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# Coating of a lignocellulosic aggregate with pectin/polyethylenimin mixtures: Effects on flax shive and cement-shive composite properties

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#### ABSTRACT

Lignocellulosic lightweight concretes are a potential contributor to sustainable development. However, lignocellulosic aggregates are not always fully compatible with cement matrices leading to setting delays, significant dimensional variations and low mechanical strengths. An aggregate treatment, reducing water absorption and water-soluble molecule release, can avoid or reduce these drawbacks. In this study a coating treatment, using a pectin/polyethylenimin (PP) mixture, was applied to flax shives, which is a lignocellulosic by-product. Before shive coating, a dilution with distilled water or a micro-wave heating were conducted to decrease PP mixture viscosity.

The PP treatment involved a decrease in shive water absorption. Compared to standard shive concrete, treated shive concrete exhibited a decrease in setting delay with an increase in cement hydration enthalpy, an increase in mechanical strengths and a significant reduction in dimensional variations. Although a slight increase in thermal conductivity and bulk density was measured, the cement-shive composite obtained still belongs to the insulating concrete category.

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# 1. Introduction

In recent years particular attention has been given to lignocellulosic lightweight concretes due to (i) thermo-physical considerations: high thermal inertia, high thermal resistance and low weight [1], but also to (ii) environmental and economical considerations: preservation of mineral aggregates, use of vegetal wastes and energy saving during fabrication and utilization. Lightweight concrete formulation including local lignocellulosic by-products is a way to contribute to sustainable development in the construction sector.

In 2008, according to FAO estimates [2], Europe produced 50% of world flax fiber and France was the second producer behind China. Flax fiber production generates by-products such as short fibers and shives. Important amounts of flax shives are available because 2.5 tons are obtained per ton of fiber produced [3]. They have found usage as animal bedding, mulching and fuel for thermal energy, or in fiberboard fabrication. Flax shives, with their lightness, honeycomb structure and annual renewal, exhibit desirable characteristics for use as lignocellulosic aggregates in lightweight concretes.

The literature relates various studies upgrading local vegetal resources in a cementitious matrix: bagasse in French Antilles [4] and in India [5], wheat straw in USA [6], oil palm shell in Malaysia [7], wood shavings in Algeria [8,9] and in Italy [1], coconut husk in

Nigeria [10], sugar beet pulp in France [11], hemp shives [12] and flax shives [13] in France. In all cases insulating concretes were obtained. However a setting delay, important dimensional variations and low mechanical strengths as consequences of high water absorption capacity and water-soluble molecule release of lignocellulosic aggregates have been encountered by most authors [14].

Therefore these lignocellulosic aggregates must be treated before their incorporation in a cementitious matrix. Treatments under high temperature and pressure allow decreasing wood hydrophilic capacity, swelling and shrinkage [15-17]. Thermal treatments in aqueous medium remove pectin and water soluble hemicelluloses [13,18] responsible for hydration defaults of cementitious matrix. Coating is another way of treatment consuming less energy and being more environmentally friendly. Thus vegetal aggregate is isolated from cement matrix preventing its water absorption and migration of water-soluble molecules. Coating can be conducted with mineral or organic substances not very permeable to water and its compatibility with cement matrix has to be controlled. Cement, aluminum sulphate and hydraulic lime have been used to coat flax shives, sugar beet pulps and wood shavings [9,11,19-21]. Polymerized oil, sucrose-cement mixture and a PEC elastomer have also been used as coating substance for lignocellulosic aggregates [11,14,19-20]. The mechanical strength and aggregate-matrix adherence of the resulted composites were improved, and their shrinkage was significantly reduced.

Oil polymerization and PEC elastomer preparation being time consuming, it is necessary to propose other coatings easy to implement and using nontoxic natural organic substances. Due to their

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water resistance and biodegradability, coating products of pharmaceutical domain could be applicable or adaptable to lignocellulosic aggregates. In that sense a mixture of pectin (a polysaccharide polymer presents in plant cell walls [22]) and polyethylenimin (a cationic polymer used as an attachment promoter [23]) has been retained [24]. According to Fattal et al. [24], pectin addition to a PEI gel leads to a decrease in drying duration and an increase in mechanical strength of final product. Pectin addition transforms PEI gel into a rigid polymer. Consequently to improve flax shive coating it is necessary to decrease mixture viscosity. Three ways have been chosen: a microwave thermal treatment, a dilution with distilled water and a combination of these two ways.

