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# LINSEED FIBRE LIME COMPOSITES – EFFECT OF FIBRE PRETREATMENT

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**Abstract:** This paper describes a study of linseed fibre lime matrix composites. A number of pre-treatments were applied to the linseed fibres to investigate the effect on composite strength. The fibres were pre-treated using a two stage process. The first stage involved separating the fibre bundles by soaking in either pure water or a mixture of water and yeast for 10 days. The second stage involved coating the fibres with a layer of calcium hydroxide. Coating was achieved by either mixing with calcium oxide and compressing or stirring the fibres in a solution of lime putty and water. Both these processes were applied to the damp fibres over a 24 hour period. The treated fibres were then mixed with either calcium lime (CL90) or natural hydraulic lime (NHL5) binders to produce composite materials. Stirring in lime putty and water produced a well adhered calcium hydroxide coating on the linseed fibres and gave high strength to the composite when mixed with hydrated lime. The composites made with fibres pre-treated with yeast and those coated using CaO showed lower strengths.

**Keywords:** *Lime composite, Linseed, SEM, XPS, Compressive Strength*

## 1. Introduction

Natural fibres have been used in lime based building materials for thousands of years, an important example being the addition of reeds to the lime mortars used in the construction of cisterns in Herod's palace at Masada (Allen et al. 2008 and Radonjic et al. 2001). More recently hemp shives have been used to produce lightweight hydraulic lime composites where a major application is non-load bearing walls in timber frame building.

The growing interest in these materials has encouraged the application of modern analytical techniques to historic and modern natural fibre-mortar composites. Examples of recent studies include the application of Environmental Scanning Electron Microscopy (ESEM) and Focused Ion Beam (FIB) imaging and sectioning to characterise the morphology of composite materials consisting of organic plant fibres embedded in calcium aluminate cement matrices and lime mortars (El-Turki et al. 2008). FIB techniques were found to be an effective method for sectioning hemp fibres encapsulated in a cementitious matrix (El-Turki et al. 2005).

Natural fibres such as linseed and hemp are often subjected to bacterial/fungal and moisture attack during and after cultivation. Such attack is beneficial in the 'retting' process of bast-fibre separation where the majority of cellular tissues are removed.

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The mechanical properties of a fibre composite material are determined by the nature of the matrix-fibre bond. The ability to enhance this bond may provide an effective method of improving the strength of a resulting composite. The separation of fibre bundles results in an increase in matrix-fibre surface area leading to enhanced mechanical bonding. The ability of the surface of organic fibres, which consist mainly of cellulose, to bond with building limes (either hydraulic or non-hydraulic) may be enhanced by the formation of a calcium hydroxide or carbonate surface layer between the fibre and matrix, which is more similar in chemical structure.

In this study, the effect of fibre pre-treatment on the mechanical properties of lime matrix composites has been investigated. Mechanical properties, microstructure and the fibre/matrix bond have been characterised using Scanning electron microscopy, x-ray photoelectron spectroscopy, Infrared spectroscopy and compressive testing.

## 2. Experimental

### 2.1. Fibre pre-treatment and sample manufacture

Linseed fibres (*Lignum Usitatissimum*, variety *Taurus*), supplied by DeMontfort University, were pre-treated as follows. The 'as received' fibres were cut into lengths between 5 and 10 mm prior to soaking for 10 days in either pure water or water containing the *saccharomyces cerevisiae* variety of yeast which acts in the most common types of fermentation. Following soaking, the fibres were coated with lime; it should be noted that the coating processes described below were applied to damp fibres immediately after removal from the soaking liquid. Coating was achieved by either; (i) stirring for 24 hours in a 1:3 mixture of lime putty and water before drying in the laboratory atmosphere for a further 24 hours, or (ii) mixing with an excess of calcium oxide (quicklime) and applying a pressure of approximately 1 Pa for 24 hours. Both processes described resulted in dry fibres which were subsequently rubbed lightly by hand over a 2.5 mm mesh sieve to remove excess lime. The fibre strands were then teased apart by hand before being incorporated into a lime matrix composite. Both the lime putty and quicklime were supplied by Singleton Birch Ltd., England.

