TOUGHNESS OF CELLULOSE CEMENT COMPOSITES

R.A. Tait* and W. Keenliside**

*Department of Mechanical Engineering, University of Houston,
Houston, Texas 77004 U.S.A.

**Department of Physics, University of West Indies
Mona, Kingston 7, JAMAICA

ABSTRACT

The results of a preliminary study to examine the applicability of low cost cellulosic fillers within an inorganic cement matrix are presented. The paper discusses the properties of three basic systems:

(i) A conventional cement-stone aggregate;
(ii) a coir reinforced cement (containing 20% by volume of coir fibres);
(iii) a pressure molded wood particle-cement composite (containing 20% by volume of wood particles).

In outline, the results show that while these three systems exhibit a conventionally linear stiffness-density relationship, the fracture toughness, K_{Ic}, of the pressure molded product is significantly higher than that of both the conventional cement-stone aggregate and the coir reinforced cement. This indicates that the loss of stiffness which results from the addition of a low density filler may be attractively offset by a marked increase in toughness, provided that the particle-matrix bonding conditions are controlled to ensure a high energy fracture path during failure. The paper addresses the mechanisms of failure in inorganically bonded composites with respect to filler geometry, properties and compatability, and further outlines some of the problems associated with water absorption.

INTRODUCTION

There is little doubt that the dramatic increases in the cost of energy have had serious consequences for the developing economies of Third World Nations. Not least of these is the effect upon low cost housing programs that have been curtailed as a result of the high price of cement, whose manufacture involves a large energy content. General concern over the need to develop and establish building materials industries, utilizing the local raw materials of these regions to the maximum extent, was first expressed in a resolution passed by the United Nations General Assembly in 1965 [1]. Straw and mud construction is of course of ancient origin. Asbestos cement has been in common usage since the beginning of this century, despite its associated health hazards, and the viability of using cellulosic fillers in cement has been realized at least since the time of the Second World War [2] . In 1978, a conference [3] devoted to the theme of efficient materials utilization in developing countries allowed a forum presentation for a wide variety of practical and cost efficient possibilities for the impregnation of an inorganic cement matrix with natural fibres from for example the coconut husk (coir) [4] the sugar cane stem (bagasse) [5] and sisal [6]. It is now clear that the "energy crisis" will cause a reappraisal of the efficiency of materials usage on a world wide basis. To this end, the development of modern plants capable of producing the bagasse - cement and wood particle - cement composites is already underway [7]. It would appear timely, therefore, for greater effort towards the understanding of these materials by the developed and technically advanced part of the world, which although somewhat insulated, at present, from all but the minor inconveniences of higher priced energy, has yet to face the inevitable alteration in the economic availability of vital resources.

The work described here is meant partly to indicate some of the advantages which may accrue to potential users of lightweight concrete, as well as the areas for future research, aimed at improving the properties and long term stability of cellulose - cement composites. The proposed use of natural cellulose reinforcement in the form of plant fibres or chopped wood particles, over the already widely researched glassy [8] metallic [9] or polymeric [10] fibres, in cement matrices is not based entirely upon economic availability. It has been established that the work of fracture of both plant fibre [11] and timber [12] may achieve exceptionally high values of around 10^5 J/m^2, which result from the uncoiling of the crystalline cellulose microfibrils that make up the major reinforcement of plant fibre and timber cell walls. The utilization of such materials as toughening agents for the inherently brittle cement is therefore sought.

THE MATERIALS

The three materials tested were a pressure molded wood-particle cement, a coir-fiber cement, and a conventional cement-stone aggregate. The wood-particle cement is a commercial product of the Bison Company of West Germany [7]. Coir is the fibrous constituent of the coconut husk, the samples reinforced with this product were manufactured in the laboratories of the University of the West Indies, Kingston, Jamaica. The aggregate samples were extracted from commercial building blocks that were manufactured locally in Jamaica. All samples had cured for at least two months before testing. The approximate composition of each product is presented in Table I.

TABLE 1. Composition of Cements
During Manufacture (wt. %)

Material	Cement	Fiber	Grit	Water	Mold Pressure
Cement Aggregate	30%	-	50%	20%	NONE
Coir Fiber Cement	60%	20%	-	20%	NONE
Wood Particle Cement	60%	20%	-	20%	0.7MPa

The coir fibre samples are actually derived from two sources. The first is as described above, the second consisted of parts extracted from a light structure which had been removed after a period of nine years. The detailed description of the manufacture of this product is described elsewhere [14].

