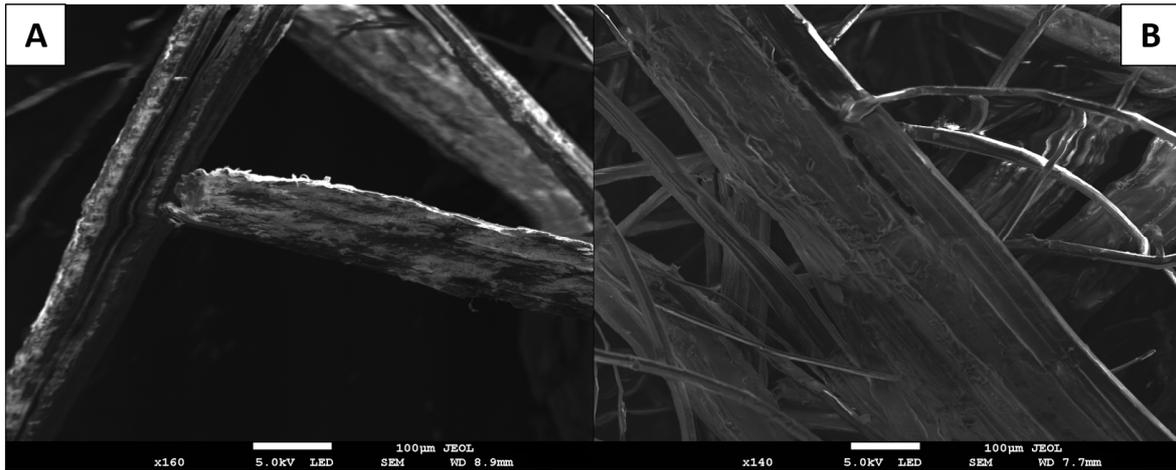
278 **Fig. 2.** ATR-FTIR analysis of raw and linseed oil treated flax fibres, and of polymerized linseed oil film.

279 **Table 2.** Main infrared absorption peaks and band for flax fibre and linseed oil between 4000 and 400  $\text{cm}^{-1}$

Wavelength ( $\text{cm}^{-1}$ )	Bonding	Associated product	Reference
3600–3000	O–H	Polysaccharides, cellulose and hemicelluloses, water	[31,58,60]
2920, 2850	C–H ( $\text{CH}_2$ )	Waxes, paraffin, oils (esterified fat acids)	[31,60–62]
1740	C=O	Dry linseed oil (ester bond in triacylglycerol)	[31,60–62]
1670–1630	–OH	Water (crystalline cellulose)	[31,54]
1460	C–H ( $\text{CH}_2$ )	Paraffin (alkanes)	[60,62]
1380	CH	Cellulose and hemicelluloses (bending)	[58]
1160	C–O	Drying linseed oil (triglycerides and aliphatic),	[60]
1120–900	C–O–C	Cellulose (bond stretching)	[58]
900	$\beta$ -links	Cellulose	[58,59]
720	$(\text{CH}_2)_n$	Paraffin (alkanes)	[60,61]
700–650	C–OH	Out-of-plane cellulose-water linkage	[58,59]

280

281 Environmental scanning electron microscope (SEM) observations of raw and LO-treated flax  
 282 fibres (ratio = 0.5) are given in Fig. 3. Fig. 3a shows several individual raw fibres naturally grouped in  
 283 bundles. On the Fig. 3b, the polymer film formed by the deposition of linseed oil gives a glossy  
 284 appearance and seems to coat all the fibres within the bundles. This will decrease the specific surface  
 285 area of the fibres and limit their separation into single fibres.



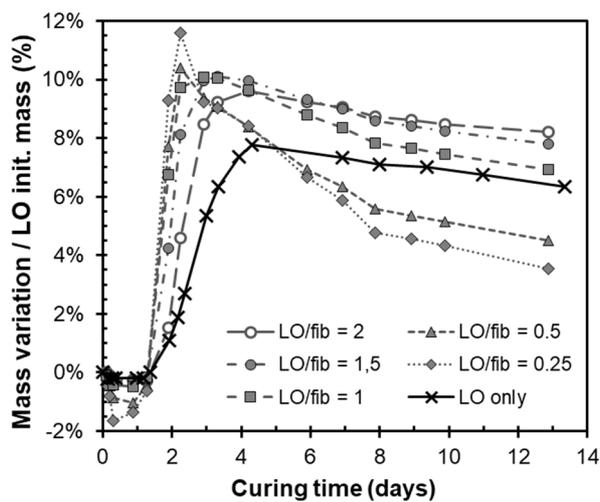
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287 **Fig. 3.** SEM observations of raw flax fibres (a) and flax fibres treated with linseed oil (ratio = 0.5) (b).

288

289 *3.1.2. Mass monitoring*

290 After treatment, the flax fibres were placed in an oven at 50 °C for 2 weeks. A portion of the  
 291 treated fibres was placed into metal cups so as to track the evolution of the mass of the samples over  
 292 time. Fig. 4 shows the mass gains of the treated flax fibres relative to the initial mass of oil, as a  
 293 function of the time after treatment, for each LO/fibre ratio.



