PERFORMANCE OF LOW-COST VEGETABLE FIBRE-CEMENT COMPOSITES UNDER WEATHERING

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ABSTRACT

Developed countries have achieved high performance wood fibre reinforced cement (WFRC) products by adopting elaborate technologies with high energy consumption processes. In attempting to reduce costs, researchers in developing countries have mainly concentrated on the use of natural strand reinforcement and simple production methods. Serious concerns have arisen regarding the durability of these lower technology products and consequently asbestos-based composites remain widely used in countries such as Brazil which has 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 more countries, research is now underway for the adaptation of vegetable fibre in fibre-cements manufactured using locally available raw materials and production systems and meeting consumer requirements in each particular application area.

The main purpose of this current collaborative work was to evaluate Australian technology - developed in the early 80's to enable the manufacture of WFRC products by the Hatschek (or wet) process - for use with Brazilian recycled raw materials in the fabrication of thin cellulose-cement sheeting. Granulated blast furnace slag (BFS), a glassy material obtained as a by-product of pig iron manufacture, was ground and used as the major component of an alternative hydraulic binder activated by a mixture of gypsum and hydrated lime. Mechanical pulps from sisal (Agave sisalana) and banana (Musa cavendishii) crop wastes and also residual kraft Eucalyptus grandis from a wood pulp mill were chosen as reinforcements on the basis of their ready availability and acceptable performance in cement. Composites were prepared using a slurry vacuum de-watering process followed by pressing and air-curing. Fibre content varied from 8 to 12% by mass, the region of optimum mechanical behaviour indicated in previous studies.

Mechanical testing revealed flexural strengths in excess of 18 MPa, moduli of elasticity between 5 and 6.2 GPa and fracture toughness values in the range 0.51 to 1.25 kJ/m² at an age of 28 days. Water absorptions of the composites were about 32% by mass and bulk densities 1.3 g/cm³. Composites were then subjected to natural ageing by external exposure in tropical (Pirassununga, Sao Paulo, Brazil) and temperate (Melbourne, Victoria, Australia) environments. Corresponding series were kept as references in controlled ambient conditions (23 ± 2°C and 50 ± 5% relative humidity). After twelve months of external exposure the flexural strengths of weathered composites had decreased to values in the range 6.6 - 10.1 MPa. Fracture toughness values, however, had either remained stable or had significantly increased (over 105% for 8% banana pulp in BFS matrix) relative to those at 28 days of age. The absence of decreases in ductility suggests that the fibres are still effective and largely free from either alkali attack or inner petrifaction. Evidence to the contrary was not apparent during microscopic examination of fracture surfaces. The results of this ongoing work indicate that the performance of the composites is currently compatible for low-cost building materials in developing countries.

KEYWORDS:

Blast furnace slag; vegetable residues; cellulose fibres; durability; alternative fibre-cements.
INTRODUCTION

The interest in vegetable fibres as substitutes for asbestos in low-cost building applications lies mainly in their competitive prices and renewable nature. However, the use of natural fibres in building materials has some disadvantages, such as low modulus of elasticity, high water absorption, a propensity to decompose in alkaline environments or under biological attack, and variability in mechanical and physical properties (Swamy, 1990). Several studies in developing countries examining alternative fibre-cement products (e.g. Agopyan, 1988) have concentrated on the use of strand fibres and simple production processes, giving rise to concerns about durability. Strands are assemblages of individual fibres bound by lignin and hemi-cellulose layers and these are readily decomposed by the alkaline pore solution of ordinary Portland cement (OPC). Strands are also stiff and generally cause mixing difficulties at volume fractions over 4%, with heterogeneous reinforcement distribution and poor packing in the composites resulting.

Pulping is the conventional procedure used in the industrial preparation of wood and other vegetable fibres for use in fibre-cements (Coutts, 1992). Beneficial aspects to the use of pulped fibres include their ability to form a network that assists in cement retention during sheet formation, good adhesion and packing in the matrix and greater resistance to alkaline attack when compared to the original strands or chips. Chemical pulps prepared by the kraft (or sulphate) process are preferred but are mainly produced for the paper industry and fetch prices of up to US$ 800/tonne (Ausnewz, 1999). This translates to significant costs for fibre-cement products. Despite its high mechanical energy consumption, and incomplete removal of non-fibrous elements, mechanical pulping possesses some advantages over chemical pulping. As noted by Coutts (1986), chemical requirements are lower, effluent treatment and disposal are less troublesome, and mills can be economically viable at a smaller scale. Pulp prices can be around half those of the corresponding kraft pulp.

