

## DEVELOPING WASTE CELLULOSE FIBRE REINFORCED COMPOSITES WITH CLINKER FREE CEMENT

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### ABSTRACT

Developed countries have achieved high performance wood fibre reinforced cement products by adopting elaborate technologies. On the other hand, researchers in developing countries concentrated mostly on the use of natural strand reinforcement and simple production methods resulting in serious concerns regarding durability. As consequence asbestos based composites remained widely used in Brazil, a market of more than two million tons/year of fibre-cements, mainly in the form of corrugated roofing elements. As growing concerns about health hazards are leading to asbestos (chrysotile) bans in several countries, research is now underway for the adaptation of vegetable fibre in fibre-cements using locally available raw-materials and production systems and meeting consumer requirements in each particular application area. The main objective of the present paper is to discuss the performance of several non-conventional materials based on cementitious matrices reinforced with cellulose pulps. Alternative binders involve clinker free cement based on blast furnace slag (BFS) ground at three different Blaine fineness, as well as ordinary Portland cement (

OPC). Fibrous resources include eucalyptus (*Eucalyptus grandis*) residual pulp, banana (*Musa cavendishii*) kraft and sisal (*Agave sisalana*) kraft, both produced in laboratory scale. Composite preparation followed a slurry vacuum de-watering process, pressing and air-curing. Fibre content varied from 8 to 12% by mass, the region of optimum mechanical behaviour as indicated in previous studies. The fibre-cements are evaluated by means of their mechanical and physical behaviour at age 28 days. The results of this ongoing work indicate that the performance of the composites is currently acceptable for low-cost building materials. As a final remark, potential solutions for housing and infrastructure can be achieved throughout the adaptation of already known technologies to the peculiarities of developing countries.

**KEYWORDS:** composites, cellulose, cement, clinker-free cement, blast furnace slag, slurry vacuum de-watering

## 1. INTRODUCTION

Developed countries have achieved high performance wood fibre reinforced cement products by adopting elaborate technologies such as pressing and autoclave curing. On the other hand, researchers in developing countries concentrated mostly on the use of natural strand reinforcement and simple production methods resulting in serious concerns regarding durability. As consequence asbestos based composites remained widely used in Brazil, a market of more than two million tons/year of fibre-cements, mainly in the form of corrugated roofing elements. As growing concerns about health hazards are leading to asbestos (chrysotile) bans in several countries, research is now underway for the adaptation of

vegetable fibre in fibre-cements using locally available raw-materials and production systems and meeting consumer requirements in each particular application area.

Natural vegetable fibres, like sisal, banana pseudo stem, coir, etc. are widely available in developing countries [1]. These macroscopic vegetable fibres are complex natural composites. Each macro-fibre is made with several micro fibres or cells. Micro fibres are constituted by four different layers each of it is have different proportions of cellulose, hemi-cellulose, lignin and extractives. The micro fibres are glued together by the middle lamella, constituted mainly by lignin.

It has been proved that ordinary Portland cement reinforced with natural fibres loses its ductility with ageing [2, 3]. One possible explanation for this degradation is the dissolution of the lignin from the middle lamella by the cements alkaline pore water [2]. The use of low alkaline cements has been proposed in order to prevent fibre degradation, among other solutions [2,4].

Cellulose production process, or pulping process, is mainly a strong alkaline attack. The main objective of these processes is removing lignin. When almost all lignin of the middle lamella is already removed by the chemical attack, the cells, constituted mostly by small cellulose, are liberated [5]

Since the pulping process removes the most soluble part of the lignin, the resulting cellulose fibres are more resistant to alkaline attack than the natural fibres. From this point of view, they are probably more suited to be used with alkaline cements. Cellulose reinforced cement is known since the Second World War but it became a commercial technology since the beginning of the 1980's [6]. Contents of up to 12% composite's weight are usual. Additionally, due to its geometry and filtration capacity, cellulose fibres can be used in the production of thin fibre-reinforced cement sheets in replacement of asbestos fibres, without replacing the Hatchek machines [6].

Even if cellulose fibre are more alkaline resistant than natural vegetable fibres, they still can be attacked by the alkaline cement pore water. So, low-alkaline cements still desirable. Oliveira [7] carried on a comprehensive investigation of pore water composition of OPC and ground granulated blast furnace slag (BFS) based cement. It has been found that BFS activated with 2% of lime and 8% of natural gypsum [4] was the one with the lowest alkaline water pore, with a pH around 11.5 against typical values 12.7 for commercial OPC [7].