The study presented herein focuses on the effect of a pectin/polyethylenimin (PP) coating of flax shives on lignocellulosic aggregate and cement composite properties according to the method used to decrease coating mixture viscosity. The goal is to use these coated shives as aggregates to prepare a light concrete with better mechanical strengths and a high capacity of thermal insulation compared to those of noncoated shive concrete. Real bulk density and water absorption capacity of treated shives are determined and their micro structural morphology is characterized by scanning electron microscopy (SEM). The impact of coated flax shives on cement matrix is evaluated through hydration enthalpy and setting time values, but also through SEM imaging. Finally mechanical, thermal and hydrous tests are conducted to evaluate the influence of the coating treatment process on the cement-shive composite properties.

#### 2. Materials and methods

#### 2.1. Raw materials

Flax shives were supplied by CALIRA (Coopérative Agricole Linière de la Région d'Abbeville, France). These lignocellulosic particles, 4–8 mm in length and 0.6–1.3 mm in width, were washed with distilled water at room temperature and dried at 50 °C for 3 days until constant weight before their use.

The cement used was Portland cement CPA CEM I 52.5 R supplied by Calcia, according to the EN 197-1 standard [25]. Water and Ordinary Portland Cement (OPC) were the only components of the cementitious matrix.

Chemical materials were pectin practice produced by Acros Organics, with 67-71% as degree of esterification and polyethylenimin produced by Aldrich, with low molecular weight (Mw  $\sim 1300$ ) and 50 wt.% in water (d = 1.08).

# 2.2. Flax shive treatment and characterization

## 2.2.1. Treatment process with pectin/polyethylenimin

A preliminary study has been conducted to determine pectin/PEI mass ratio in order to minimize drying duration of coating substance. The resulting ratio, equal to 1, was used herein. In other respects, as a viscous mixture is obtained following pectin addition in PEI gel (the resulting mixture is called PP1), it is necessary to decrease viscosity to be able to coat efficiently flax shives. Three ways have been chosen: a microwave thermal treatment (the resulting mixture is called PP2), a dilution with distilled water (the resulting mixture is called PP3) and a combination of the two previous (the resulting mixture is called PP4). Therefore, four PP mixtures were applied to treat the shives.

After addition of distilled water to obtain a half dilution of the PP mixture, a mixing was performed during 2 min at 140 rpm. For thermal treatment, a household microwave was used during 1 min/100 ml of PP mixture with a power of 900 W, then a 2 min mixing at 140 rpm was performed. For dilution and thermal treat-

ment combination, the PP mixture has undertaken the following steps: dilution, mixing, thermal treatment and mixing.

Whatever the process used to obtain a PP mixture, aggregate coating was performed with a flax shive mass equal to PP mixture mass before dilution and a mixing under the conditions described before. Then coated shives were dried at 50 °C until constant mass before composite elaboration.

To test the chemical interaction between pectin and PEI in the different mixtures, a Fourier transformed infrared spectrometer (FTIR) has been used. It was an IR Prestige 21 provided by Shimadzu equipped with an ATR Ge analyser. The data processing software was the IR-solution. FTIR analyses have not highlighted interactions between pectin and PEI after water addition and/or thermal treatment. This is consistent with Fattal et al. [24] results. However after thermal treatment, a decrease in spectra intensity at 1050 cm<sup>-1</sup> corresponding to C–O links and an increase at 3420 cm<sup>-1</sup> corresponding to O–H links were observed (data not shown). These spectra modifications may be involved by pectin hydrolysis.

# 2.2.2. Physico-chemical characterization of flax shives

Flax shives (coated or not) were dried at 50 °C until constant weight before water absorption experiments. A measured mass was immersed in distilled water for 3 h (water saturation time of non-coated shives). After elimination of surface water using absorbent paper, the gain of mass is calculated. A pycnometric method in glycerol was used to determined the real bulk density of shives (coated or not). Each measuring type was replicated threefold for each coating treatment.