EXPERIMENTAL TECHNIQUES

Measurement of fibre length, fibre diameter, grit size and porosity

Fibre length and diameter, as well as grit size, were measured to an accuracy of ± 0.01 cm using an optical binocular microscope. Coir fibres were separated from the cement matrix by

(i) the use of tweezers
(ii) crumbling the matrix around the fibre (where possible)

Only undamaged fibres were used. Porosity was determined by using a transparent graticlue under a magnification of x50. It was computed according to the formula

$$\text{porosity} = \frac{\text{volume of voids (pores)}}{\text{overall volume of specimen}}$$

which is assumed to be directly proportional to

$$\frac{\text{surface area occupied by voids}}{\text{total surface area}}$$

Measurement of Density

All four edges of the specimens used were smoothly and squarely trimmed by use of the sanding machine. Measurements on all faces were made to an accuracy of ± 0.01 cm by use of a micrometer screw gauge. Weights were measured to an accuracy of ± 0.01 g by use of an electric balance.

Measurement of Water Absorption

All specimens were prepared as described above.

Each specimen was weighted to an accuracy of ± 0.01 g and then submerged in water. It was then re-weighted after 48 hours, re-submerged, and the above weighing procedure repeated to constant weight.

The present water absorption is expressed by weight according to the formula:

$$\frac{(W_t - W_o) \times 100}{W_o} \tag{1}$$

where W_t = weight after time t (or constant weight)
W_o = original (dry) weight.

After each submergence excess water on the surface of the specimen was removed with damp tissue before re-weighing.

Specimens of a standard size were not used. W_o is not based on the oven-dry weight. Because of variable specimen sizes, specimens were submerged under variable water depths. Tap, and not distilled water was used in all experiments.

No distinction was made between absorbed and adsorbed water although the term "water absorbtion" is used.

Determination of Young's Modulus, E

To determine the value of Young's modulus for the various materials described, beam test specimens with a rectangular cross section were cut from each sample using a geological diamond cutting wheel. These smooth test specimens were loaded in a three point bending rig having a major span of 120 mm.

Overall dimensions of the specimens were 30 mm x 30 mm x 180 mm for the coir and cement aggregate specimens and 10 mm x 20 mm x 130 mm for the wood particle cement specimen. The load was applied at the center span position, and the deflection was measured at this point. A dial gauge accurate to ±.001 mm was used for these deflection measurements. Young's modulus was then calculated according to the expression:

$$E = \frac{L^3 A}{48I} \tag{2}$$

where E = Young's modulus, L = major span, I = the area moment of inertia, and A = the slope of the force deflection curve. Ten separate determinations of Young's modulus for each material type were made in this way.

Determination of Fracture Toughness

To determine the opening mode fracture toughness, K_{Ic}, beam testpieces containing notches of various depths were loaded to failure in three point bending. The span depth ratio was 4:1 for all testpieces. The value of K_{Ic} was determined at the onset of rapid crack propagation using the compliance calibration equation [13]:

$$K_{Ic} = \frac{6P}{bw^{\frac{1}{2}}}\left(1.93\left(\frac{a}{w}\right)^{\frac{1}{2}} - 3.07\left(\frac{a}{w}\right)^{3/2} + 14.53\left(\frac{a}{w}\right)^{5/2} - 25.11\left(\frac{a}{w}\right)^{7/2}\right. \tag{3}$$

where P = the failure load, b = the testpiece width, w = testpiece depth, a = the notch depth.

SEM[1] PREPARATION

Each surface examined under the SEM was coated with a thin gold layer using the sputtering technique. To use this technique effectively, all sources of moisture should be removed from the specimen. Water vapor will decompose to O_2 within the system and effectively reduce the deposition rate. This problem is significant because the sputtering process heats the specimen and the moisture retained in the cellulose fibers will be released as vapor. To solve this problem the specimens were heated in a laboratory oven for times up to 4 hours. Since some damage to cellulose fibers is risked in the 100 to 125°C range, care was taken not to overheat the specimens. The temperature was maintained at 90°C which is the normal temperature of commercial kiln drying processes. By following this procedure, a suitable coating of gold was achieved. Artefacts were minimal despite the high porosity of the surfaces.

RESULTS

Material Characterization

The nine year old sample of coir reinforced cement consisted of approximately 70% coir and 30% cement by volume. Single coir fibres of length, ℓ, in the range 0.8 to 1.3 cm and with an average diameter of around 200 μm made up some 15 - 20% by volume of the specimen. Coir fibre bundles with the same range in length as above, but with diameters in the range 3 - 4 mm constituted between 30 - 40% by volume of the sample. The majority of fibre bundles exhibited fibre decay. Single fibres were generally unaffected by decay. Fibre particles, about 200 μm in size make up a minimum of 20% of the sample by volume. These fibres were usually well coated with cement.

