Wood fibre-reinforced cement composites

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The use of wood fibres as reinforcement for a structural composite material has been investigated. Although wood fibres have relatively poor mechanical properties compared with synthetic fibres, they have the advantages of low density, low cost and low energy demand during manufacture. A number of possible matrix materials were considered and Portland cement was chosen for further investigation. An examination was made of the effect of the pulping technique used to prepare the fibre on the strength of the composite and on the stability of the wood fibres in the cement matrix. A chemically pretreated high-temperature thermomechanical pulp and a pulp produced by the kraft process were selected for further study. The effect of the water—cement ratio of the matrix and the weight of fibre in the composite on the strength of the composite and the rate of increase in strength and fracture energy of composite are reported for composites containing these pulps. The results indicate that the kraft pulp is suitable for applications where slurry dewatering can be employed during the forming operation and that the thermomechanical pulp is more suited to applications where low water—cement ratio slurry is used.

1. Introduction

In recent years there has been considerable research into developing high-performance reinforcing fibres. As a result of this work, continuous graphite, boron and aramid fibres are now used as reinforcement in organic polymer matrix composites. However, the cost of these composites will restrict their use to specialized applications for some years to come. The introduction of glass fibre-reinforced cement (GRC) has gone some way towards the goal of a cheap fire-resistant composite building material, but at present this section of the market is still dominated by asbestos fibrecement composites [1].

There is a growing awareness of the health hazards associated with the inhalation of small diameter ($< 2 \mu m$) durable fibres [2]. The use of asbestos fibres is of major concern and exposure levels in asbestos cement manufacturing plants are now being reduced with a resultant increase in the price of these products. In addition, asbestos fibres are a non-renewable resource and with the most accessible deposits being depleted the cost of these fibres is beginning to rise [2]. In Australia there has been a partial replacement of asbestos fibre in some cement sheet products by naturally occurring cellulosic fibres since the early 1960s. CSIRO is committed to a programme of replacing high-energy consuming, non-renewable resources with lower energy renewable resources. It is in this context that it is worth reviewing the role of cellulosic fibre as a reinforcing agent.

2. Cellulosic reinforcing fibres

Since Australia has limited commercial vegetable fibre resources this discussion will be restricted to naturally occurring cellulosic fibres derived from wood sources. These fibres are cylindrical with tapered ends and have a length-diameter ratio of between 50 and 60. Since the wood fibre is hollow, the reinforcing properties of an individual fibre depend on both the degree of collapse of the fibre and the thickness of the fibre walls. The tensile strength of delignified cell wall substance is between 500 and 900 MPa with 2000 MPa being recorded for selected fibres. An elastic modulus of 25 to 40 GPa is common with up to 100 GPa being recorded [3]. The fibres can be subdivided into two groups depending on whether they originate from hardwoods or softwoods. Hardwood

T	A	В	L	Е	I	Properties	of	fibres
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Material	Cost/tonne A\$ ^{††}	Specific gravity	Tensile strength (MPa)	Specific strength (MPa)	Cost/MN A\$**
kraft (P. radiata)*‡	440	1.5	500	333	1.3
TMP (<i>P. radiata</i>) ^{†‡} High Temperature	250	0.5	125	250	1.0
Asbestos (JM 5R)	460	2.6	700 [¶]	269	1.7
Glass (E)	1 800	2.5	1400 [¶]	560	3.2
Nylon [§] (Tyre Cord)	3 600	1.1	868	789	4.6
Kevlar 49	19 000	1.5	2800	1867	10

*Collapsed fibre.

[†]Uncollapsed fibre – cell wall substance strength assumed to be the same as for the delignified fibre.

⁴Based on "average fibre" with diameter = 38 μ m and lumen diameter = 33 μ m.

[§]Nylon cost includes \$A1.00 per kg + 15% import duty.

[¶]Strength is for commercial glass and asbestos fibres [2]. ^{*}Ratio of cost to load based on 1 m³ material.

^{††}Cost from Australian manufacturers or importers (May 1979).

fibres generally have a diameter of 20 to 60 μ m and a length of 0.5 to 3.0 mm, while the softwood fibres are 30 to 120 μ m in diameter and are 2.0 to 4.5 mm in length.