Granulated blast furnace slag (BFS) can provide advantages when used as a partial or total replacement of conventional Portland cement. These may include energy savings, cost reductions and abundant availability in steel production areas. The BFS hydration process involves consumption of free calcium hydroxide. This is of particular interest in cellulose fibre-cement materials, since as a result the fibres are less likely to suffer alkali attack or lumen petrifaction. However, the hydration rate of BFS-based cement is known to be lower than that of OPC and to be strongly dependent on the degree of fineness and cure procedures employed (Swamy, 1997).

The objective of the present study was to evaluate the weathering in temperate and tropical climates of vegetable fibre-BFS cements fabricated using the slurry vacuum de-watering process. Mechanical, physical and microstructural properties of non-aged and aged composites were compared in an attempt to identify the main mechanisms related to changes in composite performance over the long term.

RAW MATERIALS AND PREPARATION

Basic granulated iron BFS provided by Companhia Siderurgica Tubarao (CST), Brazil, and fully characterised by Oliveira et al. (1999), was ground to an average Blaine fineness of 500 m²/kg and employed as the main component of an alternative binder. Ground agricultural gypsum and construction grade hydrated lime were used as activators in the proportions of 0.88:0.10:0.02 (BFS:gypsum:lime) by mass.

Three different types of Brazilian fibrous residue were selected on the basis of their availability and relatively low levels of contamination: waste *Eucalyptus grandis* kraft pulp, with an estimated availability of 30,000 tonnes/year; sisal (*Agave sisalana*) field by product, with an estimated 100,000 tonnes/year available, equivalent to the amount of commercial sisal production in Brazil; and banana (*Musa cavendishii*, nanicao variety) pseudo-stem fibres, with a potential production of 95,000 tonnes per annum in Ribeira Valley, the main producing area in Sao Paulo state.
Samples were thermally sterilised (85°C, 8 h), sealed in plastic bags and forwarded to the Forest Products Laboratory of CSIRO Forestry and Forest Products, Australia. The *Eucalyptus grandis* waste pulp was used as received after a 2 min disintegration and washing in hot (90°C) water.

Chemi-thermomechanical (CTMP) pulping procedures, based on the suggestions of Higgins (1996), were employed in the preparation of banana and sisal fibres. 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. The chemical pre-treatment step consisted of boiling the strands in a 10% lime (on strand mass) liquor for a period of 1 h on completion of the soak. The strands were then mechanically defibred in an Asplund Type D laboratory defibrator and post-refined using a 20 cm Bauer laboratory disc refiner fitted with straight-patterned “rubbing” plates.

Both pulps were passed through a Packer screen (0.23 mm slots) to separate shives and a Somerville screen (0.180 mm mesh) to reduce the incidence of fines of length less than 0.2 mm. Fibre shortening and the generation of fines are typical, although undesirable, outcomes of refining procedures (Higgins, 1996) and can be controlled by the application of appropriate energies to the stock during mechanical treatment. Finally, the pulps were vacuum de-watered, pressed, crumbed and stored in sealed plastic bags under refrigeration.

The main physical attributes of the pulps produced are summarised in Table 1. The Canadian Standard Freeness (CSF) of each pulp was determined in accordance with AS 1301.206s-88. CSF is an arbitrary measure of the drainage properties of pulp suspensions and is associated with the initial drain rate of the wet pulp pad during the de-watering process (Coutts & Ridikas, 1982). Fibre length and fines content were determined using a Kajaani FS-200 automated optical analyser.

