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# Optimization of flax shive-cementitious composites: Impact of different aggregate treatments using linseed oil

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ABSTRACT

This study investigates the effect of flax shive treatments on physico-chemical characteristics of shive and physico-mechanical properties of flax shive cementitious composites. In the context of sustainable development and biorefinery, the main objectives of this work are to optimize a lightweight lignocellulosic concrete and to upgrade a product and a by-product of flax transformation, respectively linseed oil and flax shives. To improve the compatibility between cementitious matrix and vegetable aggregates, flax shives have been treated with raw and emulsified linseed oil. A more marked decrease in water absorption capacity and hydro soluble molecule release has been obtained for shives coated with raw linseed oil. Compared to standard shive composite, whatever the linseed oil treatment, the setting times have been reduced, the mechanical strengths improved and the thermal conductivity slightly increased. In the case of dimensional variations, the coating with raw linseed oil has led to a slight better improvement compared to emulsion coating. The flax shive-cementitious composites obtained exhibit the properties of insulating bearing lightweight aggregate concretes.

## 1. Introduction

The need to improve vegetable fractions behavior is necessary for almost all lignocellulosic resources in order to optimize performances of lignocellulosic cementitious composites. The main drawbacks involved by vegetable aggregates and Portland cement coexistence, concern (i) cement hydration (setting delay and inhibition) due to hydro soluble molecules release, (ii) drying and dimensional variations due to high water absorption capacity of plant material. In order to limit exchanges between matrix and plant fractions, several treatments are described in literature. They consist in extracting disturbing substances, blocking hydrophilic groups with chemical reactions or isolating vegetable aggregate from matrix with a coating. Physical extractive treatments using ultrasonic waves have been used (Cheng-yi et al., 1999; Sun et al., 2004; Vodeničarová et al., 2005; Wang and Zhang, 2006). Woods and lignocellulosic products have been thermally treated either in dry conditions (Kamdem et al., 2002; Rapp, 2001; Rowell et al., 2002; Stamm, 1964; Viitaniemi and Jämsä, 1996; Yew et al., 2014) or wet conditions (Jacobs et al., 2003; Olorunnisola, 2008). Soda treatments were conducted on plant fractions to make them water-repellent (Jähn et al., 2002; Števulová et al., 2012; Terpáková et al., 2012; Van de Weyenberg et al., 2003, 2006). Coatings have been carried out with mineral and organic substances (Bederina et al., 2009, 2012; Borkowski and Körning, 2007; Hoehn and Hoehn, 2001; Khazma et al., 2007, 2008, 2011, 2012; Ledhem et al., 2000; Merzoud et al., 2008; Monreal et al., 2011; Nozahic and Amziane, 2012).

Wood shavings and hemp shives are currently the main commercial aggregates. To be able to use wood aggregates in a cementitious matrix, several patents describe a process consisting in a pyrogenic reaction at 300 °C followed by an impregnation and mineralization of wood shavings (Aschero, 1983; Gaillard et al., 1999; Guidat, 1984). Hemp shives can be directly incorporated in a lime-based binder or after impregnation and petrifaction with several mineral substances (Rasetti, 1990, 1991). The consumptions of energy and inorganic chemicals are the drawbacks of the previous treatment processes. In the context of sustainable development and biorefinery, a study of energy-efficient treatment processes using compounds from renewable natural sources is important. The design of a linseed oil based coating process satisfies this request. Ledhem et al. (2000), Merzoud et al. (2008), Monreal et al. (2011) and Nozahic and Amziane (2012) have used raw linseed oil to

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respectively coat wood aggregates, crushed diss fibres, beet pulps and sunflower aggregates before the incorporation in a mineral matrix.

In fact, linseed oil is a drying oil susceptible to auto-oxidation and polymerization resulting in tough elastic films upon air exposure (Derksen et al., 1995; Sharma and Kundu, 2006). The polymerization reaction is a three-step radical chain reaction (induction, propagation and termination). The induction period of pure linseed oil is about two days and nearly one month is required to complete film formation (Turri et al., 2001). The reaction rate is improved with an increase in temperature and depends on the linoleic fatty acid to linolenic fatty acid ratio (Stenberg et al., 2005). Polymerization time can also be reduced by an emulsion preparation (Bravin et al., 2006). An emulsion is interesting because the coating with a more fluid substance is easier, coated surfaces are less oily, which limits product agglomeration.

In this work, in order to improve the compatibility between cementitious matrix and vegetable aggregates, linseed oil coatings are used to treat flax shives. Linseed oil is applied on its own or emulsified. The impact of the two linseed oil treatments on flax shive characteristics is analyzed. The consequences on composite performances are evaluated by measuring mechanical strengths, dimensional variations and thermal properties.

## 2. Materials and methods

### 2.1. Materials

The cement used was Portland Cement CPA CEM I 52.5 supplied by Calcia. Clinker is the main component (≥95%) of this cement according to EN 197-1 standard. No fillers and no sand particles were applied in this study so result dispersion is avoided.