The method of removal of the fibres from the parent wood (pulping) has a profound effect on the reinforcing properties of the fibres. Pulping processes form a complete spectrum varying from chemical to mechanical. Heat may also be added to the process. So far as reinforcing fibres are concerned the extremities are defined by the fully chemical processes (e.g. kraft) and the hightemperature thermomechanical processes (TMP). The kraft process produces a low yield (approximately 45%) collapsed ribbon-like delignified fibre suitable for papermaking, while the thermomechanical process conducted at temperatures greater than the glass transition temperature of the lignin binder, gives a high yield (approximately 90%) lignin-coated, uncollapsed, rod-like fibre. Disc refining can be used to produce fibres with intermediate properties. Refining operations without external heat application (e.g. Refiner groundwood process) can be used to produce high-yield partially collapsed fibres. High-temperature TMP fibres may be further refined (e.g. by a Bauer disc refiner) to produce pulps with partially collapsed lignin-coated fibres.

The properties of wood fibres prepared from Pinus radiata are compared with those of other reinforcing fibres in Table I. It is evident from the ratio of cost to load carried by the fibre that wood fibres are a highly cost-effective reinforcement. However, the relatively poor absolute properties of the fibres means that considerable care should be exercised in the selection of a matrix. Table II lists some possible matrix materials while Table III compares performance data for composites containing glass, asbestos and wood fibres. This approximate analysis is based on the law of mixtures for a two-dimensional random composite assuming that the strength is 1/3 of the unidirectional case and that the kraft fibres do not swell in the alkaline conditions of the cement mix. The most costeffective matrix materials for use with wood fibres appear, therefore, to be plaster or ordinary Portland cement (OPC). In practice, the economic advantage of either plaster or OPC matrices is further enhanced by the failure phenomena encountered when composites with these matrices are placed in bending. These brittle matrix composites do not behave in tension as they do in compression and

TABLE II Properties of matrix materials

Material	Cost/tonne A\$*	Density (tonne m ⁻³)		
Polyethylene	1000	1.0		
Polyester	1500	1.3		
Portland cement	50	2.5		
Plaster	70	2.4		

*Cost from Australian manufacturers or importers (May 1979).

Material	Cost/tonne A\$	Density (tonne m ⁻³)	Tensile strength (MPa)	Specific strength (MPa m ³ tonne ⁻¹)	Cost/MN A\$ [†]
OPC + 10% kraft bw	89	2.3	26	11	7.9
OPC + 10% TMP bw (high temp.)	70	1.8	15	8.3	8.4
Plaster + 10% kraft bw	107	2.3	25	11	9.8
Plaster + 10% TMP bw (high temp.)	88	1.7	16	9.4	9.4
OPC/Glass fibre 5% bw	138	2.1*	16^*	7.6	18
Asbestos cement sheet 15% fibre bw	112	2.1*	14*	6.7	17
Glass fibre/polyester sheet (random rein- forcement) 50% bw	1650	1.7	160	94	17
Polyethylene/kraft 50/50 bw	720	1.3	67	52	14
Polyester/kraft 50/50 bw	970	1.4	77	55	18

TABLE III Properties of composites

*Data from [1].

[†]Ratio of cost to load based on 1 m³ material.

this results in a flexural strength of up to three times the tensile strength [6].

The engery associated with the production of selected fibres and matrix materials is presented in Table IV. The data for glass fibre was unavailable but it is assumed that it will not differ significantly from that for container glass. It is evident that, from an energy conservation point of view, a combination of OPC and wood fibres would be desirable.

A review of the literature has not revealed any documented health hazards associated with the use of wood fibres except for dermatitis and allergic reactions to extractive-rich species by sawmill workers [7, 8]. Acheson *et al.* [9] have reported nasal cancer in wood workers exposed to dust from selected species. The physical dimensions of the fibres are such that they are unlikely to lodge in the lungs [2].

T A B L E $\,$ IV Energy consumption associated with composite materials

Material	Energy (GJ tonne ⁻¹)	Energy (GJ m ⁻³)		
Glass (container)[5]	13	32		
Wood fibre (TMP) [4]	8.6	4.3		
Plastics [5]	90	90		
Cement [5]	4.7	12		

There is a history of materials in which wood and cement are combined. Wood residues in the form of sawdust, slivers, shavings and "excelsior" (wood-wool) strands have been used in conjunction with cement to produce blocks, boards and floors for buildings [10-12]. In all these materials the dominant component by volume is the wood. Krenchel [13] has suggested that papermaking fibres may be used as a replacement for asbestos fibres. Vegetable fibres (e.g. sisal, cotton, bamboo) have been used with some success as reinforcement for materials with a cement or plaster matrix although they have now been replaced in most applications by polymer or glass fibres except in the developing countries [14, 15].