<table>
<thead>
<tr>
<th>Fibre</th>
<th>Freeness (ml)</th>
<th>Fines (%)</th>
<th>Length (mm)</th>
<th>Width (µm)</th>
<th>Aspect ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td><em>E. grandis</em></td>
<td>685</td>
<td>7.01</td>
<td>0.66</td>
<td>10.9</td>
<td>61</td>
</tr>
<tr>
<td>Sisal</td>
<td>500</td>
<td>2.14</td>
<td>1.53</td>
<td>9.40</td>
<td>163</td>
</tr>
<tr>
<td>Banana</td>
<td>465</td>
<td>1.55</td>
<td>2.09</td>
<td>11.8</td>
<td>177</td>
</tr>
</tbody>
</table>

1 Arithmetic basis, 2 Length-weighted basis, 3 Average of 20 determinations by SEM

**COMPOSITE PREPARATION**

Cement composite pads measuring 125 x 125 mm and reinforced with 12% by mass of *E. grandis* waste kraft or 8% of either CTM pulp were prepared in the laboratory using a slurry vacuum de-watering technique. The selection of fibre contents was based on the optimum levels found in a similar study published elsewhere (Savastano Jr. et al., 2000). Pads of each formulation were prepared in groups of three, pressed simultaneously at 3.2 MPa for 5 min, then sealed in a plastic bag to cure in saturated room temperature air for the following 7 days.

On completion of the initial saturated air cure, pads destined for testing at a total age of 28 days were each wet diamond sawn into three 125 x 40 mm flexural test specimens. Specimen depth was the thickness of the pad, which was in the region of 6 mm. The specimens were then allowed to air cure in a laboratory environment of 23 ± 2°C and 50 ± 5% relative humidity prior to the conduct of mechanical and physical tests.

Three pads of each formulation were allocated for exposure to 3 and 12 month periods of weathering in temperate Australian and tropical Brazilian environments. Additional sets of pads were stored continuously in the laboratory over the same periods to provide specimens for the determination of reference properties at the different ages. On removal from their bags, these pads were allowed to air cure in the laboratory environment to 28 days of age before being exposed to weathering or further
ageing in the same environment. At total ages of 4 and 13 months, test specimens were prepared as previously described and stored in laboratory conditions for 7 days to achieve equilibrium moisture content prior to testing.

TEST METHODS

A three point bend configuration was employed in the determination of modulus of rupture (MOR), modulus of elasticity (MOE) and fracture energy. A span of 100 mm and a deflection rate of 0.5 mm/min were used for all tests in an Instron model 1185 universal testing machine. 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. For the purpose of this study, the fracture toughness (FT) was measured as the fracture energy divided by specimen width and depth at the failure location. Nine flexural specimens were tested for each formulation and condition of exposure. The mechanical test procedures employed are described in greater detail by Savastano Jr. et al. (2000).

Water absorption and bulk density values were obtained from tested flexural specimens following the procedures specified in ASTM C 948-81. Six specimens were used in the determination of these physical properties.

Property data was subjected to one-way analysis of variance using Tukey’s multiple comparison method to determine the significance of observed differences between sample means at the 95% confidence level (α = 0.05).

Fracture surfaces of composites subjected to the different environments and periods of exposure were analysed using a Philips XL30 field emission gun (FEG) scanning electron microscope (SEM). To facilitate fibre observation some images were taken after tilting the samples 75° in relation to the horizontal plane. Observation procedures were similar to those employed by Coutts & Kightly (1984).

WEATHERING CONDITIONS

28 days after manufacture the allocated series of composites of each formulation were placed in a rack facing North at an angle of inclination of 45° to age naturally in the temperate environment of Melbourne, Victoria, Australia (37° 49’ S of latitude). Exposure commenced in April 1999 for those composites reinforced with *E. grandis* pulp, and July 1999 for the remaining composites. Corresponding series of composites were exposed in a like manner to the tropical environment of rural Pirassununga, Sao Paulo state, Brazil (21° 59’ S of latitude). Exposure of these series commenced in July 1999.

Table 2 lists the main long term climate averages for the Australian and Brazilian exposure sites.