#### 2.2.3. Micro structural characterization of flax shives

The micro structural morphology of treated flax shives was studied by means of scanning electron microscopy (SEM). The SEM analysis enables evaluating the treatment effects on shives. The micrographs were performed using a PHILIPS FEG XL 30 microscope. To facilitate shive observation, a thin layer of spray-on gold, which acted like a conductor, was applied on the beforehand dried samples. For stereoscopic visualization the secondary electron (SE) mode was used.

## 2.3. Elaboration and characterization of cement-shive concrete

To upgrade the more flax shives and to use the less OPC, a preliminary study has been conducted in order to define water to cement (W/C) and shives to cement (S/C) mass ratios to obtain a nonfriable composite. The resulting ratios were applied herein: S/C = 4 and W/C = 0.75. During the different experiments, W/C mass ratio was kept constant. Except drying steps, flax shives have not undergone any treatment before their coating and mix with cement and water. Shives, OPC and water were mixed using a standard mixing machine (according to EN 196-1 standard [26]).For each mixture, three  $4 \times 4 \times 16 \text{ cm}^3$  prismatic specimens were made. The samples were allowed to cure for 24 h, and then demoulded and cured during 28 days at a temperature of  $20 \text{ °C} \pm 2 \text{ °C}$  and 95% relative humidity inside a control room. After curing, concrete samples were dried in a 50 °C oven for 7 days (until constant weight).

Possible interactions between cement and aggregates (coated or not) can appear as soon as the mixing step and induce cement hydration enthalpy and setting time values different from an OPC paste or a standard lignocellulosic concrete. Herein a SETARAM C80 calorimeter was used, in isothermal mode at 25 °C, to record the heat flow during hydration reactions. The concrete mixture was prepared following the procedure recommended by Govin [27].

Due to the presence of vegetal aggregates, able to absorb water, the lignocellulosic concretes exhibit important dimensional variations [28]. In order to quantify shive coating impact on hydrous behavior of composites, a determination of drying shrinkage (DS) and extreme dimensional variations (EDV) was carried out according to EN 196-1 standard [26] using a retractometer (Controlab, NF P 15-433 and NF P18-427). Results are expressed in terms of millimeters per meter (mm/m).

To evaluate the effect of PP coating on mechanical behavior of shive concrete, three points bending and compression tests were carried out according to EN 196-1 standards [26] on a hydraulic machine Proviteq. Concrete bulk density was also measured because it can influence mechanical strengths.

Lignocellulosic concretes exhibit thermal insulating properties. So the thermal conductivity of composite was determined using a transient dynamic electrothermal measurement method. The technique employed was the Transient Plane Probe (TPS), which stems from developments resulting from the THS (Transient Hot Strip) probe technique derived by Gustafsson et al. [29]. Tests were carried out at dry state, normal temperature and pressure, on samples of dimensions  $100~\text{mm} \times 100~\text{mm} \times 100~\text{mm}$  placed in an enclosure at constant temperature.

Each measurement has been performed in triplicate for each coating treatment.

Pieces of the fractured surface of half specimens obtained after mechanical tests were used for SEM observation. The SEM analysis was performed with the same equipment and conditions as described previously (Section 2.2.3) and enables evaluating the influence of PP coated shives on the cementitious species development and paste-aggregate adhesion.

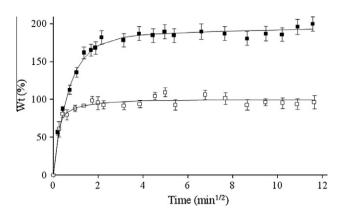
# 3. Experimental results and analysis

# 3.1. The influence of PP coating on flax shives properties

# 3.1.1. Water absorption and real bulk density of treated shives

Coating purpose is to isolate flax shive from cement matrix preventing its water absorption and migration of water-soluble molecules. To evaluate effectiveness of the PP coating towards water, water absorption tests were conducted on the different shives (PP1 to 4). A similar evolution is obtained with the four processes, so only the water absorption kinetics of nontreated and PP4 treated shives are represented in Fig. 1.

