JOURNAL OF MATERIALS SCIENCE 19 (1984) 3355–3359

Bonding in wood fibre–cement composites

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Much has been written about the mechanical properties of wood-fibre reinforced cement composites (WFRC), but little has been reported on their response to moisture or how the composite materials are bonded. The present observations are consistent with the hypothesis that hydrogen bonds and/or hydroxide bridges play a major role in the mechanical performance of wood fibre–cement composites.

1. Introduction

The effect of moisture on WFRC composites was noted when collaborative work between CSIRO and James Hardie Industries, Australia, revealed variation in the mechanical properties of samples tested under different conditions [1]. To solve this problem, a series of standard conditions for testing was established and the effect of moisture on composite performance studied [2].

Mai and co-workers [3] proposed that dry WFRC samples failed in flexure because of whole-fibre pullout. However, our scanning electron microscopy (SEM) studies [4, 5] indicated that fibre fracture is the major mechanism taking place at low moisture content with fibre pullout increasing in importance as the moisture content of the test sample is increased [2]. Mai and co-workers [6] have more recently confirmed our earlier observations and agree that fibre fracture is the likely mechanism involved with failure in flexure. This prompted us to present SEM data to support our current hypothesis on the mode of bonding in WFRC composites referred to in the earlier work [2].

2. Materials and methods

The wood fibres used in this study were from commercial *Pinus radiata* kraft lap pulp obtained from Kinleith, New Zealand.

The samples of WFRC (containing a matrix of ordinary Portland cement and ground silica of equal weight) were prepared by a slurry/vacuum dewatering process followed by autoclaving as reported in an earlier study [2]. Samples were conditioned (a) in a control room with relative humidity (r.h.) of 50 ± 5% and 22 ± 2° C; (b) by heating in an oven at 100 to 105° C for 24 h then cooled in a desiccator, and (c) by soaking in water for 48 h with excess water being removed with a cloth prior to testing.

Specimens after fracture were coated with gold under vacuum and the fracture surfaces examined with a Cambridge S600 scanning electron microscope operating at 25 kV.

3. Results and discussion

3.1. Fracture surface studies using SEM

The fracture surfaces of WFRC samples containing 2 wt% of kraft *P. radiata* fibres are shown in Figs. 1 to 3. Most of the fibres protruding from the fracture surface of a sample preconditioned in an oven at 105° C for 24 h before testing have broken ends (Fig. 1a). The matrix material attached to their surfaces (Fig. 1b) suggests a strong fibre-to-matrix bond [5].

By contrast, Fig. 2a shows the fracture surface of a sample from the same preparation, which has been preconditioned in water for 48 h before testing. Fig. 2a suggests fibre pullout has taken place. Mai and co-workers [6] and Mindess and Bentur [7] stated that a pullout mechanism took place with broken fibres being rare. Both groups examined composites with high fibre loadings (> 8%). At high fibre loadings, difficulties in coating the numerous fibres evenly, achieving sufficient depth of field and eliminating discharge from the electron beam all contribute to poor SEM data. To overcome this, samples of low fibre