The resulting fibres were used to manufacture a 1:1 fibre to lime volume ratio composite material with a matrix of either CL90 or NHL5, both supplied by Hanson Cement Ltd. The fibre and lime were mixed by hand, in a dry state for around 3 minutes prior to the addition of 60 % de-ionised water by volume. Three cylindrical specimens measuring 18 mm in diameter and 36 mm in height were cast from each mixture in a purpose made mould. The 1:2 aspect ratio is the standard for specimens of this shape (Bevan and Woolley 2008). Specimen mixes are detailed in Table 1. The specimens were removed from the mould after 7 days and cured in the laboratory environment of temperature 20~24 °C, relative humidity ~50 % and CO<sub>2</sub> concentration ~400 ppm for 28 days.

**Table 1** : Linseed mix designs

<b>Matrix</b>	<b>Linseed pre-treatment</b>	
	<b>Soaking</b>	<b>Coating</b>
CL90	Water	Lime Putty solution
CL90	Water and yeast	Lime Putty solution
CL90	Water	CaO powder
CL90	Water and yeast	CaO powder
NHL5	Water	Lime Putty solution
NHL5	Water and yeast	Lime Putty solution

NHL5	Water	CaO powder
NHL5	Water and yeast	CaO powder

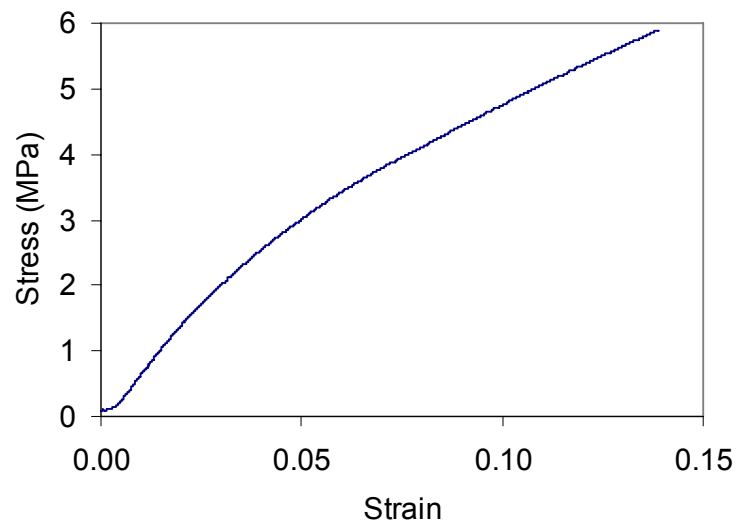
## 2.2 Characterisation techniques

The force and displacement of each specimen when compressed was determined using a Zwick Roell Z010 testing machine. A  $20 \text{ N s}^{-1}$  loading rate was applied to the specimens up to a maximum displacement of 5 mm, equivalent to a 14 % strain. The elemental composition of the material on the fibre surface in each specimen was determined using a Kratos XSAM800 X-ray photoelectron spectrometer equipped with an Al  $K_{\alpha}$  (1486.6eV) x-ray source. Spectra were obtained using an X-ray power of 240W, emission current of 20 mA and acceleration potential of 12 kV. The atomic percentage was calculated for each specimen from spectra analyzed in the narrow scan mode which used a step size of 0.1 eV and duration of 3 s per step. Fourier transform infrared spectroscopy (FTIR) was carried out using a Perkin Elmer Spectrum 100 spectrometer within the wave range 650 to 4000  $\text{cm}^{-1}$ . Spectra were generated from an average of 4 acquisitions. Microstructural analysis of linseed fibre surfaces from each mix design were obtained using a Hitachi S2300 scanning electron microscope operating at an accelerating voltage of 20 kV. Specimen charging was reduced by gold coating using an Edwards Scancoat Six sputter coater for 90 s prior to insertion into the SEM.