The water saturation of treated shives is very fast compared to the one of nontreated shives. It is reached in 3 min against 135 min in the case of standard shives. Moreover the maximum of



**Fig. 1.** Comparison between water absorption (Wt) kinetics of nontreated ( $\blacksquare$ ) and PP4 treated flax shives ( $\sqcap$ ).

absorption is less than 100% against over 213% obtained with the standard shives. This result shows the efficiency of PP treatment on shive hydrophobisation. A decrease in water absorption of a same range (50–60%) has been observed by Khazma et al. [14] for flax shives with a PEC coating and by Monreal et al. [11] for sugar beet pulp with a linseed oil coating. A better reduction in water absorption, yielding 75–80% has been obtained with a linseed oil coating of wood shavings [19] and with a cement-sucrose coating of flax shives [21].

In other respects, due to the biodegradable character of the PP coating, fears could exist towards its efficiency for a long time. Durability experiments have not been conducted but the mixture is enough stable for concrete elaboration because it still coats shives after 28 days of composite cure (Fig. 2).

Flax shives can be used as light aggregates due to their low real bulk density  $(127.7~{\rm kg~m^{-3}})$ . With the four coating processes, no significant difference in real bulk density is observed (Fig. 3). A 60% increase in real bulk density is obtained. Except Khazma et al. [14] who have obtained only a 40% increase with a PEC coating of flax shives, the other authors do not give real bulk density values for their coated lignocellulosic aggregates [11,19,21].

# 3.1.2. Shive coated surface characterization

Although there is no difference in water saturation and real bulk density between the shives coated according to the four processes, the SEM analysis shows differences in the quality of shive coating (Fig. 4).

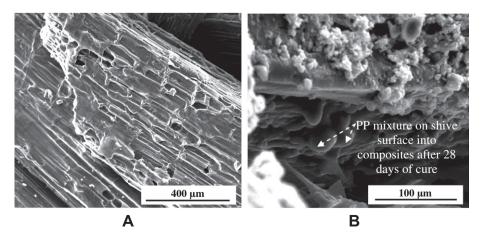


Fig. 2. Coated shives according to PP4 process (A: magnification × 250) before concrete elaboration and into the cement matrix (B: magnification × 1000).

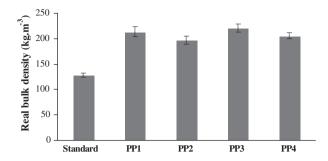


Fig. 3. Real bulk density evolution of flax shives according to PP treatment.

PP accumulations are visible when the treatment process is performed without water addition (delimitated spaces in A and B micrographs of Fig. 4). These accumulations could be the result of a nonhomogeneous dispersion of pectin in the PEI network. In fact the microscopic observation of a pectin/PEI mixture has shown drops of pectin reticulated with polyethylenimin [24]. Furthermore the high viscosity of PP mixture prepared without adding water results in an incomplete coating of shive surface.

With addition of water in the treatment process (PP3 and 4), the formation of PP accumulations is inhibited as shown in C and D micrographs of Fig. 4. Finally when a thermal treatment is combined with a dilution (PP4), a PP film covers the whole shive surface and does not fill the honeycomb structure of flax shives (Fig. 4D).

Thereafter only the composites including PP1 and PP4 coated shives were characterized.

# 3.2. The influence of shive PP coating on composite properties

3.2.1. Concrete setting time and cement hydration enthalpy evaluation

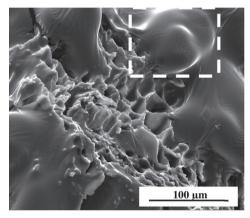
The hydrophilic character of flax shives and their ability to release water-soluble molecules lead to setting delay and modification in hydration reactions. In order to evaluate the impact of PP processes on cement matrix, the time of setting was determined by the evaluation of cement hydration heat flow thanks to a microcalorimetric method. The analysis has been conducted on an Ordinary Portland Cement (OPC) paste and concretes prepared with nontreated standard shives, PP1 and PP4 treated shives. Resulted curves are presented in Fig. 5. They offer the possibility to calculate

the times of setting beginning and end. More the integration of

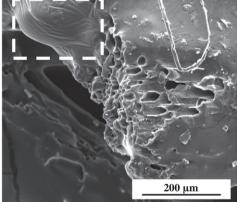
delimited peak gives the enthalpy of cement hydration in the mix-

ture. Calculated results are presented in Table 1.