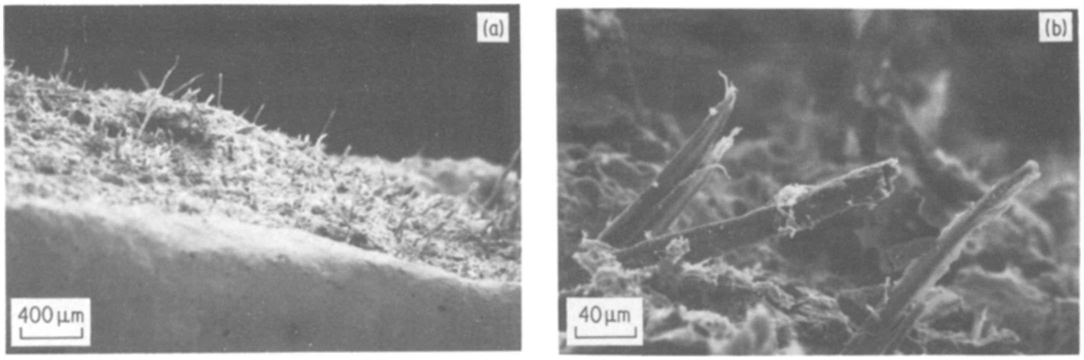

Figure 1 (a) SEM showing fracture surface of WFRC which had been preconditioned at 100 to 105° C for 24 h. (b) As (a) but higher magnification.

content were examined. Mindess and Bentur [7] used only photographic evidence and stated that even at 18× magnification, lack of focus restricted their observations due to the unevenness of the surface.

Mai and co-workers [6] concluded that broken fibres were rarely observed in wet samples, yet typified this by showing an SEM ([6], Fig. 8) which contained three fibres, *one of which had a fractured tip.*

We observed, as have others [6], that when examining the fracture surfaces of samples preconditioned in water, some fibres are twisted (Fig. 2b). This phenomenon has been observed by Page *et al.* [8] with isolated single wood fibres tested under axial tensile strain, and is due to collapse of the hollow fibre. In the cement matrix, it has been established that the collapsed kraft fibres remain collapsed and do not swell open during fabrication [4, 9]. But more important than the observation that the wet fibres were twisted and largely devoid of adhering matrix (weak fibre-to-matrix bond) is the fact *that many fibres were broken* (even the tips of some of the "long fibres" (see Fig. 2b).

Fig. 3a shows the fracture surface of the WFRC containing 2 wt% of kraft *P. radiata* fibres, conditioned at 50 ± 5% r.h. and 22 ± 2° C. Both fibre fracture and fibre pullout appear to have taken place (Fig. 3a). Higher magnification (Fig. 3b) shows that considerable damage to some fibres of relatively short length has taken place while other fibres have been pulled out from the matrix.

Andonian *et al.* [3] assumed linear elastic fracture mechanics (LEFM) could be applied to WFRC composites and proceeded to derive the critical length of a wood fibre. Their findings suggested a value of 18 to 23 mm and, as the *P. radiata* fibres have an average length of 3 mm, all fibres should experience pullout and not be fractured during sample failure. Our observations [2, 5] suggest that the non-uniform nature of the wood cellulose fibre does not lend itself to analysis by conventional LEFM, and as fibre fracture does take place the

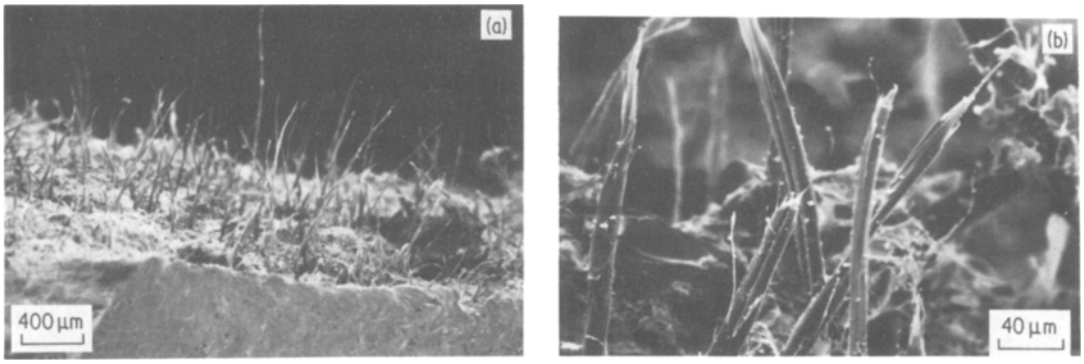

Figure 2 (a) SEM showing fracture surface of WFRC which had been preconditioned by soaking in water for 48 h. (b) As (a) but higher magnification.