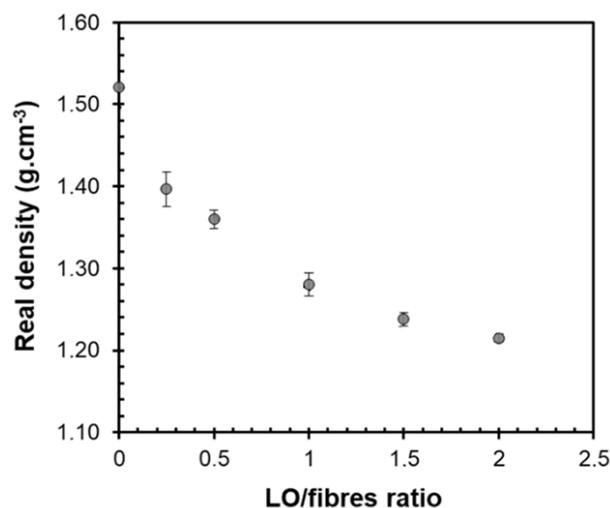
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295 **Fig. 4.** Mass monitoring of fibres treated with linseed oil during the curing time.

296 For all samples, linseed oil mass increased between 7 % and 14 %. The loss of mass in the first day  
297 at the very beginning of the treatment corresponds to the departure of the moisture naturally  
298 present in the fibres. Low LO/fibres ratio led to a significant and faster oil mass increase. This mass  
299 increase is the result of the oxidation of linseed oil. The polymerization of this boiled linseed oil  
300 seems to occur between 2 and 3 days after application of the treatment. This is in keeping with the  
301 results obtained by Lazko *et al.* who observed a mass increase of linseed oil of between 10 and 15%  
302 [63].

### 303 3.1.3. Density

304 The linseed oil coating treatment of fibres reduces the overall density of the treated fibres (Fig. 5).  
305 The specific density of the raw flax fibres is  $(1.52 \pm 0.01) \text{ g.cm}^{-3}$ . As a result of the treatment, the density  
306 of fibre + LO coating is  $(1.21 \pm 0.01) \text{ g.cm}^{-3}$  for a LO/fibres ratio of 2. This is explained by the low density  
307 of the linseed oil, equal to  $0.93 \text{ g.cm}^{-3}$ . Thus, when the LO/fibres ratio increase, the proportion of oil is  
308 greater, resulting in a decrease in the treated fibre densities. Also note the very small standard  
309 deviations, indicated by the error bars in Fig. 5. This suggests good homogeneity of the application of  
310 the polymeric coating.



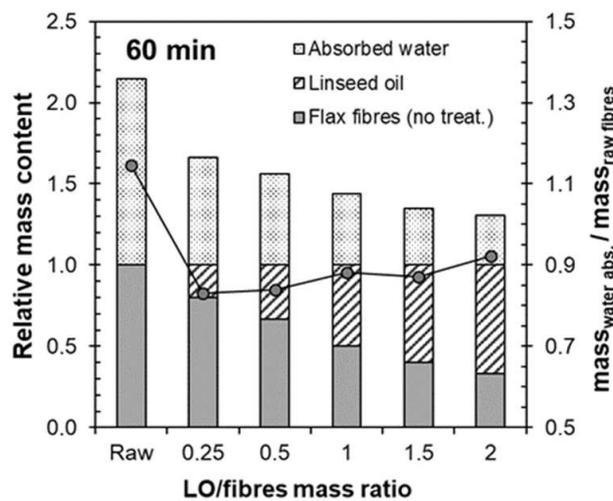
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**Fig. 5.** Density of the coated fibres as a function of the LO/fibre ratio.

313 3.1.4. Water absorption

314 The main purpose of the linseed oil treatment is to reduce the water absorption of the flax fibres.  
315 The linseed oil (LO) / fibres ratio will therefore be optimised mainly according to this criterion.  
316 Water absorption measurements were therefore carried out on these treated fibres for 60 min of  
317 immersion. The results of these water absorption tests are presented in terms of mass ratios, based  
318 on the mass of flax fibres (Fig. 6).



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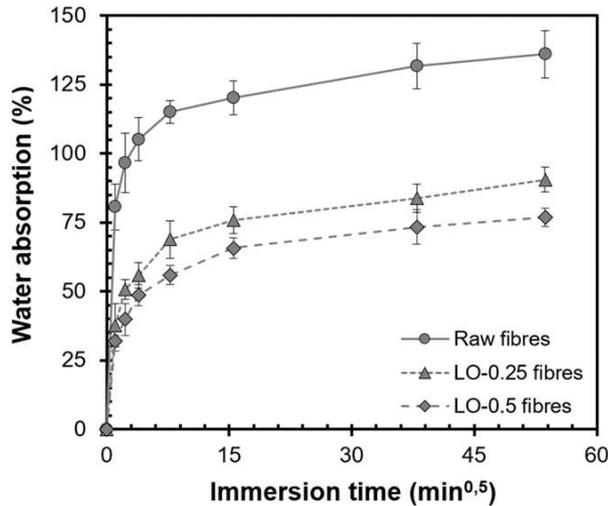
320 **Fig. 6.** Absorbed water mass relative to the mass of treated fibres, according to the LO/fibres ratio after soaking  
321 for 60 min.

