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ASSESSMENT OF INTERFACIAL MICROSTRUCTURE AND BOND PROPERTIES IN AGED GRC USING A NOVEL MICROINDENTATION METHOD

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ABSTRACT

Changes of microstructure and properties in the interfacial zone of glass fibre reinforced cement (GRC) under the effect of ageing were investigated. A novel technique based on a microindentation apparatus was developed and successfully used to carry out microstrength testing in the interfacial area and push-in tests on selected individual fibres within a strand. By continuously monitoring load vs. displacement, the new technique allowed the microstrength to be measured in small, porous areas of the fibre-matrix interfacial zone, and particularly within the glass fibre strand/bundle. The results showed that the embrittlement of aged GRC was closely associated with a substantial increase of the microstrength values within the fibre bundle during the ageing process. It was also revealed that a wide range of bond properties existed within the fibre strand. The resistance to fibre sliding was much greater at the outer filaments than at the inner central filaments of the fibre strand/bundle. © *1997 Elsevier Science Ltd*

Introduction

One of the major differences between GRC and most other cement-based composites is its very complex microstructure. The basic reinforcing element in GRC composites is a strand or fibre bundle, instead of a single fibre as in many other composites. An important feature associated with the bundle structure of the basic reinforcing elements is the continuous change of interfacial microstructure with age when exposed to a wet environment. It has been recognised that the microstructural changes at the fibre strand-matrix interface and fibre-fibre interface within the fibre strand contribute significantly to the ageing embrittlement and the reduced long-term performance of GRC, as discussed previously (1–4).

Assessing which changes of the microstructure and interfaces in GRC may lead to the embrittlement phenomenon is not an easy task. Most of the previous research involved the examination of fractured surfaces or exposed interfaces using the scanning electron microscope (SEM). Though being convenient to use, the SEM technique does have its own

limitations. The SEM observations can provide only qualitative information, which is difficult to relate to the interfacial properties. In addition, Cohen and Constantiner (5) noted that the detection of interfacial films with a SEM is difficult because the film seems to be very brittle and fragile, thus can fracture and shatter during sample preparation, leaving no traces. Majumdar et al. (1) also pointed out that different types of interfacial microstructure may be observed in different parts of the sample.

Pullout tests of a single glass rod or fibre strands from a cement matrix were carried out by a few researchers (6–9) in order to assess the actual magnitude of the interfacial bond. However, it was found that after a period of wet ageing a substantial portion of the fibres ruptured rather than pulled out during the test, thus limiting the applicability of the pullout test to young/unaged composites. Another significant shortcoming of the pullout test is its inability to evaluate the bond at the fibre-fibre interface within a fibre bundle.

In an attempt to overcome the above mentioned difficulties, a new technique based on an unique microindentation apparatus was developed and successfully used in this study to carry out microstrength testing in the fibre strand-matrix interfacial zone and particularly within the fibre strand/bundle, as well as push-in tests on selected individual filaments. This paper describes the principles of the test methods and presents preliminary test results for GRC specimens with different matrix modifications. The change of the flexural properties of composites due to the various matrix modifications and ageing periods was related to the microstructural properties in the interfacial area and especially within the fibre strand/bundle.

Materials and Methods

Specimens

A standard-grade ordinary Portland cement (Blue Circle) and an alkali-resistant glass fibre (Nippon Electric Glass) were used as the basic materials for matrix and reinforcement, respectively. A microsilica slurry (Elkem Chemicals), an acrylic polymer latex (Forton BV), and a metakaolinite powder (CR-PAM, St. Gobain Co.) were used for different modifications of the OPC matrix. Details of the test series used are given in Table 1.

Two types of specimens were prepared, one for a four-point bending test and one for the microindentation test. The specimens for the four-point bending test were 110 × 20 × 9 mm in size and contained two parallel layers of glass fibre strands spaced equally across the 9-mm depth of the section. Ten fibre strands were evenly spaced across the 20-mm width of the specimen in each of the two layers, thus representing a continuous reinforcement. Arrange-

TABLE 1
Test Series

Test series	Matrix	Fibre reinforcement
A	OPC as a reference	AR-glass fibre in the form of continuous strand
M1	OPC + 10% Acrylic polymer (addition)	
M2	OPC + 10% Microsilica (replacement)	
M3	OPC + 25% Metakaolinite (replacement)	

ment of the four-point bending test was closely related to that used by Bentur (10), and the details were given previously (11).