This paper presents some basic investigation carried on by University of São Paulo, Brasil, partially carried on with collaboration of CSIRO, Australia, to develop a more durable and cheap cellulose fibre reinforced cement aiming to substitute asbestos reinforced composites.

## **2. MATERIALS AND METHODS**

### *2.1. Experimental design*

#### *2.1.1. Comparing alternative waste fibres*

The performance of composites made with residual *Eucalyptus grandis* Kraft pulp and laboratory made pulps from residues of natural fibres of sisal (*Agave sisalana*) and banana (*Musa cavendishii* nanicao variety) pseudo-stem were compared. Australian commercial OPC and BFS clinker free binder were used as matrixes. The content of sisal and banana pulp fibre was 8% (w/w). Since the Eucalyptus pulp presented a lower aspect ratio and higher fines content, the selected fibre amount was 12% (w/w).

The samples produced by slurry vacuum de-watering technique were immediately pressed to 3,2MPa for 5 minutes. After that they were wet cured in sealed plastic bags at room temperature, for 7 days time period. After this period, they were removed from the bags and diamond sawn into three 125 x 40 mm test specimens. The specimens were conditioned in the laboratory conditions environment until they were tested at age 28 days. Specimen's depth was the thickness of the pad, which was in the central region about 6 mm.

### 2.1.2. Optimisation of BFS clinker free binder

The effect of the variation of Blaine fineness and curing on composite short-term performance was investigated. Three levels of Blaine fineness (250, 500 and 820m<sup>2</sup>/kg) and three levels curing times (7, 14 and 25 days) inside of sealed plastic bags. A additional series of samples, with Blaine fineness of 820m<sup>2</sup>/kg, was cured in plastic bags for only two days and subsequently immersed in lime-saturated water at room temperature until it completed 7, 14 and 25 days.

After the curing period all samples were store in laboratory condition until they are tested at age 28 days. The samples were produced as described previously.

## 2.2. Materials

### 2.2.1. BFS clinker free binder

The BFS clinker free binder was defined in previous studies by Agopyan and John [4] and Agopyan et al. [8]. It is a low alkaline, low energy cement and can be formulated using only residues. It is made with 88% BFS, 10% of CaSO<sub>4</sub>.2H<sub>2</sub>O and 2% of hydrated lime. All ingredients were dry mixed before water addition.

#### BFS

BFS, provided by Companhia Siderúrgica Tubarão (CST), Brazil, was used. It was grounded, using a laboratory ball mill, to the desired Blaine fineness (250, 500 or 820m<sup>2</sup>/kg) and stored in sealed plastic bags. Chemical composition follows at Table 1. Glass content was 97%.

Table 1. BFS chemical composition (% by mass)

Loss on ignition	1.67	SiO <sub>2</sub>	33.78	CaO	42.47
Free CaO	0.1	Al <sub>2</sub> O <sub>3</sub>	13.11	MgO	7.46
Insoluble residue	0.53	K <sub>2</sub> O	0.32	SO <sub>3</sub>	0.15
Fe <sub>2</sub> O <sub>3</sub>	0.51	S <sup>2-</sup>	1.14	Na <sub>2</sub> O	0.16

#### Gypsum

Commercial natural ground gypsum, agricultural grade, from Araripina (PE) was used in optimization of BFS binder studies. Its main chemical composition was 88,9 % CaSO<sub>4</sub>.2H<sub>2</sub>O and 7,12% CaSO<sub>4</sub>.

Natural ground gypsum, agricultural grade, supplied Garden King, Australia (calcium as calcium sulphate, 18.5%w/w; sulphur as calcium sulphate, 14.5%w/w) was used in the experiments aiming to compare different alternative fibres.

#### Lime

Hydrate lime (CH I type, NBR-7175), supplied by Supercal (Brazil), was used in the binder optimisation. Its chemical composition was 90% of Ca(OH)<sub>2</sub> and 5,6% do CaCO<sub>3</sub>.

For the study of that compares different waste fibres an Australian commercial hydrated lime (AS 1672), supplied by Adelaide Brighton was used.

### 2.2.2. OPC binders

OPC CII-F 32, Itaú brand, was used in the binder optimisation. Adelaide Brighton brand OPC, Type GP (AS-3972-1991), was used as the reference matrix when comparing the alternative waste fibres.