322 This type of representation was chosen to highlight the absorption results with respect to the  
323 linseed oil present on the treated fibres. Indeed, it is considered that linseed oil does not absorb  
324 water; it is therefore obvious that the absorption of the treated flax fibres is reduced in comparison  
325 with raw fibres. It is then necessary to calculate the ratio of the mass of water absorbed to the mass  
326 of raw fibres ( $\text{mass}_{\text{water,abs.}}/\text{mass}_{\text{raw,fibres}}$ ), without the linseed oil mass. This allow to assess whether  
327 linseed oil is really effective in reducing this absorption. The mass of linseed oil in the treated fibres  
328 is known from the masses of oil and fibers introduced into the mixer. This curve (in black on the  
329 graph) is used to determine the optimal ratio corresponding to the minimum mass of linseed oil.  
330 Logically, the raw fibres are those that absorb the largest amount of water:  $115\pm 4\%$  after soaking for

331 60 min. It is also noteworthy that the higher the LO/fibres ratio, the lower is the amount of absorbed  
332 water.

333 However, it is important to compare the mass of absorbed water and the amount of raw fibre  
334 present in the sample to identify the most effective LO/fibres ratio. Therefore, the curve (relative to  
335 the right Y-axis in Fig. 4) shows the mass of absorbed water relative to the mass of raw fibres. Thus,  
336 the LO/fibres ratios of 0.25 and 0.5 seem to be the most effective in decreasing the water absorption.  
337 These two ratios will therefore be employed for the rest of this study.

338 The water absorption kinetics of raw and treated flax fibres are presented in Fig. 7. Two distinct  
339 stages are evidenced for all fibres. First, a rapid mass increase of the fibres is observed for a short  
340 time. At the beginning of the water sorption, this process is driven by capillary forces. In less than 5  
341 min, the raw fibres absorbed about 50 % of their maximum water retention capacity. A second  
342 absorption stage shows a less steep raise toward the saturation level. In the second stage, the  
343 absorption is driven by water diffusion. The highly hydrophilicity of plant fibres, due to the nature of  
344 their components, explains the strong dependency of their morphology and their physical properties  
345 on the relative humidity [64,65]. This essential characteristic also gives rise to fierce competition  
346 between the flax fibres and binder in terms of water demand, which starts with concrete mixing.  
347 Thus, raw flax fibres can absorb water up to 150% of their dry mass because of their hydrophilicity  
348 [28]. As Fig. 7 shows, the linseed oil treatments (0.25 and 0.5 LO/fibres ratios) greatly reduce water  
349 absorption by the fibres. The slopes at very short times confirm that the hydrophilicity of fibres is  
350 reduced by the linseed oil treatment, which is hydrophobic. The maximum water absorption after  
351 immersion for 24 h WA(24) is reduced to  $(83\pm 5)$  % of the dry mass for the 0.25 LO/fibres ratio and  
352  $(73\pm 6)$  % for fibres coated with linseed oil with a ratio of 0.5.



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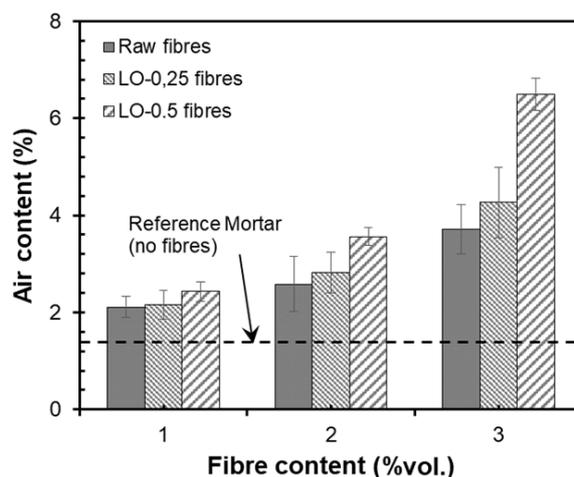
Fig. 7. Water absorption of raw and treated flax fibres.

355 **3.2. Comparison of the properties of cement composites reinforced with raw and linseed oil**  
 356 **treated flax fibres**

357 Linseed oil treatment significantly decreased the water absorption of the fibre, especially for 0.25  
 358 and 0.5 ratios. Mortars incorporating raw and treated fibres (with these two ratios) were  
 359 manufactured and characterised in both fresh and hardened states. Three volume ratios of flax fibres  
 360 were used: 1.0, 2.0 and 3.0 %vol. (Table 1).

361 **3.2.1. Air content**

362 The incorporation of flax fibres into mortar mixes increases the air content due to entrainment of  
 363 additional air (Fig. 8). The reference mortar has the lowest air content, equal to  $(1.7 \pm 0.1)$  %. As  
 364 expected, increasing the fibre content increases the amount of trapped air, whichever the fibre state  
 365 (raw or treated). This increase in air content was also underlined by other authors [6]. It is mainly  
 366 assigned to the low ability of fibres to compact. Indeed, the fibres have a significant impact on the  
 367 granular compactness of concrete mixtures [66].