The aggregates used were flax fibre shives. This main by-product of flax transformation (50% of biomass) is made of elementary particles 6.82 ± 3.82 mm in length and 0.81 ± 0.24 mm in width. The size distribution of bulk flax shives (average length and width) was studied using image analysis. The incremental and cumulated distributions of shive lengths and widths are presented in Fig. 1. Ninety percent of the shives exhibit a length between 1 and 11 mm and a width between 0.1 and 1.1 mm. Almost 60% of the shives show a length between 2 and 6 mm and a width between 0.6 and 0.9 mm. This analysis shows a tight distribution of shive size. In the typical honeycomb structure of shives, the voids account for about 65–70% volume (Dupré, 2005). These lignocellulosic particles were dried at 50 °C until constant weight before their use.

The water used was tap water.

The linseed oil used was commercial grade oil (Mieuxa) sold in a do-it-yourself store as a wood preservative. As natural oil, it is triglyceride esters of fatty acids. The main fatty acids in linseed oil are in descending order linolenic acid, oleic acid, linoleic acid, palmitic acid and stearic acid (Khot et al., 2001). It exhibits a specific gravity between 0.931 and 0.936 and an iodine value between 175 and 204 showing a high degree of unsaturation (Derksen et al., 1995). The linseed oil was used on its own or under a more fluid form by emulsification.

### 2.2. Elaboration of cement-shives composites

Cement, shives and water were mixed using a standard mixing machine according to EN 196-1 standard. For each mix, three 4 cm × 4 cm × 16 cm prismatic specimens were made. The mixture was introduced into the mold in two layers. Between each addition, the mold was placed on a shock table for 60 hits. After mixture, leveling the samples were allowed to cure for 24 h, then demolded and cured during 28 days at a temperature of 20 ± 2 °C and 95% relative humidity inside a controlled room. After curing, concrete samples were dried in a 50 °C oven until constant weight before micro structural, mechanical, thermal and hygral characterization.

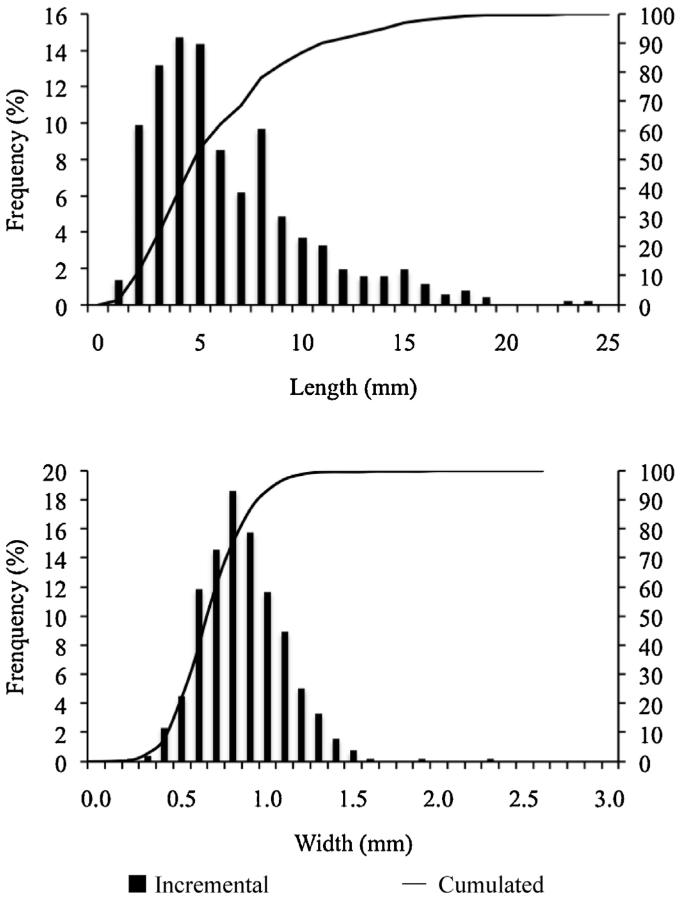

**Fig. 1.** Incremental and cumulated distributions of flax shive length (top) and width (bottom).

The cement composite was prepared with a shives to cement (S/C) volume ratio of 4 and a water to cement (W/C) mass ratio of 0.50. These ratios arise from a preliminary study conducted in order to obtain a composite containing the more lignocellulosic aggregates without being friable. The W/C mass ratio is maintained constant for all composites with coated and not coated flax shives.

### 2.3. Composites characterization

In order to evaluate oil-coating impact on composite micro structural morphology and cementitious species development, scanning electron microscopy and X-ray diffractometry were conducted. To stop cement hydration after 28 days curing, samples were placed in an anhydrous ethanol solution during 24 h and then dried at 50 °C until constant weight. For SEM spray-on gold samples were used. In the case of X-ray diffraction samples were ground before dehydration.

Scanning Electron Microscopy was performed using a PHILIPS FEG XL 30 microscope. For microscopic visualization the secondary electron (SE) mode was used. X-ray diffractometry was performed using a Bruker D8 Advance. Rietveld refinement analysis was carried out on X-ray diffractograms using SIRO-QUANT$^{TM}$ software (Sierotonics) in order to quantity anhydrous and hydrated phases in the cementitious matrix.

Possible interactions between cement and shives (coated or not) can appear as soon as the mixing step starts and induce setting time delay. So isothermal calorimetry was undertaken to