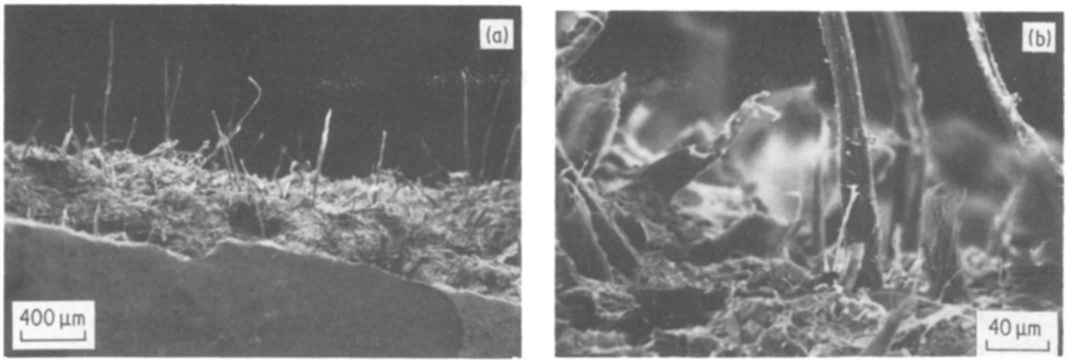

Figure 3 (a) SEM showing fracture surface of WFRC which had been preconditioned in an atmosphere with a relative humidity of 50 ± 5% at 22 ± 2° C. (b) As (a) but higher magnification.

"critical length" must be close to or less than 3 mm.

Mindess and Bentur [7] conclude from their studies of WFRC that in the wet state such composites are completely notch insensitive and at the least only slightly notch sensitive to the dry state. Thus they confirm the view that the use of LEFM is not a valid approach as it has been shown [10] that notch sensitivity is a necessary though not sufficient condition for the applicability of LEFM to cementitious materials.

To model this system we are currently studying the pullout of single slivers of sisal from a cement matrix [11]. It has been demonstrated from load deflection curves for single slivers that as the length of fibre embedded is increased the mode of failure changes from one of fibre pullout to one of fibre fracture. The change of failure mechanism occurs when the aspect ratio of a sliver has a value of 150 ± 50. The aspect ratio of the single *P. radiata* fibres is found within this range of values and confirms the fibre fracture mechanism.

The fracture surface of the commercial product Hardiflex II* (> 8 wt% wood fibre), tested at 50% r.h. and 22 ± 2° C, resembles a "forest" of fibres (Fig. 4a) and could be ascribed to fibre pullout; however at higher magnification (Fig. 4b) large numbers of broken fibres are observed in agreement with laboratory samples (Fig. 3b).

3.2. Mechanical properties

The testing of laboratory, pilot-scale and production-run WFRC samples has led to a number of conclusions which have been reported in a series of specific studies [2, 9, 12–14].

There is slight variation between the flexural strength of samples of the same composition when tested after being dried in an oven at 100 to 105° C for 24 h, or after curing at 50 ± 5% r.h. and 22 ± 2° C. However, samples soaked in water