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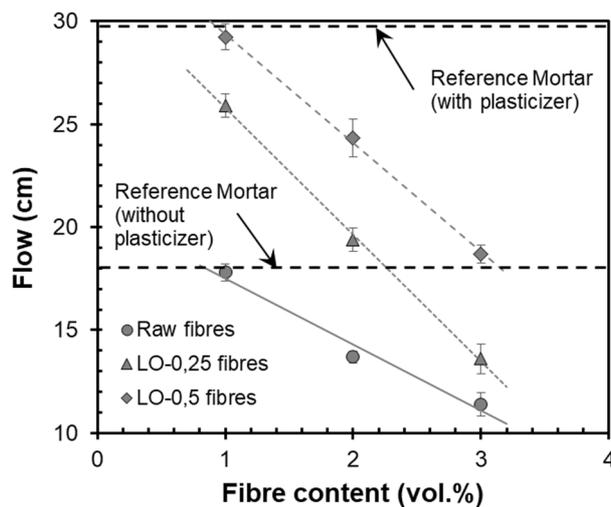
**Fig. 8.** Air content of fresh mortars.

370 For mortars reinforced with 1.0 %vol. of fibres, the linseed oil treatment does not seem to have a  
 371 significant impact on the air content, which is between  $(2.1 \pm 0.2)$  % and  $(2.4 \pm 0.2)$  %, depending on  
 372 the fibre state. For mixtures with 2.0 %vol. of fibres, the linseed oil treatment (especially with the  
 373 0.5-LO/fibres ratio) results in a noticeable increase in air content compared to raw fibres. The effect  
 374 of the linseed oil treatment on air content is even more pronounced for mixtures containing 3.0 %vol.  
 375 fibres. The air proportion represents  $(3.7 \pm 0.5)$  %,  $(4.3 \pm 0.7)$  % and  $(6.5 \pm 0.3)$  % with raw fibres, and  
 376 with 0.25 and 0.5 LO/fibres ratio, respectively. Thus, the linseed oil treatment increases the amount  
 377 of entrapped air in fresh mixtures. A previous study had also shown that linseed oil treatment has the  
 378 effect of increasing the air proportion in plant fibre reinforced cementitious composites [14]. This  
 379 could be explained by the coating characteristics specific to linseed oil. Indeed, linseed oil sometimes  
 380 seems to coat individual flax fibres and sometimes bundles of fibres. In the latter case, air could be  
 381 entrapped in fibre bundles coated with linseed oil. Once these fibres are mixed with the mortar, the  
 382 air remains entrapped, increasing the air content of the material.

### 383 3.2.2. Consistency of fresh mixes

384 Fig. 9 shows the flow diameters measured for the flow table test. Firstly, the reference mortar has  
 385 a plastic consistency, with a flow of  $(18.0 \pm 0.4)$  cm. Note that this mortar does not contain a  
 386 superplasticizer. In contrast, the mortars with flax fibres all contain a superplasticizer and a

387 viscosity-modifying admixture to overcome the problems encountered with the use of flax fibres in  
 388 cementitious composites [66]. If chemical admixtures are added to the reference mortar mixture (in  
 389 the same proportion that flax-fibre reinforced mortars), the consistency is very fluid, with a flow  
 390 diameter of  $(29.8 \pm 0.2)$  cm. Moreover, despite the use of admixtures, the raw fibre-reinforced mortars  
 391 have lower diameter flows than the reference mortar (with and without plasticizer):  $(17.8 \pm 0.3)$  cm  
 392 with 1.0 %vol. of raw flax fibres,  $(13.7 \pm 0.6)$  cm with 2.0 %vol. and  $(11.4 \pm 0.6)$  cm with 3.0 %vol. Thus,  
 393 as the fibre content is increased, the flow decreases, reflecting a reduction of workability. This trend  
 394 has also been highlighted by other studies [11,66]. The linseed oil treatment of flax fibres  
 395 significantly enhances the workability of fresh fibre-reinforced mortars. Indeed, with the LO-0.5  
 396 treated fibres, the measured diameters are 69% higher, on average, than raw-fibre-reinforced  
 397 mortars. With these treated fibres, the diameter is always greater than 18 cm, even with 3.0 %vol. of  
 398 fibres in the mixture. With 1.0 %vol., the measured flow is near 30 cm, i.e. a very fluid consistency.  
 399 With the LO-0.25 treated fibres, the workability of the mortars is also improved compared to raw-  
 400 fibre-mortars, but to a lesser extent. For 1.0 %vol. fibres, the diameter is greatly increased, up to  
 401  $(25.9 \pm 0.6)$  cm - i.e. 45% higher than raw-fibre-reinforced mortar. On the other hand, the  
 402 improvement is less significant with 3.0 %vol. of fibres, with an improvement of 19%.