## 3. Results

### 3.1 Mechanical properties

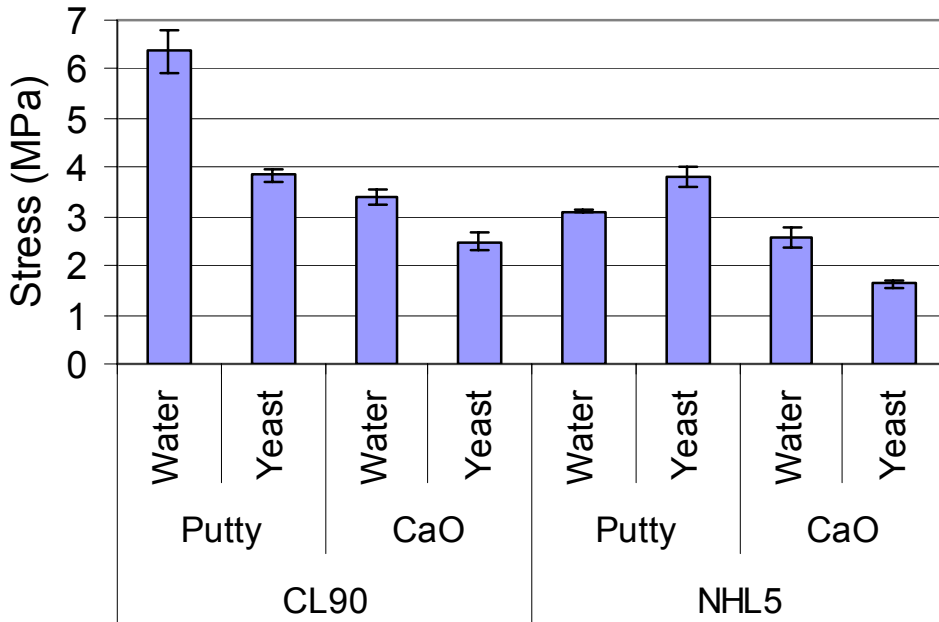
A stress versus strain curve of the specimen comprising a CL90 matrix and linseed fibres pre-treated by soaking in pure water and mixing with a lime putty solution is shown in Figure 1. The behaviour show is typical of all the composites tested. The test was terminated at a displacement of 5 mm which corresponded to a strain of 14 %. No maximum stress corresponding to failure was observed and this was the case for all the samples tested. This indicated that the specimens were too flexible to fracture.



**Figure 1:** Typical stress - strain curve for CL90 matrix composite containing linseed fibres soaked in water and coated using lime putty solution

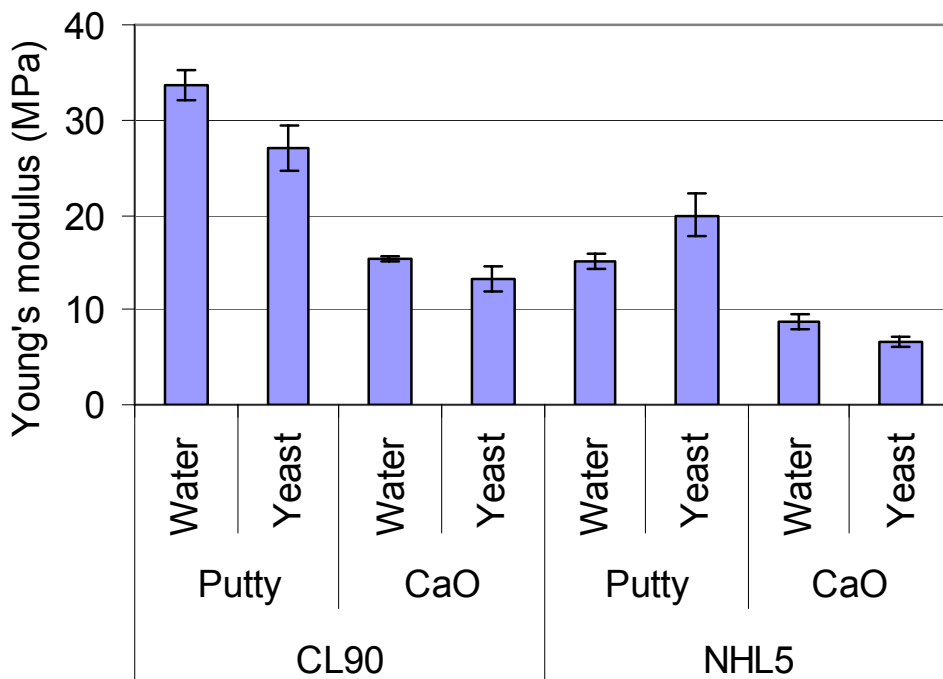
The average stress at 14 % strain for each mix design is shown in Figure 2, where bars represent the standard error. The specimens made with CL90 were stronger when

compared with the equivalent sample made with NHL5. The CL90 specimen with the fibres pre-treated with water and lime putty showed the highest strength of all the specimens. The specimens with the fibres pre-treated in lime putty showed overall higher strength than the specimens pre-treated with CaO. All the specimens with fibres soaked in yeast containing water, apart from the NHL5 specimen treated using lime putty, were weaker than the ones soaked in pure water.