A composite material composed of a cement or plaster matrix reinforced by a small volume of individual wood fibres appears to offer potential as a low cost, low-energy consuming building material, with reasonable mechanical properties. We have selected ordinary Portland cement as the matrix for this investigation.

3. Compatibility of wood fibre with cement

Before a composite of wood fibre and cement can be considered it is necessary to examine both the chemical and physical compatibility of the materials. The chemical compatibility of the fibre and the cement will depend to a large extent on the type of pulping process employed to produce the fibre.

It is well known that when setting cement paste contacts some species of wood the cure of the cement is inhibited. This effect is believed to be due to the leaching of the tannins and sugars contained in the wood into the cement [12]. In the case of low-yield pulps produced by alkaline chemical processes, interaction between the cement and the wood fibre is unlikely. However, if high-yield mechanical pulps are used, the cure of the matrix adjacent to the fibres may be impaired.

The long-term stability of the wood fibrecement composite must also be considered. Unlike glass fibres, wood fibres appear to be inherently stable under the highly alkaline conditions of the cement matrix. Cement-sawdust floors laid in a CSIRO building in 1947 are still in service and Huffaker [11] has reported good results for cement-wood shavings blocks after 12 years of natural weathering. Asbestos-cement sheets containing wood fibre are still in exterior service after 13 years under Australian conditions with almost no loss in mechanical properties [16]. Examination of samples taken from a 30 year old cement-sawdust floor revealed mineralization of the wood sawdust. It is possible that a wood fibre cement composite may, in time, become brittle due to this reaction; however, this was not observed in the asbestos-cement sheets containing wood fibre [16]. Attack on the wood fibres by micro-organisms is unlikely since it has been reported [17] that cotton fibre-cement composites are highly resistant to this form of degradation.

The elastic modulus of wood fibres is of the order expected for Portland cement (7 to 28 GPa) and is low when compared with other reinforcing fibres such as steel and glass. This means that the addition of wood fibres is unlikely to increase the elastic modulus or limit of proportionality of the cement. The amount of shrinkage is also unlikely to be affected. However, an increase in the fracture energy and modulus of rupture, and a decrease in shrinkage cracking could be anticipated.

4. Experimental details

The objectives of the experimental programme were to determine a pulping technique suitable

for the production of wood fibres for cement matrix composites and to examine the effect on the mechanical properties of the composite of time, the water-cement ratio of the matrix and the weight of fibre in the composite.

A selection of high-temperature thermomechanical wood fibre pulps were prepared in the laboratory and these along with commercial pulps were assessed for their mixing and reinforcing properties in cement matrix composites. The laboratory pulps were prepared by defibrating 300 g air-dried commercial chips that had been fully saturated in 1.5 litre water or water containing 25 g NaOH or 25 g Na₂SO₃. The chips were loaded into an Asplund type D laboratory defibrator and presteamed for 4 min at 0.69 MPa gauge pressure then run at 1440 rpm for a further 30 sec. Pulps produced using these conditions consisted of individual uncollapsed fibres that did not readily bond together upon drying. One pulp was further refined under atmospheric conditions by one pass through a 300 mm Bauer disc refiner at 0.13 mm plate clearance. The matrix was fresh commercial type A OPC.

Preliminary trials indicated that all the pulps could be successfully incorporated into a cement water slurry in the ratios 40 OPC:16 water:1 fibre by weight. The kraft and groundwood pulps required soaking overnight in water and dispersion in a high-speed mixer before being added to the composite mix. The slurry was mixed in a heavy duty paddle mixer for 5 min, placed into steel moulds and hand trowelled to a flat surface.

All specimens were cured for 24 h under polyethylene sheeting, then removed from the moulds, dampened with water and wrapped in polyethylene for a further 6 days. At the end of this time the samples were unwrapped and stored in the laboratory until tested. The day of manufacture was defined as day zero for the time trials.