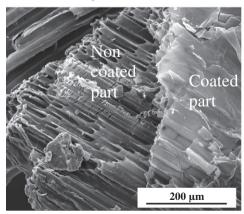
With standard flax shives, cement hydration is slow, a setting delay is detected and the hydration enthalpy value is very low. The comparison of enthalpy values obtained with standard shives concrete and OPC paste shows that cement hydration is inhibited by the lignocellulosic aggregates. This inhibition is accentuated when PP1 treated shives are present in the concrete. In fact the characterization of shive coated surface has shown nonhomogeneous dispersion of pectins in the PEI network for PP1 coating process and pectins are known for their high capacity to cause a delay in setting time [30]. With PP4 process, a more important enthalpy



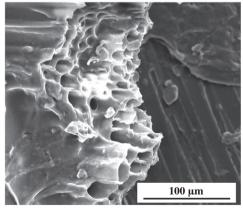
A- Shive coated according to PP1 process (magnification x1000)



B- Shive coated according to PP2 process (magnification x 500)

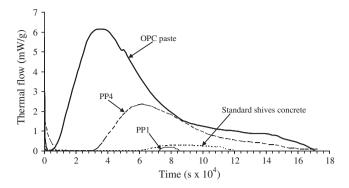


C- Shive coated according to PP3 process (magnification x 500)



D- Shive coated according to PP4 process (magnification x 1000)

 $\textbf{Fig. 4.} \ \ \textbf{Micro characterization of coated shives according to different processes}.$ 



**Fig. 5.** Microcalorimetric curves of OPC paste (—), standard shives concrete  $(\ldots)$ , PP1 (-) and PP4 (- -) concretes.

**Table 1**Setting times and enthalpy obtained from Fig. 5 curves.

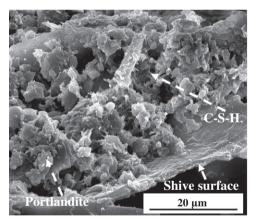
		OPC paste (52.5 R)	Concrete prepared with		
		,	Standard shives	PP1 treated shives	PP4 treated shives
Setting time (h)	Beginning End	1.99 9.36	17.06 22.38	20.16 22.89	8.30 16.24
Cement hydration enthalpy (J/g)		377.06	6.03	2.18	154.62

value is obtained indicating a better hydration of the cement. A high quantity of Portlandite tablets and a modest development of C–S–H molecules are detected into composites prepared with shives treated with PP1 coating (Fig. 6A). The development of a high quantity of C–S–H and Portlandite can be observed in composites prepared with shives treated with PP4 coating (Fig. 6B). The delay in setting time is shorter when PP4 treated shives are used instead of standard shives. However it is still important compared to OPC paste. FTIR spectra modifications, probably linked to pectin hydrolysis, have been observed after thermal treatment of PP mixture (cf Section 2.2.1). However some molecules of non-hydrolysed pectins could be still present at the shive surface and act on setting time. This delay in setting time can be corrected by a complementary matrix treatment using a setting acceleration agent.

The other studies on organic coating treatments found in the literature did not always characterize lignocellulosic concretes in the young age [11,19]. Khazma at al. [14] have obtained shorter times of setting beginning (4.69 h) and end (12.02 h) but a lower value of hydration enthalpy (113.21 J/kg) with a PEC coating of flax shives. With a cement-sucrose coating of flax shives [21], the same authors have not calculated the hydration enthalpy, but, although they could not detected the time of setting beginning, they have measured a time of setting end fourfold shorter (4 h).

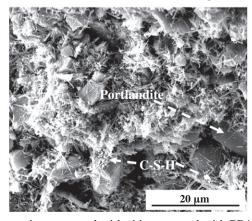
## 3.2.2. Mechanical and thermal behavior

Lightweight aggregate concretes are classified in three categories according to their apparent bulk density, compressive strength and thermal conductivity [31]. The aim of this study is to obtain a



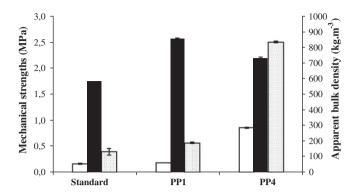


A- Composites prepared with shives treated with PP1 coating (left) and a closer view (right)



B – Composites prepared with shives treated with PP4 coating

Fig. 6. C-S-H and Portlandite development in composites prepared with PP1 and PP4 treated shives.