403

404

**Fig. 9.** Flow diameters measured with flow table for the different mortars.

405 The significant reduction of the water absorption of flax fibres due the linseed oil treatment can  
406 account for these enhancements in workability. Moreover, the presence of the coating can also  
407 explain this improvement. Indeed, the linseed oil treatment has the effect of increasing the apparent  
408 cross-sectional area (due to coating) of the fibrous reinforcement, and therefore their specific surface  
409 area is reduced. Thus, the demand for cement paste to meet the workability criteria is greatly  
410 reduced.

### 411 3.2.3. Porosity

412 As it can be seen in Fig. 10, the addition of flax fibres increases the porosity of mortar, as reported  
413 for raw flax fibre reinforced concrete [3]. The increase in porosity correlates strongly to the air  
414 content of fresh mortars. This suggests that the increase of porosity is mainly due to the increase of  
415 the entrained air previously observed on mortars. A previous study had highlighted that the  
416 increased air content is probably caused by the decrease of the granular compactness induced by the  
417 addition of fibres into the mixture [66]. The porosity raises from  $(13.9\pm 0.2)$  %, for the reference  
418 mortar up to  $(19.9\pm 0.2)$  % for mortar reinforced with 3.0% of linseed oil treated flax fibres (ratio =  
419 0.5). The treatment of flax fibres with linseed oil induces a higher porosity in the composite in  
420 comparison with raw flax fibres, especially with LO/fibre ratio of 0.5. For the LO-0.25 treatment, the  
421 increase in porosity ranges from 3% to 9%, depending on fibre content. Obviously, the higher the  
422 fibre content is, the more the linseed oil increases the porosity. Coating with linseed oil results in  
423 higher porosity for LO/fibre ratio of 0.5; the relative increase in porosity ranges from 9% to 22%. As  
424 the mechanical properties of brittle materials are governed by the porosity, significant reductions in  
425 mechanical performances of these mortars are expected due to the high porosity.

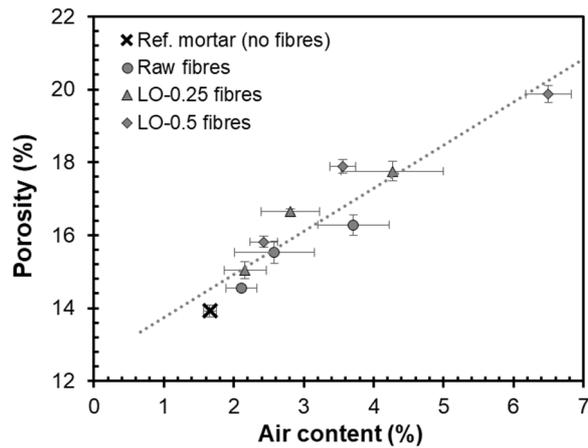
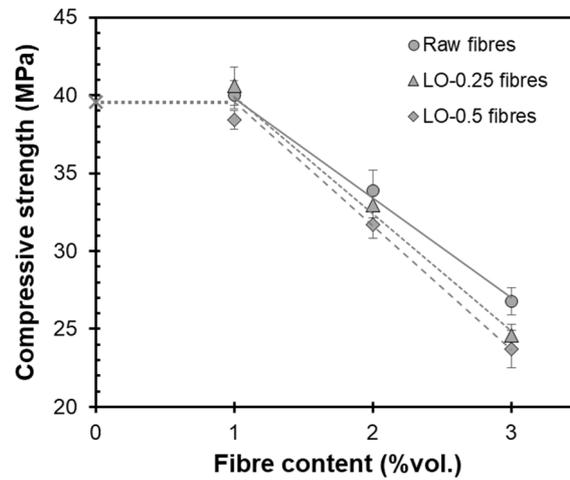


Fig. 10. Porosity of composites as a function of the air content.

### 3.2.4. Mechanical properties

The results of the compressive tests carried out on the various mortars are presented in Fig. 11. Firstly, the reference mortar has a compressive strength of  $(39.6 \pm 1.7)$  MPa. For most fibre-reinforced mortars, a decrease in compressive strength is noted in comparison with the reference mortar (0 %vol. of flax fibres). For 1.0 %vol. of fibres, the compressive strengths of the flax-fibre-reinforced mortars are quite the same as the reference mortar. The compressive strengths then decrease significantly when the fibre content is raised. The values are between 31.7 and 33.9 MPa with 2.0 %vol. of flax fibres, and between 23.1 and 26.8 MPa with 3.0 %vol. of fibres. Linseed oil treatment appears to depress the compressive strength. Indeed, the strengths of the mortars reinforced with treated fibres are lower than those of the mortar with raw fibres, and in particular the linseed oil treatment with 0.5-ratio. A trend seems to be emerging with the fibre content. It appears that increasing the fibre content results in a decrease in compressive strength. The reduction of the compressive strength was highlighted by other authors on bio-fibre cementitious composites [3,21]. This is attributed to the increased air content in the concrete, which reduces its compactness, and thus the compressive strength. Indeed, it was previously noted (Fig. 8) that the air content increases with fibre content. Other authors have also noted decreases in compressive strength with the use of linseed oil as a treatment for plant aggregates [27]. These authors have observed that treating the plant particles with linseed oil created a lower quality interface between the matrix and the plant aggregates, which explains the observed decrease in compressive strength. One can think that during

447 a compression test, the coalescence of the interfacial cracks quickly leads to the ruin of the sample  
448 (low value of the tensile strength).