Tests were carried out on samples of the composite materials to determine their modulus of rupture and fracture energy. The modulus of rupture was measured in three-point bending at a span of 100 mm and a deflection rate of 0.5 mm min⁻¹ on an Instron testing machine. The fracture energy was determined using a General Electric puncture tester [18]. This test measures the energy required to force a pyramid-shaped indenter completely through a sample of specified thickness and thereby gives a comparative indication of the dynamic fracture behaviour of the

Τ.	A	В	L	E	V	Effect of	of	type	of	fibre	on	composite	proj	perti	es
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Type of fibre		MOR	(MPa)
		14 days	28 days
TMP, P. radiata (170° C), unwashed		5.9	7.1
TMP, P. radiata (170° C), washed		6.7	8.2
TMP, P. radiata (170° C), Na ₂ SO ₃ pretreated, washed		8.3	8.5
TMP, P. radiata (170° C), NaOH pretreated, washed			7.9
TMP, P. radiata (170° C), Na ₂ SO ₃ pretreated, washed, Bauer refine		9.6	
TMP, mixed eucalypt (170° C), $Na_2 SO_3$ pretreat, washed		8.1	8.6
Commercial Asplund TMP, P. radiata (170° C) unwashed*		6.3	7.1
Commercial Asplund TMP, mixed eucalypt (170° C) unwashed $*$		-	7.0
Commercial refiner groundwood P. radiata, washed [†]	Р	8.3	9.4
Commercial kraft <i>P. radiata</i> [‡]	Р	10.1	10.7
Commercial kraft mixed Eucalypt [‡]	Р	_	8.5
Cement [§]		4.7	5.5

P: indicates a paper making pulp. These pulps required reslushing before being added to the matrix slurry.

*Supplied by Hardboards Australia Limited.

[§]ARC Brand Type A.

composite material. The test is suitable for flat sheet materials but because the crack path is uncontrolled, it is not a measure of fracture surface energy.

The specimens for flexural testing were approximately 6 mm thick, 50 mm wide and 110 mm long while the specimens for fracture testing were approximately 6 mm thick and 150×150 mm. At least four specimens were tested for each value reported in the graphs or tables.

5. Results and discussion

The modulus of rupture at 14 and 28 days of 40 OPC:16 water:1 fibre by weight composites manufactured from a range of wood fibre pulps is presented in Table V. All the composites have improved strength compared with the cement paste matrix. In addition, the ductility of the composites is greater, although failure still occurs due to the propagation of one large crack. The fibres denoted as papermaking pulps all show a tendency to clump in the matrix slurry making their even distribution difficult.

The composite containing unwashed *P. radiata* TMP fibres had low strength and whole-fibre pullout occured during fracture. Extractives leached from the fibres into the surrounding matrix have inhibited the cure of the cement, resulting in poor matrix-fibre adhesion. Thoroughly washing the fibres increases the strength of the composites and changes the failure mode to a combination of fibre fracture and fibre pullout. Pretreating the chips with Na₂SO₃ enhances the strength of the composite while the use of NaOH has a deleterious effect. Since both the pulps were thoroughly washed it is unlikely that residual chemicals or extractives are causing this behaviour. It is more probable that the chemicals modify the surface of the fibre and in the case of Na₂SO₃ this results in an improved compatability between the fibre and the matrix. This result indicates that the chemical modifications of the surface of the wood fibre by the addition of a coupling agent may have a beneficial effect. In addition the composites containing the Na₂SO₃ treated fibres show a more rapid increase in strength than those containig washed fibres.

Partially collapsing the fibre, as occurs during Bauer disc refining has the effect of further increasing the composite strength and reducing the workability of the fibre-cement slurry. However, the almost fully collapsed high-yield refiner

[†]Supplied by Caxton Paper Mills Limited, New Zealand.

[‡]Supplied by Australian Paper Manufacturers Limited.



Figure 1 Development of composite strength with time.

groundwood pulp does not show the same strength increase. This is probably due, at least in part, to the large number of damaged (shortened) fibres in this type of pulp.

The high strength of the composite containing P. radiata kraft fibres demonstrates the advantages of having an extractive free pulp and fully collapsed fibres. Scanning electron microscopy of the fracture surfaces [19] revealed that the kraft fibres remained in a collapsed state in the cured composite while the lumens of the TMP fibres were fully open and empty. It follows, therefore, from the earlier analysis (Table I) that the kraft fibres have a higher effective tensile stiffness and strength in the composite than the TMP fibres. The relative yields of the two pulping processes (~45% kraft, ~90% TMP) means that, for equal weight of reinforcement, there are approximately twice the number of reinforcing fibres present in the case of the kraft pulp. However, the volume occupied by these kraft fibres is only one third that of the uncollapsed TMP fibres.

The eucalypt and pine TMP, and the eucalypt kraft composites have similar strengths while the pine kraft composite has markedly superior properties. The similar performance of the eucalypt kraft and TMP pulps may be due to the short length of the fibres limiting the width of crack that can be bridged. This is believed to be likely since the wood fibre-cement composites fracture by the propagation of one large crack with accompanying fibre pullout.