**Fig. 7.** Variations of composite apparent bulk density ( $\blacksquare$ ), flexural strength ( $\square$ ) and compressive strength ( $\square$ ) according to coating process of flax shives.

cement-shive composite belonging to the insulating class (class III), so exhibiting a compressive strength above 0.5 MPa and a thermal conductivity below 0.3 W m $^{-1}$  K $^{-1}$ . RILEM recommendations [31] specify a value of apparent bulk density below 2000 kg m $^{-3}$  only for bearing class (class I).

The mechanical behavior of composite is assessed by measuring the compressive and flexural strengths. Fig. 7 shows the variations of apparent bulk density and strengths of concretes according to coating process applied to flax shives.

Whatever the coating process used, mechanical strengths of composites are improved. Except Ledhem et al. [19], with a clayey-cement matrix, who have obtained less resistant concrete

with a linseed oil coating of wood shavings, the same trend has been observed with other organic coatings [14,21]. Compared to standard shive concrete, concrete prepared with PP1 treated shives exhibits 11% and 25% increases in flexural and compressive strengths, respectively, and a 46% increase in apparent bulk density. In other respects, values of flexural and compressive strengths are multiplied, respectively, by 4 and 5, when apparent bulk density only undergoes a 25% increase for concrete prepared with PP4 treated shives. So, the increase in strengths is not proportional to the increase in apparent bulk density. This phenomenon is very unusual. Indeed, the use of pectin/PEI mixture without either water addition or thermal treatment (PP1) makes the mixture very compact during composite preparation. After 28 days of curing, PP1 composites are soft with a very compact structure that can explain the important value of apparent bulk density. Moreover the mechanical strengths of PP1 composites remain low compared to those of PP4 composites. This is consistent with the previous statements. Actually in the case of PP1 coated shives, coating observations have shown accumulations of pectins not well dispersed in PEI gel (Fig. 4A and B) at shive surface and these accumulations have been related to the important setting delay, the very low cement hydration enthalpy (Table 1), the abnormally abundant Portlandite and the very small quantity of C-S-H detected (Fig. 6A). On the other hand, in the case of PP4 process, the microcalorimetric study has shown an improvement in the development of cement hydration products (Fig. 6B). In other respects the adherence between aggregates and cement matrix can explain the increase in mechanical strengths [14,32]. In the PP1 and PP4 cases, the adherence between cement matrix and flax shive is improved with the

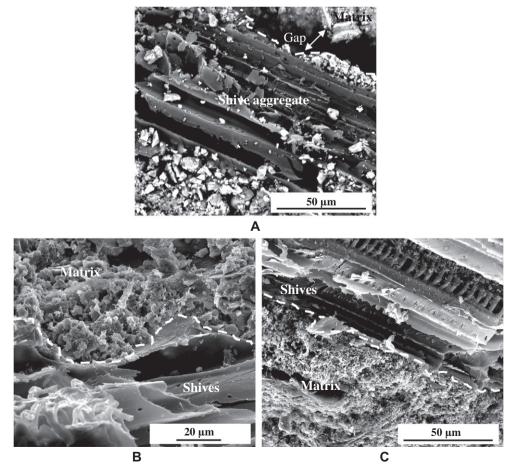


Fig. 8. Interfacial zone between shives and cement matrix for concrete prepared with standard shives (A), PP1 shives (B) and PP4 shives (C).

**Table 2**Thermal conductivity, drying shrinkage (DS) and extreme dimensional variations (EDV) of cement-shive composites according to coating process of flax shives.

	Concrete prepared with			
	Standard shives	PP1 treated shives	PP4 treated shives	
Thermal conductivity $(W m^{-1} K^{-1})$	0.104	1	0.182	
DS (mm $m^{-1}$ )	8.49-8.75	6.02-6.32	4.15-4.30	
EDV (mm $m^{-1}$ )	7.79-8.64	6.23-6.64	4.99-5.24	

treatment: no gap zone is observed in concrete prepared with treated shives (Fig. 8).

The concrete prepared with the PP4 treated shives exhibiting compressive strength greatly above 0.50 MPa (in agreement with RILEM Class III), its thermal conductivity has been measured (Table 2). An increase in thermal conductivity is observed and is probably a consequence of the increase in apparent bulk density [14]. However the value obtained is below 0.3 W m<sup>-1</sup> K<sup>-1</sup> so in accordance with the insulating class of RILEM.