### 2.2.3. Fibres

The residual *Eucalyptus grandis* kraft pulp was supplied by Aracruz Celulose Espírito Santo State, Brazil. It was partially bleached and used as received after a simple disintegration in tap cold water during 2 min.

Pulps from sisal (*Agave sisalana*) and banana (*Musa cavendishii*, nanicao variety) pseudo-stem fibres were produced by chemi-thermomechanical (CTMP) pulping process [9]. Slivers of by-product sisal and banana were initially chopped to 30 mm in length and soaked for at least 16 hours in tap water. After soaking the strands were boiling in a 10% lime liquor for 1 h. The strands were then mechanically defibred in an Asplund Type D laboratory defibrator and refined with a 20 cm Bauer laboratory disc refiner fitted with straight-patterned “rubbing” discs. Both pulps were passed through a Packer screen (0.23 mm slots) to separate shives and through a Somerville screen (0.180 mm mesh) to reduce fines content. Finally, the pulps were vacuum de-watered, pressed, crumbed and stored in sealed plastic bags under refrigeration until they were used.

The main physical attributes of the pulps are summarised in Table 2. The Canadian Standard Freeness (CSF) was measured following AS-1301.206s-88. Fibre length and fines content were determined using a Kajaani FS-200 automated optical analyser.

Table 2. Pulp and fibre physical properties

Fibre	CSF (ml)	Fines (%) <sup>1</sup>	Length (mm) <sup>2</sup>	Width (µm) <sup>3</sup>	Aspect ratio
E. grandis	685	7.01	0.66	10.9	61
Sisal	500	2.14	1.53	9.40	163
Banana	465	1.55	2.09	11.8	177

<sup>1</sup> Arithmetic basis, <sup>2</sup> length-weighted basis, <sup>3</sup> average of 20 determinations by SEM

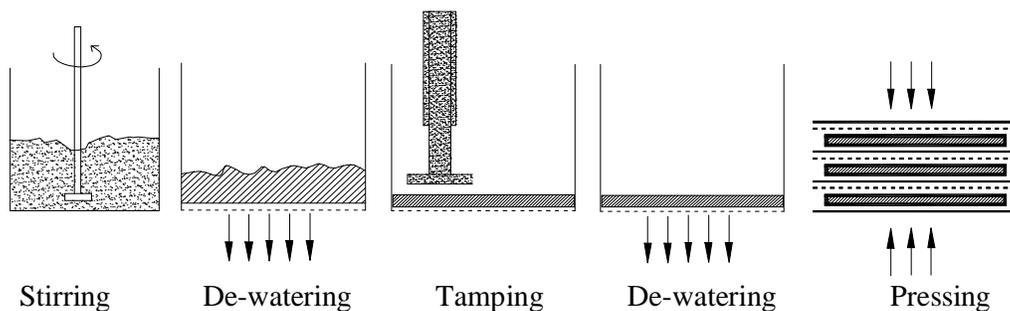


Figure 1 -Steps of pad preparation.

### 2.3. Composite Production

Figure 1 summarizes the steps of composite production. Binder was added to the appropriate amount of moist fibres, pre-dispersed in water, to form a slurry with 20% (w/w) of solids. After stirring for 5 minutes using a high-speed laboratory mixer, the slurry was rapidly transferred to an evacuable 125 x 125 mm casting box. An initial vacuum of up to 80 kPa (gauge) was applied until the bulk of the excess water was removed and a solid surface

formed. The moist pad was tamped flat and vacuum reapplied for 2 minutes. The consolidated pad was then removed from the casting box, transferred to an oiled steel plate and a fine wire mesh placed on top. Three pads per composite formulation were prepared in this manner, stacked on top of each other and submitted to pressing of 3.2 MPa for 5 minutes.

#### 2.4. Mechanical and Physical Tests Methods

The flexural properties of the materials were measured 28 days after production. A three-point bend configuration, span of 100 mm and a deflection rate of 0.5 mm/min, was used for the determination of flexural strength (MOR), modulus of elasticity (MOE) and fracture energy properties. The fracture energy was calculated by integration of the load-deflection curve to the point corresponding to a reduction in load carrying capacity to 50% of the maximum observed, divided by the transversal area. Nine flexural specimens were tested for each composite formulation and test condition.