**Figure 2:** Stress exerted by composites at a strain of 14 %

The stiffness of the specimens was investigated by calculating Young's modulus in the extension region 0.2 mm to 1 mm. The average modulus for each specimen type is shown in Figure 3.



**Figure 3:** Young's Modulus calculated within the extension region 0.2 mm to 1 mm for each composite type.

The composite materials containing a CL90 matrix showed greater stiffness and amongst these those with fibres pre-treated with water and lime putty performed best. The difference in Young's modulus between CL90 specimens pre-treated with 'water & putty' and 'yeast & putty' was less than the difference in their strengths. The overall trend for the Young's modulus was in good agreement with the compressive stress data.

### 3.2 X-ray photoelectron spectroscopy (XPS)

Wide scan survey spectra performed on the specimens revealed peaks at binding energies of 285, 347 to 349, 198 to 200 and 531 to 533 eV corresponding to signals from carbon ( $C_{1s}$ ), calcium ( $Ca_{2p}$ ), chlorine ( $Cl_{2p}$ ) and oxygen ( $O_{1s}$ ) respectively. An additional peak around 398 eV was observed in the narrow scan spectra which corresponded to the nitrogen ( $N_{1s}$ ) photoelectric peak. A weak peak at 531 eV was attributed to chlorine contamination.

Atomic percentages for each element were calculated from the narrow scan spectra using Pisces software (version 2006.0 Dayta Systems Ltd.) and its supplied relative sensitivity factors. The normalized atomic percentages are shown in Table 2.

**Table 2 :** Normalized atomic percentages obtained from XPS data

<b>Matrix</b>	<b>Fibre coating</b>	<b>Fibre soaking</b>	<b>Carbon</b>	<b>Calcium</b>	<b>Chlorine</b>	<b>Nitrogen</b>	<b>Oxygen</b>
CL90	Lime Putty	Water	36.03	8.27	6.53	2.04	47.13
		Yeast	30.88	16.41	5.96	2.07	44.68
	CaO	Water	39.73	9.00	3.20	2.14	45.92
		Yeast	40.01	8.24	5.83	3.24	42.68
NHL5	Lime Putty	Water	35.63	11.28	2.54	1.12	49.43
		Yeast	35.28	6.20	3.13	2.76	52.62
	CaO	Water	41.89	7.09	4.49	2.84	43.69
		Yeast	40.34	9.34	5.60	3.26	41.46