---

[1] Scanning Electron Microscope

The percentage by volume of coir and cement in the two month old samples of coir reinforced cement was estimated to be the same as in the nine year old sample. Fibre bundles constitute only 10 - 20% of the sample by volume. Fibre lengths for both single fibres and fibre bundles were observed in the range 3 - 10 mm. Diameters remain the same as for above. Nodules of hardened, unreacted cement having diameters in the range 5 - 10 mm made up about 5% by volume of the two month old sample.

The conventional cement aggregate sample consisted of approximately 70 - 80% grit and 20 - 30% cement. Grits of size greater 10 mm in diameter constituted 10 -15% by volume; those having diameters in the range 2 - 10 mm constituted 50 - 60% by volume and those having diameters less than 2 mm about 5% by volume.

The commercial wood particle cement, well compressed and compacted consisted of 70% by volume of wood particles whose size ranged from about 0.5 mm to 3 mm maximum dimension. These particles were more plate-like than fibre-like.

Measurements of porosity and density of the samples are presented in Table 2.

TABLE 2. Comparison of approximate porosity and density of materials examined

Specimen type	Approximate Porosity (% Volume)	Density (kgm⁻³) Range	
		Range	Overall Mean
Coir reinforced Cement Aged for 9 years	20 - 30	740.0- 862.3	795.0
Coir reinforced cement Aged for 2 months	5 - 15	775.6- 857.0	823.0
Wood particle cement	negligible	1149.4- 1233.3	1182.7
Conventional agglomeratic concrete	30 - 50	1795.2- 1933.5	1876.4

Water Absorbtion

The results of the water absorbtion tests are presented in Table 3.

As these results are expressed in terms of weight percentages they do not reflect exactly the volume percentages used to describe porosity. In this regard attention is directed to the wood particle samples which were observed to have very low porosity but nevertheless absorbed a great deal of water.

Mechanical Properties

The results of the mechanical tests are summarized in Figures 1 & 3. Figure 1 indicates that the Young's Modulus is reduced at lower density. The coir system has

TABLE 3. Comparison of Water absorbtion (% Weight

Specimen Type	Water Absorbtion (% Weight)			
	After 48 hours		To Constant weight	
	Range	Overall Mean	Range	Overall Mean
Coir reinforced cement Aged for 9 years	38.3- 52.9	46.6	60.6- 77.9	69.2
Coir reinforced cement Aged for 2 months	42.5- 50.7	46.6	50.2- 58.7	54.1
Wood Particle Cement	18.2- 25.4	22.3	39.5 40.7	40.1
Conventional agglomeratic cement	7.1- 9.6	8.6	7.9- 10.5	9.1

a density of about one-half that of the aggregate, but Young's modulus shows a decrease of nine times. The wood particle system has a higher density and Young's modulus than the coir system. The increase in Young's modulus is due to the decrease in the number of voids that are present due to pressure molding as well as the increased stiffness of the filler materials. Voids will lower the Young's modulus by reducing the efficient of stress transfer between the matrix and fiber. Figure 2 shows that if a fiber of length L extends into a void its effective length is reduced to Lm. The presence of the voids decreases the matrix volume fraction without a corresponding increase in the fiber volume, thus reducing the stress transfer efficiency. Racines and Pama [5] have indicated that voids will affect bond strength and that the number of voids will increase with increases in fiber volume.

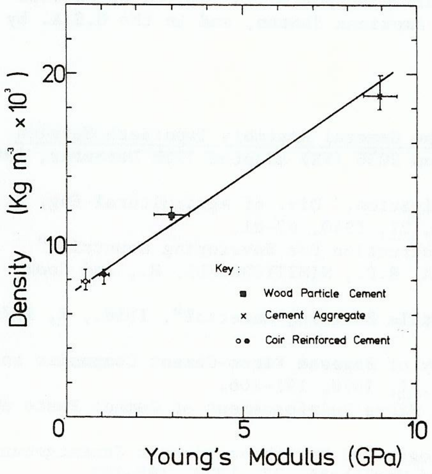
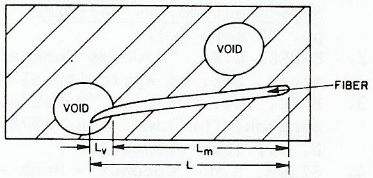


Fig 1. Young's Modulus vs. Density



ROLE OF VOIDS IN DETERMINING THE EFFECTIVE FIBER LENGTH

Fig. 2

Figure 3 summarizes the values of the opening mode fracture toughness, K_{Ic}

The wood particle system has a significantly higher value than the coir system. The aggregate obtained a value about half that of wood particles system. Figure 3 also shows that the values obtained for K_{Ic} are consistently independent of the $\frac{a}{w}$ ratio for the experiments performed here.