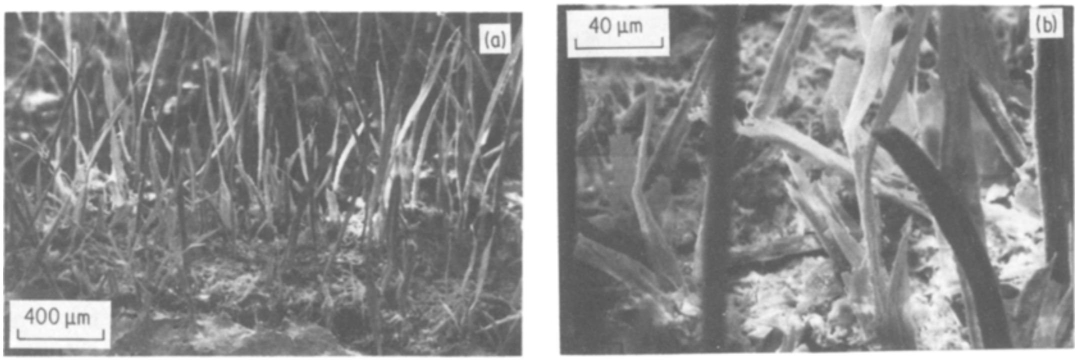

Figure 4 (a) SEM showing fracture surface of WFRC produced commercially and preconditioned in an atmosphere with a relative humidity of 50 ± 5% at 22 ± 2° C. (b) As (a) but higher magnification.

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*Hardiflex II is a trademark for a commercial product manufactured by James Hardie Industries Limited, Australia. The product is made from ordinary Portland cement, silica, cellulose fibre, water and is free of asbestos fibre.

for 48 h and then tested were found to retain between 60 and 80% of the dry strength, depending on the fibre content.

By contrast, load/deflection curves showed that the post-cracking ductility or fracture toughness of a range of WFRC samples is very low (i.e. brittle) for samples dried in an oven and high (i.e. ductile) for samples tested either when wet or after curing at 50 ± 5% r.h. and 22 ± 2° C.

Oven dried samples appeared strong but brittle with mainly fractured fibres, whereas the samples tested wet were weak but tough with considerable fibre pullout [2].

The samples tested under ambient conditions possessed both strength and toughness; this result suggests that a number of micromechanisms might be taking place during fracture of the sample.

3.3. Bonding mechanisms

The interfacial bond itself can be physical or chemical in nature, or a combination of both. Too strong a bond between fibre and matrix results in a brittle material which has strength, whereas a weak bond results in a tough material lacking strength.

The mechanical performance of a WFRC composite is therefore directly related to the nature and properties of the fibre–matrix interface and to achieve a strong yet tough material, a compromise must be sought.

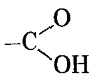
The chemistry and morphology of the matrix material have been well documented [15] and will not be considered further, apart from stating that cement is a strongly alkaline material (pH > 12.5) and contains metal hydroxy groups, such as

$$-\underset{|}{\overset{|}{\text{Ca}}}-\text{OH},\ -\underset{|}{\overset{|}{\text{Si}}}-\text{OH},\ -\underset{|}{\overset{|}{\text{Al}}}-\text{OH} \text{ and } -\underset{|}{\overset{|}{\text{Fe}}}-\text{OH}$$

(due to hydration and hydrolysis of silicates, aluminates and to a lesser extent ferrites of calcium that are present in the cement matrix). Cellulosic fibres such as wood fibres contain covalent hydroxyl groups,

$$-\underset{|}{\overset{|}{\text{C}}}-\text{OH}$$

either phenolic (from residual lignin) or alcoholic (from the cellulose component) and carboxylic groups,



$$-\text{C}\begin{matrix}\nearrow \text{O} \\ \searrow \text{OH}\end{matrix}$$

due to oxidation of end groups [16]. Hydrogen bonding and/or hydroxide bridges may play a major role in the bonding of WFRC composites. The chemical implications will not be considered quantitatively; it suffices to say that hydrogen bonds form *between fibres* or *between fibres and matrix.*

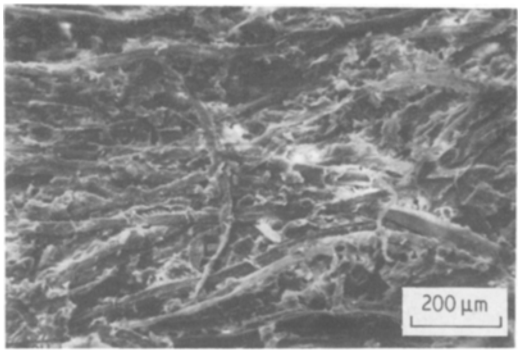


Figure 5 SEM of a cleaved section in the plane of the commercial fibre cement sheet showing numerous fibre-to-fibre crossings.