<table>
<thead>
<tr>
<th>Place</th>
<th>Temperature (°C)</th>
<th>Relative humidity (%)</th>
<th>Average rainfall (mm/year)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Ave max / month</td>
<td>Ave min / month</td>
<td>Ave max / month</td>
</tr>
<tr>
<td>Melbourne, Vic, AU a</td>
<td>25.8 / Jan</td>
<td>5.9 / July</td>
<td>82 / June</td>
</tr>
<tr>
<td>Pirassununga, SP, BR b</td>
<td>30.1 / Jan - Feb</td>
<td>9.5 / July</td>
<td>77 / Jan - Feb</td>
</tr>
</tbody>
</table>

Source: a Bureau of Meteorology, Australia; b Air Force Academy, Defence Ministry, Brazil.
RESULTS AND DISCUSSION

Mechanical properties
Non-aged composites presented statistically undifferentiated flexural strengths in excess of 18 MPa, representing a 120% improvement over a plain BFS matrix of similar formulation. As shown in Figure 1, one year of external exposure to tropical or temperate weather resulted in a considerable reduction in strength, which had fallen to 6.6 MPa in the case of the 12% *E. grandis* formulation exposed in Brazil. The loss in mechanical strength of composites subjected to either natural weathering or ageing in the controlled environment is attributable to matrix carbonation. Such a mechanism (Wang et al., 1995) consumes calcium ions from ettringite and from the calcium hydroxide occasionally present in BFS cement, and simultaneously lowers the Ca/Si ratio of the calcium silicate hydrate (C-S-H) as discussed by Taylor (1997). Qualitative evaluation using an indicator solution of 2% phenolphthalein in anhydrous ethanol revealed that the aged composites were completely carbonated. The greater severity of the natural environment on composite properties can be attributed to interfacial damage resulting from volume changes of the porous and hygroscopic vegetable fibres inside the cement matrix (Savastano Jr. & Agopyan, 1999).

Non-aged composites possessed 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 low modulus of the cellulose fibres employed and the additional porosity resulting from their inclusion. MOE dropped to the interval of 1.8 - 4.8 GPa after 13 months of ageing. 12% *E. grandis* in BFS provided the worst performance (Figure 2), as was observed in the case of MOR.

Fracture toughness (FT) is the matrix property most often enhanced by the presence of fibres, which in these materials produced a 15-fold or greater increase. As shown in Figure 3, after a period of weathering or laboratory ageing, composites demonstrated ductilities similar to or even higher than those at 28 days. 8% banana fibre in BFS, exposed in any of those environments, possessed a toughness of ~1.0 kJ/m² which corresponds to a near doubling of the short term value. The improvements in ductility can be linked to the losses in MOR and MOE, confirming the expected compromise between strength and ductility in such composites.

![Figure 1. Variation in composite MOR with age and conditions of exposure. Error bars indicate single standard deviations of the means.](image-url)
FT values in the range of 1.0 - 1.2 kJ/m² after a year of weathering indicate that the integrity of the fibres within the BFS matrix has not been significantly reduced by decomposition or petrifaction. In a previous study of sisal, malva and coconut strands in OPC, Savastano & Agopyan (1999) reported reductions of at least 50% in ductility after only 6 months in a laboratory ambient. Toledo Filho et al. (2000) and Bentur & Akers (1989) noted similar embrittlement in aged vegetable fibre-OPC composites and found that it could be directly attributed to the petrifaction of the reinforcement through the migration of hydration products to the fibre lumens and pores.

Figure 2. Variation in composite MOE with age and conditions of exposure.

Figure 3. Variation in composite FT with age and conditions of exposure.
Physical characterisation
Short term water absorption (WA) and bulk density values of the composites were in the ranges of 32 - 33% by mass and 1.3 - 1.4 g/cm³ respectively, regardless of the fibre type or content (Figures 4 and 5). Plain BFS matrix, produced using an analogous process to that reported in the present study, was found to have a WA of 18% and density of 1.8 g/cm³, confirming the influence of the cellulose fibres on the volume of capillary voids in fibre-cements.

WA values had fallen after 4 months of external weathering but rose to exceed those at 28 days in the longer term. This behaviour may partially be due to matrix carbonation and subsequent leaching under the action of rainwater. Propagation of microcracks, generated by the cyclic action of temperature, radiation and moisture, may also have contributed to the increases in WA which, in the case of 12% *E. grandis* in BFS, reached 37% with a corresponding density of 1.25 g/cm³ after Brazilian exposure.
Microstructure properties

A fracture surface of non-aged banana fibre reinforced BFS, in which numerous broken filaments can be observed, is shown in Figure 6. The inclusion of these long and supposedly strong fibres (tensile strength ~800 MPa, comparable to that of *Pinus radiata* as reported by Coutts, 1990) could be expected to give rise to considerable fibre pullout. The predominance of fibre fracture suggests that either the fibres were damaged during pulping or that the fibrillation imparted resulted in sufficiently improved anchorage in the matrix to significantly reduce the critical fibre length (Beaudoin, 1990). The high incidence of fibre fracture led to the composite absorbing relatively little energy in the post-cracking stage despite possessing a flexural strength similar to that of the sisal composite.