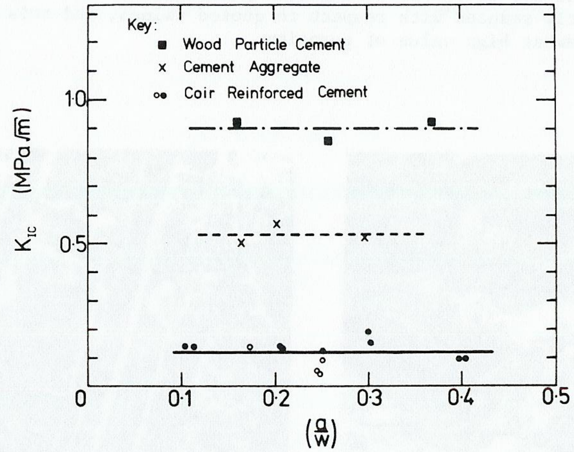


Fig 3. Fracture Toughness

DISCUSSION

Accounts describing the reinforcement of cement with polymeric, metallic or glassy fibres have usually attributed the observed toughening to an increase in the frictional work of fibre pull-out [15]. Figure 4 shows the coir reinforced cement fracture surface in which there are several pulled out fibres and further indications of fibre/matrix interface cracking. The generally poor bonding between fibres and matrix is probably due in part to fibre degradation but also to the high void content. The material in this condition is therefore both brittle and compliant. Figure 5 illustrates a broken fibre which because of its decayed condition was unable to provide any toughening. Indeed, because of the poor condition of both the fibres and their bonding, their presence is most probably detrimental to the properties of the cement.

Fig 4. Coir-cement illustrating fibre pull out

Fig 5. Broken decayed coir fibre

Figure 6 illustrates a typical view of the cement/aggregate fracture surface in which a high proportion of the crack path follows an inter-particle course. The toughness of this sample (0.5 MPa $\sqrt{m}$) is in the range frequency reported by other workers [16] for unreinforced cement. This observation is regarded as further support for the use of K_{Ic} as a toughness parameter for these materials. The stiffness is slightly reduced with respect to quoted values, and this probably results from the somewhat high value of porosity.

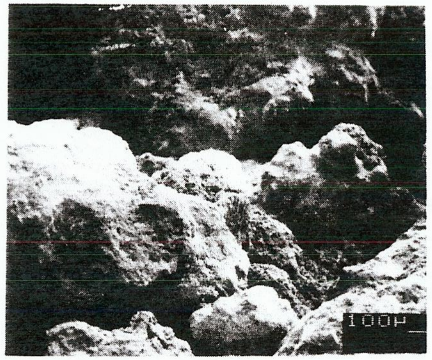


Fig 6. Cement-aggregate fracture surface

Fig 7. Wood-particle cement fracture surface

The wood particle cement fracture surface is shown in Figure 7. It is clear that the increased toughness in this system has arisen as a result of the high energy crack path *through* the wood particles. Individual cells have been ruptured and there is evidence of cell wall buckling and micro-fibril pull out. In the past, consideration of cellulose/cements has always been forstalled because the hygroscopic nature of the fibres or particles have caused difficulties in manufacture due to incomplete hydration of the cement, and rapid deterioration during service because of fibre rotting. In the short-time water tests performed here no evidence of fibre rotting was observed but an indication of incomplete cement hydration [17]was seen in the form of crystallite formation, shown in Figure 8 after only seven days of immersion. This observation is consistent with the high degree of water absorbtion observed for this system, when compared with the apparently negligible porosity, and indicates the present requirement for protective coatings to obviate bonding degradation in wet or humid environments.

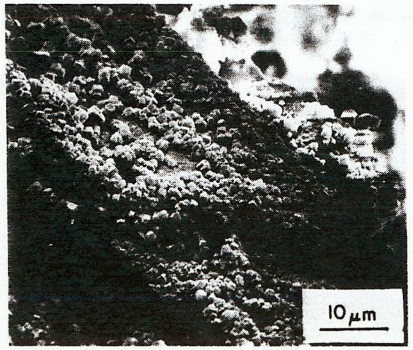


Fig 8. Surface of wood-particle cement after seven days immersion in water.