#### 3.2.3. Dimensional variations

Lignocellulosic aggregates are known for their high capacity to absorb water. This characteristic involves aggregate swelling during moistening and shrinkage during drying, so it is responsible for important dimensional variations of concrete. Drying shrinkage (DS) and extreme dimensional variations (EDV) results are shown in Table 2. After PP treatment, the reduction in hydrophilic capacity of shives has been demonstrated (cf 3.1.1). Treated shives do not exchange water as standard flax shives. The amount of water absorbed reduces but does not disappear. This smaller amount of absorbed water into coated shives will impact the DS. In other respects as the hydrophilic capacity of treated shives is not equal to zero, inside the cement matrix the lignocellulosic aggregates may be still able to absorb water and consequently to swell involving EDV. Concretes prepared with treated shives exhibit a decrease in DS and EDV compared to standard shive concrete. The best drying shrinkage and EDV reductions are obtained for PP4 process associating a water dilution and a thermal treatment (-50% in DS and -38% in EDV compared to standard shive concrete). Actually this process leads to a decrease in mixture viscosity providing a better distribution of the treatment substance on shive surface and ensuring, at the same time, the dissolution of chemicals. However dense and lightweight aggregates concretes have to exhibit dimensional variations below 1 mm m<sup>-1</sup> according to EN 771-3 standard [33]. So although the important reduction in EDV obtained with PP4 composite, EN 771-3 standard threshold is not reached, just as in the case of cement composites including cement-sucrose [21] or PEC [14] coated shives. A linseed oil coating of lignocellulosic aggregates seems to be more efficient because EDV values of 1.3 and 2.3 mm/m<sup>-1</sup> were obtained respectively with beet pulps [11] and wood shaving [19] concrete. These findings can be related to the lower water absorption capacity of beet pulps and wood shavings after oil coating (respectively 80% and 40%) compared to the one of flax shives after PP coating (close to 100%).

# 4. Conclusion

The results of this study have shown that a pectin/polyethylenimin mixture can be used as coating substance for flax shives in order to design lightweight cement-shive composite with improved mechanical and hydrous properties. The PP mixture being very viscous and not easy to handle, water dilution and/or thermal treatment have been carried out before application on shives. The coating process had two aims: (i) to minimize hydrophilic capacity of flax shives and (ii) to prevent release of water-soluble molecules into the cement matrix. The first aim has been reached because a 50% decrease in water absorption capacity has been obtained compared to nontreated shives. Although release capacity of water-soluble molecules has not been measured, the results of the microcalorimetric study seem to show that the second aim has also been reached. In fact, the best decreases in beginning and end setting times, and the best increase in cement hydration enthalpy have been obtained for concrete prepared with shives coated with a PP mixture having undergone a water dilution and thermal treatment compared to standard shive concrete.

Coating effectiveness has also been evaluated towards the mechanical strengths of the resulted composites. Compressive and flexural strengths of treated shive concrete were higher by factors of 5 and 4, respectively, compared to standard concrete. These results can be explained by the increase in apparent bulk density, the good adherence between cement matrix and flax shives, but also, and certainly mainly, by a better development of the matrix proved by the increase in hydration enthalpy, the decrease in setting times and the high quantity of C–S–H and Portlandite observed by SEM.

As observed for other lignocellulosic concretes including coated flax shives, the increase in apparent bulk density has induced an increase in thermal conductivity compared to standard shive concrete.

In other respects composite behavior towards water has been improved after the PP treatment because 50% reductions in drying shrinkage and extreme dimensional variations have been obtained compared to standard shive concrete. These results are equivalent to those obtained with cement-sucrose and PEC coatings but higher than those obtained with oil coatings.

Pectin/PEI treatment is easy to implement and, as it uses non-toxic natural organic components, it is environmentally friendly. The resulted composites belong to the insulating class (class III) of RILEM but they do not meet dimensional variations standard. Consequently their uses have to limit water contact. Furthermore a biodegradability study should be conducted to test the water resistance of the PP coating inside the cement matrix.

# Acknowledgements

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