![Figure 6. SEM image (75° tilt, carbon coat) of the fracture surface of banana CTMP in BFS. Hydration age: 32 days.](image)

Brittle fracture can be expected in air-cured OPC-based composites after ageing (Bentur & Akers, 1989). However, the lumens of fibres in the fracture surfaces of BFS-based composites, such as those...
in Figure 7, appeared to be free of deposited hydration product (or fibre petrifaction) when examined at a relatively young age. Figure 7 also shows partially pulled-out fibres encrusted with matrix material, attesting to significant energy dissipation in the interfacial zone during fracture progression.

Examination of the fracture surfaces of composites exposed for 12 months to the Melbourne climate revealed still no evidence of fibre petrifaction and confirmed that the fibres remained in good condition. Fibre pullout rather than fracture was predominant in all of the composites, as would be expected in light of their toughness values. A fracture surface of 8% banana CTMP in BFS is shown in Figure 8. In contrast to the surface of the non-aged specimen (Figure 6), the fibres remain largely intact. This suggests that the fibre-matrix interface or the matrix itself has weakened, in turn weakening the composite but improving its toughness by allowing greater dissipation of energy through the mechanism of fibre pullout.

Figure 8. SEM image (75° tilt, gold coat) of the fracture surface of banana CTMP in BFS after 12 months exposure to a temperate climate.

Figure 9. SEM image (75° tilt, gold coat) of the fracture surface of sisal CTMP in BFS after 12 months exposure to a temperate climate.
Figure 9 shows a fracture surface of the corresponding 8% sisal CTMP-BFS composite. Again the pulled-out fibres are largely intact. A fragment of sisal strand not removed by the screening step can be seen at the top of the image and contrasts the nature of sisal reinforcement when incorporated as individual fibres or in the strand form.

CONCLUSIONS

BFS cement reinforced with pulp fibres obtained from agricultural or industrial by-products showed increased flexural strength and toughness (up to 19 MPa and 1.2 kJ/m² respectively) relative to the plain matrix at 28 days of age. Production involved a slurry vacuum de-watering process, press compaction and air curing. However, as a consequence of the large amount of vegetable fibres employed, the modulus of elasticity values for the composite materials were relatively low, between 5 and 6 GPa, and water absorption exceeded 30% by mass.

Exposure to temperate or tropical climates caused a severe reduction in the flexural strength and modulus of composites and can be attributed to matrix carbonation followed by leaching and progressive microcracking. At 13 months of age under external weathering the MOR fell to between 36 and 55% of that of the non-aged composites and MOE to 37 - 65% of the initial values. The high porosity associated with water absorption in the region of 30% by mass is expected to be a significant factor in this undesirable behaviour and the refinement of pore structure may be an effective approach to material optimisation.

Contrarily, ductility remained stable in the long term for 12% E. grandis in BFS, with an outstanding fracture toughness (FT) in the vicinity of 1.2 kJ/m². In the case of 8% sisal or banana CTMP, FT values of 1.0 kJ/m² or greater exceeded the short term results, indicating that the fibres continue to be capable of providing reinforcement. The low alkalinity of the BFS cement, use of pulped fibres, good fibre-matrix adhesion and an absence of fibre petrifaction are assumed to be the major factors in achieving this improved result relative to previous studies on similar materials.

Specimens from the three analysed formulations remain exposed to weathering to allow the examination of properties at greater ages to be carried out in the future. After 15 months exposure, all of the composites appear to be retaining their structural integrity, with no evidence of cracks in exposed surfaces and only isolated signs of superficial erosion. The performances presented indicate a potential for the use of these types of materials on an industrial scale for the production of thin construction panels that are accessible to developing areas with a critical lack of housing and infrastructure.

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