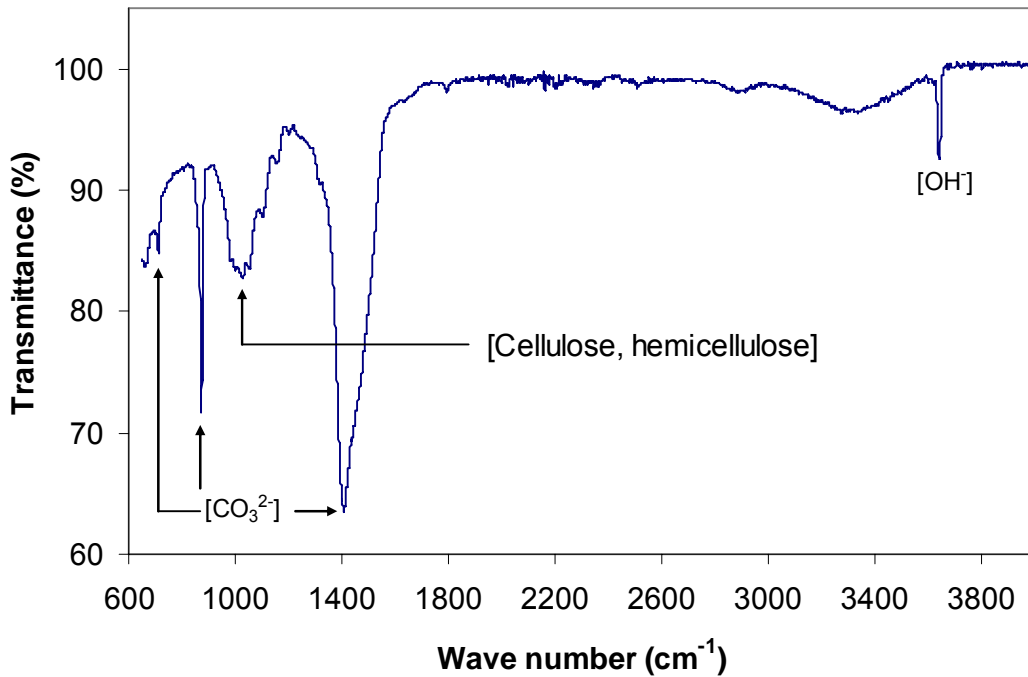
No direct correlation was observed between the atomic percentages and specimen strength; however a number of qualitative observations were made. Specimens mixed with CaO had a higher atomic percentage of carbon in comparison to their lime putty solution treated counterparts. This could be the effect of heating of the outer fibre surface caused by the elevated temperatures produced during the slaking of quicklime.

It was noted that the atomic percentages of oxygen were higher in the specimens containing water impregnated fibres compared to those pre-treated with a yeast solution. The only exception to this observation was the NHL5 specimen in which the fibres were coated with lime putty. This trend corresponded to the strength and stiffness of the specimens, where the specimens containing the yeast treated fibres exhibited lower strength/stiffness compared to those containing the water soaked fibres. This observation is consistent with a higher degree of carbonation (El-Turki et al. 2006).

### 3.3 Fourier transform Infrared spectroscopy (FTIR)

A typical FTIR spectrum taken from the CL90 specimen with the fibres pre-treated with water and lime putty is shown in Figure 4. Peaks at 1404, 872 and 711  $cm^{-1}$  are attributed to calcium carbonate (Zeng et al. 2008). The peak located at 3643  $cm^{-1}$  is attributed to the

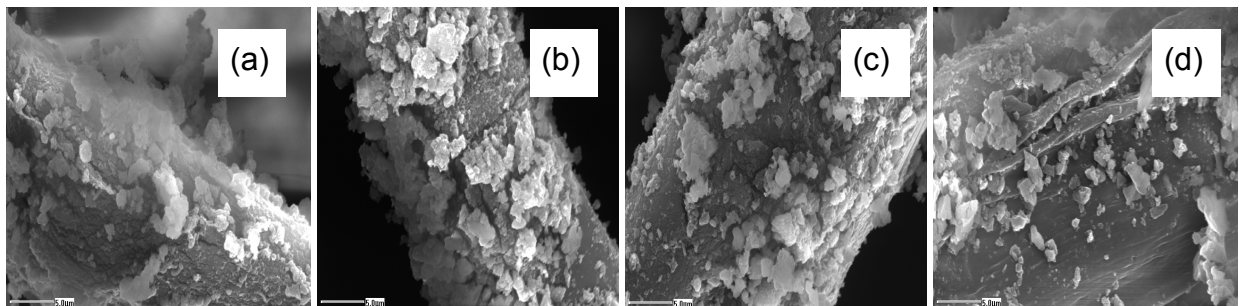
OH<sup>-</sup> stretching frequency and those located around 1026 cm<sup>-1</sup> are assigned to C-O-C asymmetrical stretching in cellulose and hemicellulose within the linseed fibre (Troedec et al. 2009).