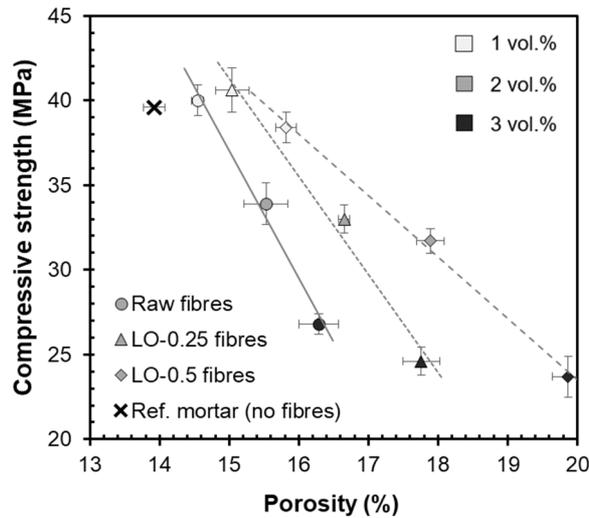


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**Fig. 11.** Compressive strength of composites after curing for 28 days.

451 Fig. 12 illustrates the reduction of the compressive strength as a function of the porosity of  
452 mortars. For an addition of 1.0 %vol. of flax fibre, the strength decrease is very slight, even negligible.  
453 For a compressive resistance practically unchanged compared to the unreinforced mortar which has  
454 a porosity of 13.9%, with 1.0 %vol. of fibers the porosity increases according to the treatment  
455 applied: 5% for the raw fibers, 8% for LO-0.25 and 14% for LO-0.5. The same trend is observed for  
456 the reinforcement rates of 2.0 %vol. and 3.0 %vol. However, the relative increase in porosity is  
457 greater with higher fibre contents.



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**Fig. 12.** Compressive strength as a function of the composite porosity.

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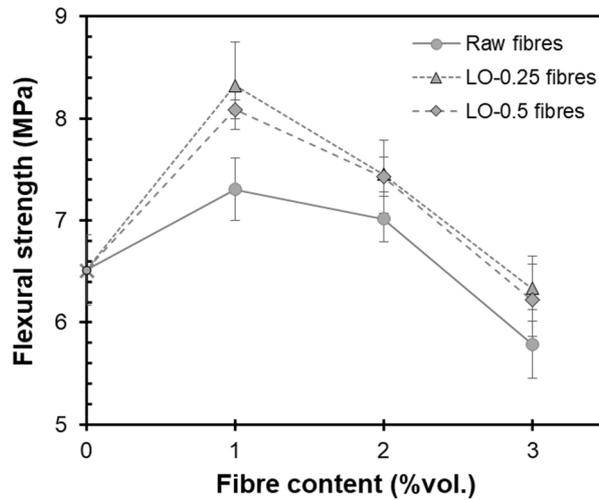
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The results of three-point bending tests after 28 days of curing are presented in Fig. 13. While the reference mortar has a flexural strength of  $(6.5 \pm 0.4)$  MPa, it appears that the addition of flax fibres into mixtures has a significant influence on the flexural strength. Indeed, with the addition of 1.0 %vol. of fibres, the flexural strength is between  $(7.3 \pm 0.3)$  MPa with raw fibres and  $(8.3 \pm 0.1)$  MPa with LO-0.25 fibres – i.e. relative increases of 11% and 28%, respectively. It is for this fibre content that the highest flexural strengths are observed. The flexural strengths of mortars with 2.0 %vol. of fibres are lower in comparison with those with 1 %vol. of fibres, while remaining higher than the reference mortar. In contrast, flexural strength of treated flax fibre reinforced mortars with 3 %vol. are much lower, with values close to that of the reference mortar, ranging from  $(6.2 \pm 0.4)$  MPa to  $(6.3 \pm 0.4)$  MPa). In the latter case then, the flax fibres do not seem to have any beneficial effect on the mortar's strength. With fibre contents of 1.0 %vol. and 2.0 %vol., at which flax fibres seem to significantly improve the composite flexural strength, coating with linseed oil can significantly improve the mortar's strength in comparison to raw flax fibres. However, both linseed oil treatments have similar results. The use of LO/fibres ratio of 0.5 instead of 0.25 does not seem to have a significant effect on the mortar' strength. Similar results were obtained in another study dealing with linseed oil-coated diss fibres, with significantly improved flexural strengths [30].



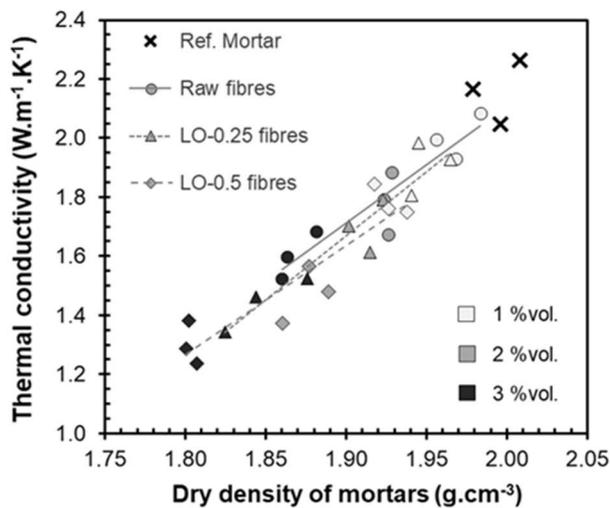
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Fig. 13. Flexural strength of composites after curing for 28 days.