If a commercial sheet of WFRC is cleaved between laminations, (see Coutts [12] for production method) SEM shows there are considerable fibre to fibre contacts (Fig. 5). The matrix material is seen to be firmly bonded to the cellulose fibres (Fig. 6a) and a higher magnification it can be observed that even the very fine microfibrils (diameter of < 0.2 μm) are bound to the matrix.

Wet or dry, a wood fibre has about the same tensile strength *but* its stiffness is about ten times greater when dry. Thus, an oven dried WFRC has stiff, highly contorted fibres (see Fig. 5) locked into a rigid cement matrix and held together by a large number of hydrogen bonds or hydroxyl bridged sites. This system when stressed can transfer the stress from the matrix to the fibres via the many interfacial bonds and hence sufficient stress may be passed onto the fibre after matrix cracking to cause the reinforcing fibre to fracture in tensile load. On the other hand, in a soaked sample, the hydrogen bonds between fibres or between fibre and matrix are destroyed (by insertion of water molecules between the bridging hydroxyl groups); and, at the same time, the cellulosic fibres are swollen by water absorption. Under stress this system allows the fibres to move relative to the matrix. However, due to the pressure of swelling and the highly contorted assemblage of fibres, considerable frictional forces are developed. If the forces are effective over suf-

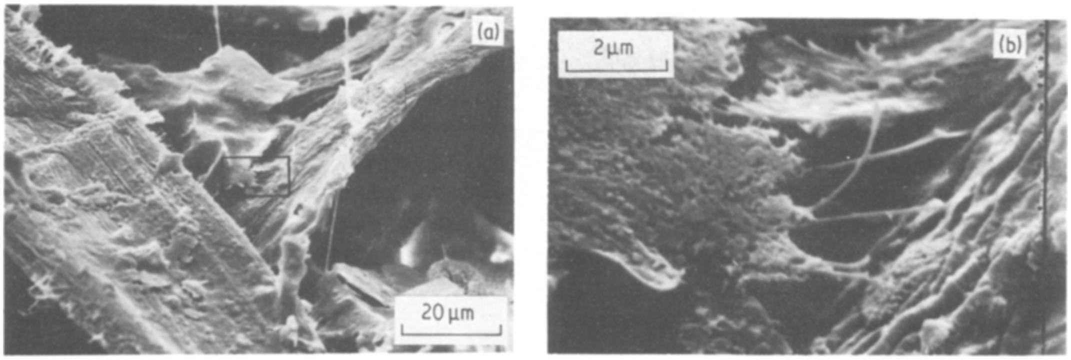


Figure 6 (a) SEM showing fibre-to-fibre and fibre-to-matrix bonding in a commercial fibre cement sheet. (b) As (a) but higher magnification.

ficient length of a fibre they can result in the fibres being loaded to failure [12]. Hence we see in Fig. 2b considerable numbers of fractured fibres even in samples tested when wet.

4. Conclusions

The effect of moisture on the mechanical properties of WFRC composites can be explained in terms of hydrogen bonds and/or hydroxide bridges having a major role in the bonding of fibre to matrix.

Oven dried WFRC composites are strong but brittle with mainly fibre fracture taking place, whereas composites tested wet are weaker but tougher with mainly fibre pullout taking place.

The forces involved under ambient conditions are such that the WFRC composites can display high values of both flexural strength and fracture toughness. The balance of fracture mechanisms between the extremes of fibre fracture and fibre pullout are fortuitously balanced in the WFRC system allowing maximum utilization of the fibre reinforcement.

Acknowledgement

We wish to thank James Hardie Industries Limited of Australia for financially supporting this work. This article is published by permission of the Commonwealth Scientific and Industrial Research Organisation (CSIRO).

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Received 14 November 1983
and accepted 16 January 1984