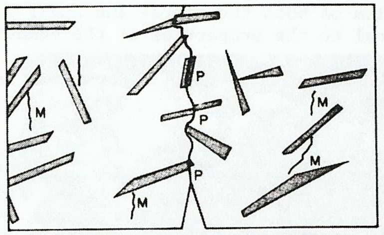


Fig 9. Schematic crack path for wood-particle cement.

CONCLUSIONS

(i) The toughening of wood particle cement is achieved by causing the crack path to be directed through the tough and relatively compliant wood-particles, as shown schematically in Figure 9.
(ii) The passage of the crack through the wood particles was accompanied by cell wall buckling and micro-fibril pull out.
(iii) Some evidence of incomplete cement hydration was observed in the commercial wood particle system.
(iv) The poor performance of the coir-fibre reinforced cement resulted primarily from very bad fibre/matrix bonding and also from the unfavorable stress transfer conditions due to a high void content.
(v) The use of K_{Ic} as a measure of toughness in these systems may be regarded as a sensitive but qualitative guide. The value of such a measure as a quantitative parameter in the detailed assessment of fracture mechanism remains to be established.

ACKNOWLEDGEMENTS

The contribution of Dr. T. P. Hughes in introducing the study of natural fibres at U.W.I. is gratefully acknowledged. Brian Baptiste and Jeff Bunch performed some of the experiments. This work was partly supported in Jamaica by various equipment grants from the Organization of American States, and in the U.S.A. by the Energy Foundation of Texas.

REFERENCES

1. UNITED NATIONS, Official Records of the General Assembly Twentieth Session Supplement No. 14 (A/6018). Resolution 2036 (XX) adopted 17th December, 1965, pp. 39 para (c).
2. BAUER, L.W., "Sawdust Concrete Investigation," Div. of Agricultural Eng. J. of Ameri. Soc. of Agricultural Engineers, 21, 1940, 63-81.
3. PROC. INT. CONF. on "Materials of Construction for Developing Countries" Bangkok, Thailand, Aug. 1978 (Eds PAMA, R.P., NIMITYONGSKUL, P., and Cook, D. J.) (two volumes).
4. SINGH, S.M. "Coconut - Husk - A Versatile Building Material", Ibid., I, 1978, 207-220.
5. RACINES, P.G. and PAMA, R.P., "A Study of Bagasse Fibre-Cement Composite as Low Cost Construction Material, Ibid., I, 1978, 191-206.
6. SWIFT, D.G. and SMITH, R.B.L., "Sisal Fibre Reinforcement of Cement Paste and Concrete", Ibid., I, 1978, 221-234.
7. PAMPEL, H. and SCHWARZ, H.G., Technologie und Verfahrenstechnik Zementgebundener Spanplatten", Holz als Roh - und Werkstoff, 37, 1979, 195-202
8. JOHNSTON, C.D., "Steel - fibre - reinforced mortar and concrete: A review of Mechanical Properties", Fibre reinforced Concrete, ACI Publ. SP 44, pp.127-142
9. HANNANT, D.J., ZONSVELD, J.J., and HUGHES, D.C., "Polypropylene fibres in cement based materials", Composites, 9, No. 2, April 1978, I.P.C.

10. BIRYUKUKOVICH, K.L., BIRYUKUKOVICH, YU L., and BIRYUKUKOVICH, D.L., "Glass - fibre - reinforced Cement," Publ. Buidvel'nik, 1964, CERA Translation, NO. 12, Nov. 1965.
11. McLAUGHLIN, E.C. and TAIT, R.A., "Fracture Energy of Plant Fibres," J. Matls. Sci., 14, 1979, 998-999
12. GORDON, J.E. and JERONIMIDIS, G., "Work of Fracture of Natural Cellulose," Nature, London, 252, 1974, 116.
13. BROWN, W.F. and SRAWLEY, J.E., ASTM STP 381, 1965, 133.
14. TAIT, R.A., unpublished report.
15. SWAMY, R.N., MANGAT, P.S., and RAO, C.V.S.K., "The Mechanics of Fibre Reinforcement of Cement Matrices," ACI Publ. SO 44, pp. 1-28.
16. MAZARS, J., "Existence of a Critical Strain Energy Release Rate for Concrete Adv. in Strength and Fracture of Materials, (Ed. D.M.R. Taplin') III, 1977, 1205-1209, (Pergamon Press).
17. DOUBLE, D.D. and HELLAWELL, A., "The Solidification of Cement," Scientific American, July 1977, 82-89.