478 3.2.5. Thermal conductivity

479 The thermal conductivity of mortars was measured in the dry state (drying at 60°C in an oven for  
 480 24 h, prior to the test). Fig. 14 presents the variation of thermal conductivity as a function of the dry  
 481 density of mortars. A quasi-linear relationship seems to fairly describe the thermal conductivity as a  
 482 function of the mortar density. This relationship has already been highlighted by several authors who  
 483 investigated the thermal characteristics of bio-based building materials [67]. Note that this  
 484 relationship can be considered linear in the domain of interest to us here, but this is not true in  
 485 general.



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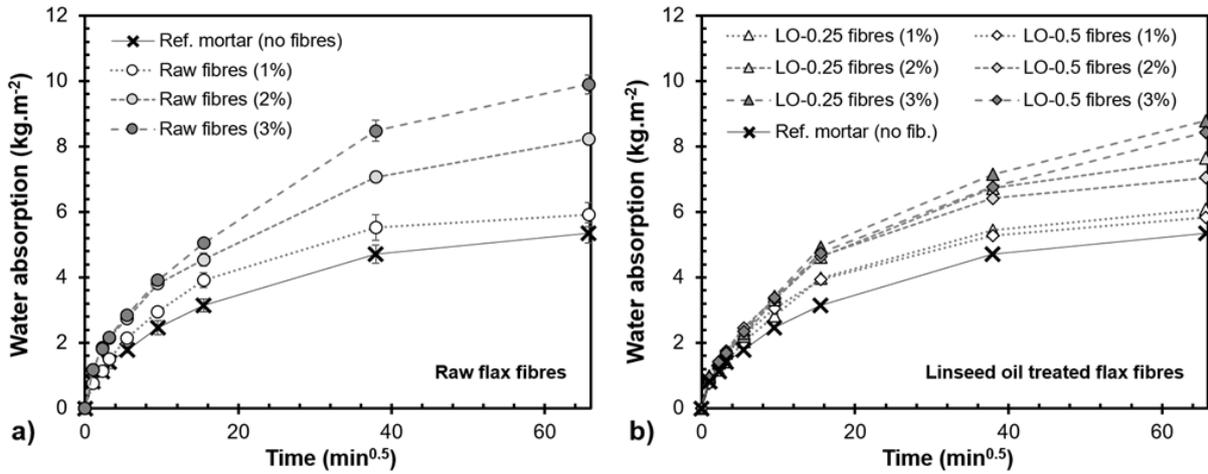
**Fig. 14.** Thermal conductivity of composites according to their dry densities.

488 The reference mortar exhibits the highest thermal conductivity, with values between 2.05 and  
489 2.37  $\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ . The increase in fibre content leads to a decrease of the thermal conductivity. Thus, the  
490 mortars incorporating 1.0 %vol. of flax fibres show thermal conductivities slightly lower than the  
491 reference mortar, ranging from 1.75 to 2.08  $\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ . On the other hand, mortars with 3.0 %vol.  
492 fibres have the best thermal characteristics, with conductivity between 1.23 and 1.68  $\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ . Thus,  
493 flax fibres can slightly improve the thermal insulation properties of the material. In addition, for  
494 equal fibre content, mortars with linseed oil-treated flax fibres have lower thermal conductivities  
495 than those with raw fibres. Linseed oil therefore is beneficial to this characteristic, because it allows  
496 reducing the density of the material, which leads to a decrease in thermal conductivity.

#### 497 3.2.6. Capillary water absorption

498 Some authors have highlighted that plant fibre-reinforced cement composites are vulnerable to  
499 weathering and water absorption [68,69]. Therefore, the water absorption of bio-based cement  
500 composites is an important parameter for the durability of this material. The capillary water  
501 absorption kinetics for the reference mortar and raw fibre-reinforced mortars are illustrated in Fig.  
502 15a, while those of treated fibre-reinforced mortars are presented in Fig. 15b. In addition, the  
503 capillary water absorption coefficients of all mixes are provided in Fig. 16. It can be seen that the  
504 reference mortar shows the slowest and lowest water absorption behaviour, with a coefficient of 194  
505  $\text{g}\cdot\text{m}^{-2}\cdot\text{min}^{-0.5}$ . The addition of flax fibres to the cement matrix leads to an increase in the water  
506 absorption capacity. Obviously, higher fibre content leads to higher water absorption. After 72 h (*i.e.*  
507 65.7  $\text{min}^{0.5}$  in Fig. 15), the absorption of the 3.0 %vol. raw fibre mix is almost twice as high as that of  
508 the reference mortar, 9.90  $\text{kg}\cdot\text{m}^{-2}$  and 5.35  $\text{kg}\cdot\text{m}^{-2}$ . This increase can be attributed to two phenomena:  
509 the high-water absorption capacity of plant aggregates and the increase in the water accessible  
510 porosity of the composites. However, the absorption coefficients remain within acceptable values in  
511 comparison with other studies incorporating plant particles [70]. This is due in particular to the  
512 relatively low fibre content of the composites investigated in this study.