Water absorption, values at 28 days were obtained from tested flexural specimens following the procedures specified in ASTM C 948-81. Six specimens were used in the determination of each of these physical properties.

### 3. RESULTS AND DISCUSSION

#### 3.1. Comparison between different residual pulps

Table 3 depicts the mechanical and physical properties of the various composites. Non-aged composites presented modulus of rupture (MOR) in excess of 18 MPa, representing a 120% improvement over a plain BFS matrix of similar formulation.

Table 3 – Effect of different fibres on composite performance (average +/- standard deviation)

Fibre		Binder	MOR (MPa)	FT (kJ/m <sup>2</sup> )	MOE (GPa)	Water absorption % w/w
Type	% w/w					
Nil	-	BFS	8.1 ± 2.2	0.03 ± 0.01	11.6 ± 1.7	17.6 ± 0.9
<i>E. grandis</i>	12	BFS	18.2 ± 2.8	1.25 ± 0.20	5.0 ± 0.6	32.3 ± 1.7
		OPC	22.2 ± 1.3	1.50 ± 0.18	8.0 ± 1.1	24.8 ± 0.8
Sisal	8	BFS	18.4 ± 1.4	0.85 ± 0.10	5.9 ± 0.5	32.9 ± 0.6
		OPC	21.7 ± 1.1	0.79 ± 0.17	9.9 ± 0.3	22.9 ± 1.2
Banana	8	BFS	18.9 ± 1.9	0.51 ± 0.10	6.2 ± 0.6	31.7 ± 0.6
		OPC	20.9 ± 2.0	0.51 ± 0.10	9.8 ± 0.5	23.6 ± 0.9

Fracture toughness (FT) is also greatly increased by the presence of fibres, and resulted in a minimum 17-fold increase. *Eucalyptus grandis* composites showed better results than the others in the initial age of 28 days. This could be explained by the higher content of eucalyptus used. But *Eucalyptus* fibres' lower anchorage length (Table 8) could also explain the difference. BFS based composites presented modulus of elasticity (MOE) values between 5.0 and 6.2 GPa, approximately 50% of that of the plain BFS matrix. The reduction is associated with the porosity of the cellulose fibres.

All combinations tested resulted in suitable short-term performance composites. Nevertheless the residual eucalyptus fibre seems to lead to be better than the other ones, because it leads to better short-term performance and because it a residue that does not require any industrial

processing. Around 17,000 metric tonnes of this fibre is generated by Aracruz Celulose each year and sold at a price of about US\$15/ton [10].

### 3.2. Effect of BFS fineness and curing method

Increasing the wet curing period above 7 days seems not to improve the MOR when the binder is formulated using slag with Blaine fineness above 500m<sup>2</sup>/kg (Figure 2) and causing a significant reduction of fracture toughness. By the other side, for the samples produced with slag grounded to fineness of 250m<sup>2</sup>/kg, it seems to cause an increasing in both MOR and FT. For finely ground BFS the increase of degree of hydration due to an increase in the curing period probably causes only fibre-matrix transition zone densification [11], reducing the fibre pullout process and negatively affecting FT. Since the resulting pore volume reduction is probably not big enough to be measured by the bending test, MOR was not significantly affected.

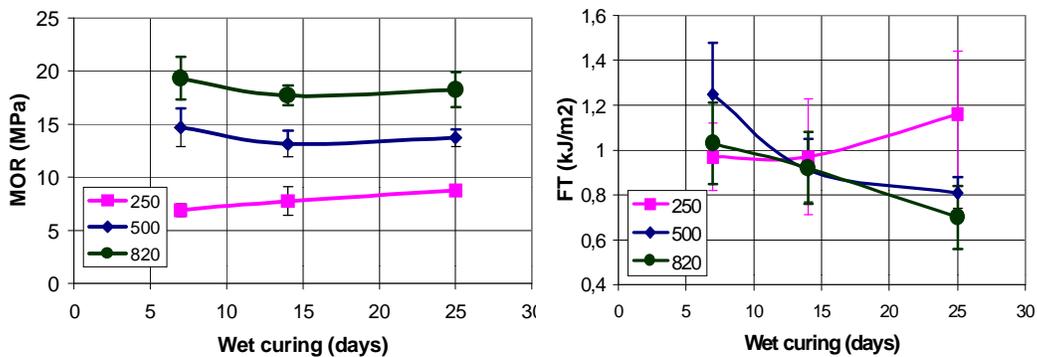


Figure 2 – Effect of wet curing time on MOR (left) and FT (right). The vertical lines indicate the standard deviation of each sample. All samples are 28 days old.