The specimens for the microstrength and the fibre push-in tests were prepared in four stages:

1. Slices were cut (approximately 15–20 mm thick) from the specimen of the four-point bending test perpendicular to the fibre axis;
2. Slices were embedded in a moulding resin to form a disc specimen (the exposed fibre ends were covered using adhesive tapes to avoid penetration of the resin into the fibre area);
3. Specimens were ground and polished with silicon carbide papers and diamond particles to obtain a very flat and smooth surface finish; and
4. Specimens were cleaned in an ultrasonic bath to remove the dust and diamond particles left in the voids or porous areas after the polishing.

All of the specimens used in this study had a fixed w/c ratio of 0.35. Following an initial curing of 7 days in water at 20°C, the specimens were stored in water at 60°C to accelerate the ageing. Tests on the specimens were carried out after 7 days and 42 days of the accelerated ageing. The specimens were identified as 7da and 42da, respectively.

Microindentation Apparatus

The apparatus MicroTest 200 is an instrument used in the continuous monitoring of a load-displacement relationship in the load range of 0–20 N and displacement range of 0–50 μm with resolutions of 0.7 mN and 1.7 nm, respectively. The operating principle of the test apparatus and the Continuous Depth Recording measurement are illustrated in Figure 1.

As shown in Figures 1a and 1b, the MicroTest 200 continuously monitors the displacement of a probe/indenter in contact with a surface during a programmed load cycle. A pendulum pivoted on frictionless bearings was used for the application of load. When a current is passing through a coil mounted at the top of the pendulum, the coil is attracted towards a permanent magnet, producing motion in the indenter towards the specimen and into the specimen surface. The displacement of the indenter or the depth of the penetration into the specimen is continuously monitored and recorded by the change in the capacitance of a parallel plate capacitor. A Load-Depth curve, as shown in Figure 1c, is thus generated during a loading-unloading cycle, which can be used to evaluate the micromechanical properties of the test area. Other features of the instrument include:

1. Different sizes and shapes of indenters, allowing tests to be carried out on a very small porous area as well as on different sizes of fibres or fibre groups;
2. An optical fibre-linked microscope, high-resolution video camera, and monitor, which were used to locate the fibres or the test points and align the specimen beneath the indenter;
3. Precision X-Y-Z movement control for mounting the specimen, the X-Y-Z resolution/travel being 0.02 μm/50 mm; and
4. Personal computer control, monitoring, and data manipulation.

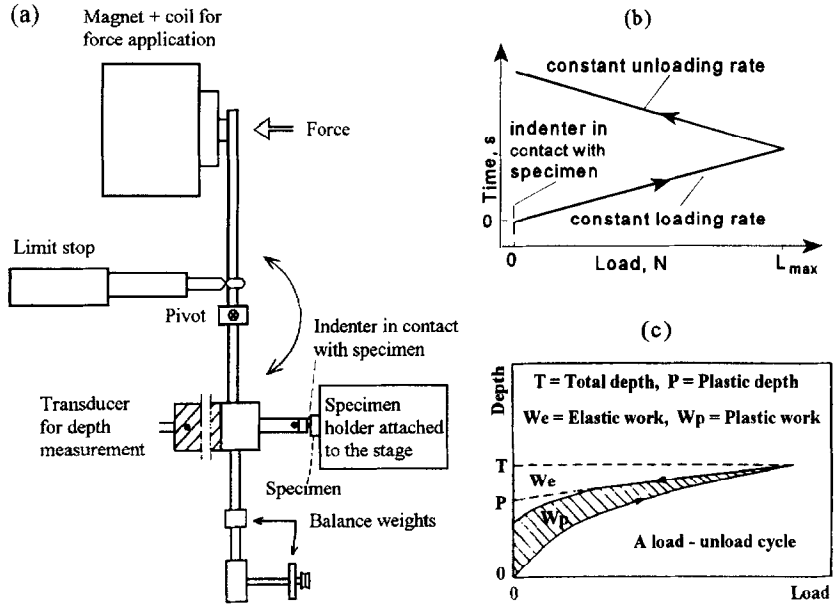

FIG. 1.
Illustration of the operating principle and the Continuous Depth Recording measurement of the MicroTest 200 apparatus.