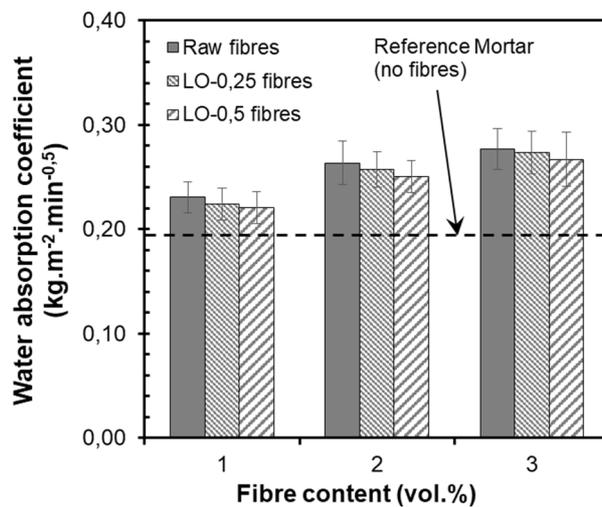
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514

515 **Fig. 15.** Capillary water absorption of mortars with raw flax fibres (a) and with linseed oil treated fibres (b).

516 It seems that the linseed oil treatment reduces the capillary water absorption capacity of mortars  
517 (Fig. 15 and Fig. 16). This treatment decreases both the absorption kinetics and the maximum water  
518 retention, especially in mixes containing 2.0 %vol. and 3.0 %vol. of fibres. The use of a higher  
519 LO/fibre ratio further decreases the absorption capacity of the mortars. However, the gain between  
520 LO-0.25 and LO-0.5 ratios remains marginal.



521

522 **Fig. 16.** Coefficient of capillary water absorption of mortars with raw flax fibres and with linseed oil treated  
523 fibres.

#### 524 **4. Conclusion**

525 This work has investigated a way to improve the physical properties of flax-fibre-reinforced  
526 mortars using a linseed oil coating. The linseed oil treatment has been optimized by using low linseed  
527 oil content while minimizing water absorption. The following main conclusions can be drawn:

528 • The water absorption of flax fibres disrupts the rheological behaviour of the cementitious  
529 composites in the fresh state and also affects the mechanical properties in the hardened state. The  
530 hygroscopic behaviour of raw and linseed oil coated flax fibres was studied to begin with. It appeared  
531 that the flax fibres very rapidly absorb a large amount of water (up to 130% by mass). The linseed oil  
532 treatment decreases the water absorption capacity. Thus, the LO/fibres ratios of 0.25 and 0.5 seem to  
533 be the most effective for decreasing the water absorption.

534 • Linseed oil treatments reduced the water absorption of fibres, resulting in a significant  
535 improvement in workability for composites made with these treated fibres. Furthermore, these  
536 coating treatments increased the fibres' diameter, leading to a decrease of the specific surface area  
537 and therefore the water demand of the flax fibres.

538 • The mechanical tests carried out on hardened mortars have shown a continuous decrease of the  
539 compressive strength as fibre content increases. The higher air content and porosity of mortars with  
540 high fibre content may explain this decreasing trend in strength. Decreases in compressive strength  
541 were also noted with the use of linseed oil.

542 • With fibre contents of 1.0 %vol. and 2.0 %vol., for which flax fibres seem to significantly improve  
543 the mortar's flexural strength, linseed oil coating can significantly improve the mechanical strengths  
544 in comparison with raw flax fibres.

545 • Flax fibres can slightly improve the thermal insulation properties of the material. In addition, for  
546 equal fibre content, mortars made with flax fibres treated with linseed oil have lower thermal  
547 conductivity than those made with raw fibres. Linseed oil is therefore beneficial to this characteristic,  
548 because it helps reducing the density of the material, which leads to a decrease in thermal  
549 conductivity.

550 This study has therefore highlighted that linseed oil could be an interesting treatment solution for  
551 composites reinforced with plant fibres. However, microstructural observations will have to be  
552 carried out in order to study the interfacial transition zone at the fibre-oil-matrix interface. Future  
553 work will investigate the possible chemical interactions between polymerized linseed oil and the  
554 cement matrix. In addition, longer term experiments would give additional information about the  
555 effectiveness of the treatment over time and therefore its durability.

## 556 **Acknowledgments**

557 The authors would like to acknowledge Vicat for supplying the cement used in this study, Argeco  
558 for the Metakaolin and BASF for supplying the superplasticizer and VMA admixture. In addition,  
559 sincere thanks go to the Depestele Company for the supply of flax fibres.

## 560 **Data Availability**

561 The data that support the findings of this study are available from the corresponding author, JP,  
562 upon reasonable request.

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