There was a complete absence of shrinkage cracking in all the composites containing wood fibre. This was in contrast to the unreinforced cement samples that were often so badly cracked in the moulds that testing was not possible.

It was decided that since the *P. radiata* kraft and the Na_2SO_3 pretreated and washed thermomechanical pulps represented the extreme cases of mechanical behaviour, the remainder of the study would be centred about these two types of fibre.

Fig. 1 shows the strength gain with time up to 28 days for the cement matrix and composites containing the two types of fibre. The rapid development in strength of the composites is marked and the flatness of the curves between 14 and 28 days is a further indication that neither of the pulps significantly inhibits the cure of the matrix.

The effect of the water-cement ratio on the strength of composites containing 2.5% by weight fibre was investigated and the results are presented in Fig. 2. These results have been corrected to take account of the amount of water absorbed by the fibre during the mixing of the slurry. This was done by saturating kraft and TMP fibres under vacuum and then drawing off all excess water with



Figure 2 Effect of water-cement ratio on composite strength.

a Buchner funnel. The water pick up was 140% for the kraft pulp and 220% for the TMP based on the dry weight of the fibres. The results show the marked superiority of the TMP fibres at low watercement ratios. This is believed to be due largely to mixing effects as is evidenced by the reduction in the spread of the results at high water-cement ratios. At the low water-cement ratios the kraft fibres clump badly while the TMP fibres disperse readily. In addition, the water contained in the lumens of the TMP fibres appears to be squeezed out during the mixing operation further lubricating the slurry. After mixing, this water is reabsorbed by the fibre before the cement hardens and therefore does not effect the water-cement ratio of the matrix.

In Fig. 3 the effect on strength of the weight of fibre in the composite is presented. Samples con-



Figure 3 Effect of fibre content on composite strength.



Figure 4 Development of composite impact energy absorption.

taining more than 6% kraft fibre were too weak to test. These results are for a corrected watercement ratio of 0.35. The superiority of the kraft fibre is seen at low fibre contents but at high fibre content the TMP fibre dominates.

The fracture energy of standard composites containing kraft and TMP fibres was measured (Fig. 4.) Although there is an error in these results due to thickness variations of up to $\pm 8\%$ in the samples, it is evident that the two composites behave in a similar manner. This result is unexpected since both composites fail by a combination of fibre fracture and pullout and the number of kraft fibres present in the composite is approximately twice that of the composite containing TMP fibres. The extra energy absorbed by the TMP fibres can probably be accounted for by the buckling of the fibres during pullout and fracture. Gordon and Jeronimides [20] have shown that the buckling of an open lumen wood fibre during tensile loading can absorb extremely large amounts of energy due to the collapse of the helical array of microfibrils in the fibre walls.

The strengths of the experimental composites fall well short of the theoretical results presented in Table III. This is due at least in part, to the difficulty of uniformly mixing large volumes of wood fibre into the cement matrix and to the high water-cement ratio of the matrix. Both these problems could be overcome in commercial production by using a high water—cement ratio slurry for mixing followed by dewatering and compaction. Since the wood fibres do not have a natural affinity for cement as do asbestos fibres some additives or surface treatment of the fibres may be required to prevent leaching of the cement from the composite during dewatering.

6. Conclusions

Although wood fibres have relatively poor mechanical properties compared to synthetic fibres their low cost, low density and low energy demand during manufacture make them attractive as reinforcement for cement matrix composites. The pulping process selected to remove the fibres from the parent wood will depend on economic and energy constraints and the type of composite material. The open lumen TMP fibre is the cheapest, consumes less resource, produces a lower density product and does not require prepulping before incorporation into the cement slurry. Composites produced using TMP fibres may not have the absolute properties of the kraft fibre composites but when allowance is made for density and pulp yields the reinforcing capacity of the open lumen fibre is similar to that of the collapsed fibre.

The closed lumen kraft fibre is more expensive, requires prepulping, is less efficient in terms of resource utilization but produces a product with higher absolute strength. This type of fibre would be more suited to products manufactured in a factory where prepulping and slurry dewatering techniques could be employed (e.g. manufacture of flat sheet). The open lumen fibre is probably more suited to field applications (e.g. use in concretes and mortars).

The use of coupling agents or chemical modification of the wood fibre surface in order to optimize the mechanical properties of the composites is worthy of further investigation.

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Received 9 October 1979 and accepted 8 January 1980.