Figure 3 show that increasing the fineness of the slag increases the MOR of 7 days wet cured sampled, but does not significantly affect its ductility. This conclusion applies to all curing treatments investigated (Figure 2), with the exception of samples produced with 250kg/m<sup>2</sup> slag.

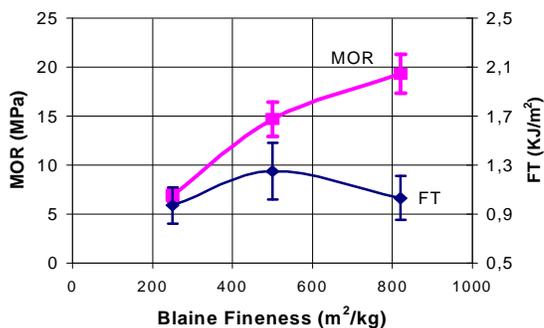


Figure 3 –Effect of fineness on MOR and FT for samples 28 days old, submitted 7 days wet curing inside sealed plastic bags. The vertical lines show the standard deviation of each sample.

Figure 4 shows MOR and FT of samples cured both wet (inside sealed plastic bags) and water immersed in lime-saturated water. Water curing seems to give better modulus of rupture, but toughness was no significantly affected. Previous results had shown that the two days curing

inside the sealed plastic bags are very important previously water curing in order to prevent composites to swell. Then, water curing is a additional activity and increases the work required to produce samples.

Because long curing times are expensive in a full-scale production and grinding must be minimized, slag fineness of  $500\text{m}^2/\text{kg}$  combined with 7 days wet curing inside plastic bags, seems to be the most appropriate combination for the production of composites using this BFS clinker-free cement.

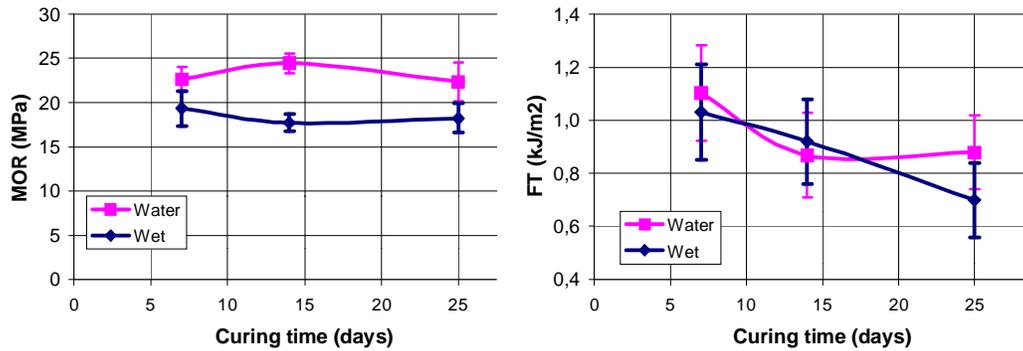


Figure 4 – Effect of the total curing type and period over modulus of rupture (left) and fracture toughness (right). Curing time for water-cured samples includes two days inside plastic bags (wet curing). Slag ground to  $820\text{m}^2/\text{kg}$ .

## 4. CONCLUSIONS

The residual eucalyptus fibre seems to be the better option to produce cellulose reinforced cement sheets for two reasons. First, it allows producing composites with the better ductility than the other two fibres. Second, it is a Kraft pulp residue and can be used without any significant processing while the two others need to be pulped.

Low alkaline slag based binder seems also to be a feasible option. It results in almost the same mechanical performance at 28 days than the OPC. Also, presents environmental advantages, since it can be fully manufactured using only residues, slag, residual lime and a residual gypsum like phosphogypsum. Because it is low alkaline, it can present better results in long-term performance as well.

Because long curing times are expensive in a full-scale production and grinding must be minimized, slag fineness of  $500\text{m}^2/\text{kg}$  combined with 7 days wet curing inside plastic bags, seems to be the most appropriate combination for the production of composites using this BFS clinker-free cement.

The use of cellulose-reinforced cement can become a potential solution for housing and infrastructure for developing countries

## 5. ACKNOWLEDGMENTS

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