**Figure 4:** Typical FTIR spectrum taken from CL90 specimen containing linseed fibres soaked in water and coated with lime putty solution

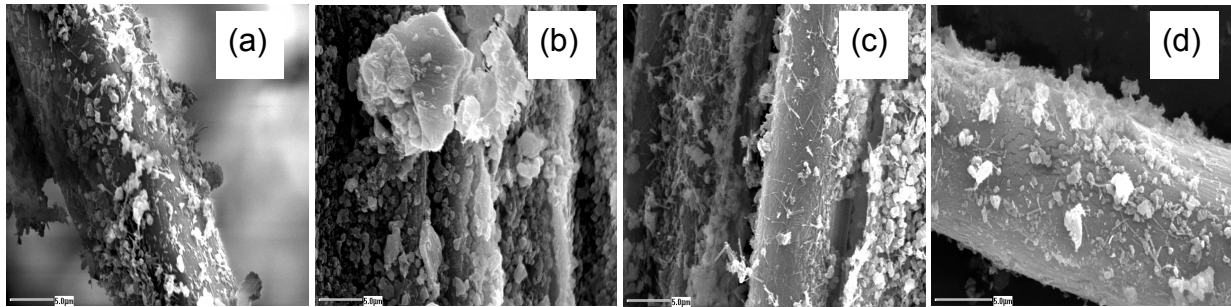
### 3.4 Scanning electron microscopy (SEM)

The SEM images of fibres extracted from each specimen are shown in Figure 5 and 6. Calcium hydroxide and calcium carbonate were observed on the surface of the fibres.



**Figure 5:** SEM images of fibres in CL90 matrix; fibres soaked and coated in (a) water and lime putty (b) yeast water and lime putty (c) water and CaO (d) yeast water and CaO

The surface of the fibres extracted from specimens with a NHL5 matrix contained calcium hydroxide and various amounts of high aspect ratio needle shaped crystals, as shown in Figures 6 (a), (c) and (d). These resembled calcium silicates observed by El-Turki et al. (2009). The NHL5 specimen made with the fibres pre-treated in yeast and lime putty (Figure 6 (b)) did not appear to have as many silicates compared with other NHL5 specimens



**Figure 6:** SEM images of fibres in NHL5 matrix; fibres soaked and coated in (a) water and lime putty (b) yeast water and lime putty (c) water and CaO (d) yeast water and CaO

#### 4. Discussion

None of the stress-strain curves obtained from the composites exhibited a maximum peak stress, usually associated with failure, within the 14 % strain range. The stress-strain behaviour of these composites suggested that their application in building would be limited to non-load bearing components where strains of up to 10% would be acceptable (Bevan and Woolley 2008). Examples may include those applicable to lime hemp in blocks, cast walls and renders. Despite the absence of a peak in the stress strain curve, the force at the end of the test, 14 % strain, varied significantly depending on fibre pre-treatment.

The Young's modulus calculated at the beginning of the compression process was in good agreement with the compressive stress.

Soaking the fibre in yeast water and coating with CaO reduced the strength of the composites compared to their water-soaked, lime putty coated counterparts. It was not possible to identify the structural or morphological changes responsible for this effect by scanning electron microscopy. However the more aggressive soaking environment attributed to the yeast and elevated temperatures reached during treatment with CaO may have weakened the fibre structure making it less rigid.

SEM examination showed that the surface of fibres taken from CL90specimens maintained a well bonded and uniform coverage of calcium hydroxide and calcium carbonate. The fibres pre-treated with lime putty appeared to be bonded more strongly with calcium hydroxide in CL90 than in NHL5. SEM images for the NHL5 specimen with the fibres pre-treated in yeast water and lime putty showed less silicate growth than the rest of the NHL5 specimens. Treatment with yeast may have modified the fibre surface leading to the preferential growth of calcium hydroxide and calcium carbonate over the silicate phase which in turn led to the higher strength of the specimen. However the silicate structure was easily observed in the NHL5 specimen with the fibres pre-treated with yeast water and CaO. This suggests that the effect of the yeast is preserved when the fibres are treated with CaO but eliminated when soaked in lime putty solution.

#### 5. Conclusions

The following conclusions have been drawn from the results;

- The specimens with a CL90 matrix were stiffer and stronger than the NHL5 specimens. Among the CL90 specimens, those containing fibres pre-treated with water and lime putty showed the highest strength/stiffness.
- The NHL5 specimens pre-treated with water containing yeast and lime putty showed similar strength, but lower stiffness compared to the CL90 specimens pre-treated in the same way.



- All the specimens made from the fibres soaked in water containing yeast were weaker than those manufactured from the water impregnated fibres. The only exception to this was the sample with a NHL5 matrix containing fibres pre-treated with yeast and lime putty.

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