Microstrength Measurement

Using the microindentation apparatus, microstrength measurements were made in the fibre-matrix interfacial zone, as well as within the fibre strand/bundle. As shown in Figure 2a, a sharp 90° diamond indenter tip (i.e., corner of a cube) was moved to be just in contact with the surface of a selected area. The indenter was then pushed against the specimen until a designated maximum load of 500 mN was reached before being unloaded to zero. A special materials micromechanical parameter (microstrength, as defined in Eq. 1) could be calculated from the Load-Depth curve with the following equation:

$$\text{Micro-strength} = L_{max}/(kP^2) \tag{1}$$

where, L_{max} = Maximum load; P = Plastic depth of the indentation; k = indenter tip geometry factor.

The indenter tip geometry factor k in Eq. 1 is a constant independent of the depth for an ideally perfect indenter tip geometry. In the case of imperfect tip geometry, k becomes a function of the depth and calibrations have to be made to determine the values of k at different depths. To avoid the determination of k, or in the absence of information about the indenter tip geometry, relative microstrength values can be easily calculated instead by comparing one Load-Depth curve with another obtained from a reference sample at the same plastic depth. A homogeneous perspex material was used as the reference for the determination of relative microstrength values in this study.

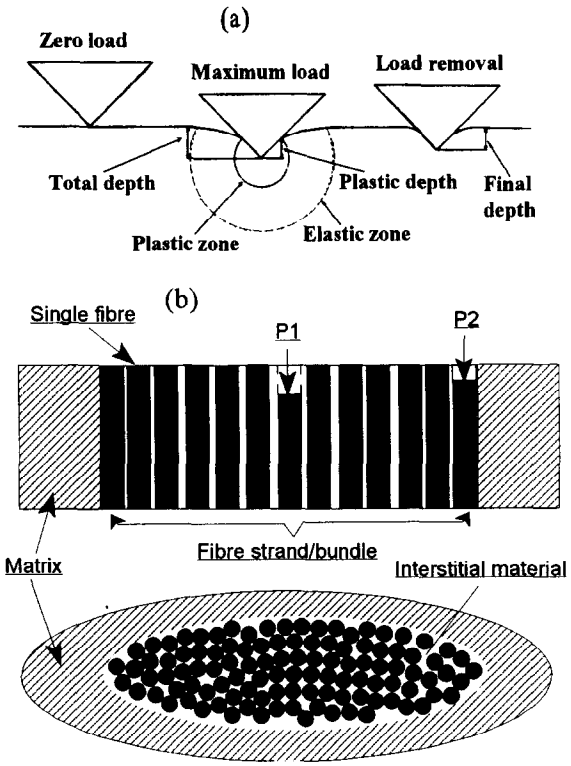

FIG. 2.
Schematic diagram of (*a*) the microstrength test and (*b*) the fibre push-in test.

Fibre Push-in Test

The fibre push-in test differs from the fibre pull-out test mainly in that the fibre is pushed into or through the matrix rather than pulled out. In many cases, particularly for very small and brittle filaments or fibre bundles, the push-in test offers a clear advantage over the conventional pull-out test. For the fibre push-in test, the specimen can be cut from a practical, commercially produced composite that has been subjected to different ageing/service conditions. Generally, the application of load leads to a progressive debonding that starts at the top end of the fibre and a sliding of the fibre in the debonded region against frictional resistance at the interface.

The resulting Load-Depth curve in the push-in test can be used to assess the bond properties of the fibre tested. In this paper, three parameters from the Load-Depth curve (T, total depth; P, plastic depth; and Wp, plastic work; Figure 1c) were used to compare the bond properties of various fibres tested. Alternatively, calculation of interfacial properties from the Load-Depth curve could be done by fitting theoretical predictions derived from micromechanical modelling of the debonding and sliding process to the experimental data. Several theoretical models with various degrees of approximation have been proposed and used for such a purpose recently (12–14).

As shown in Figure 2b, quantitative information about the bond properties at the different

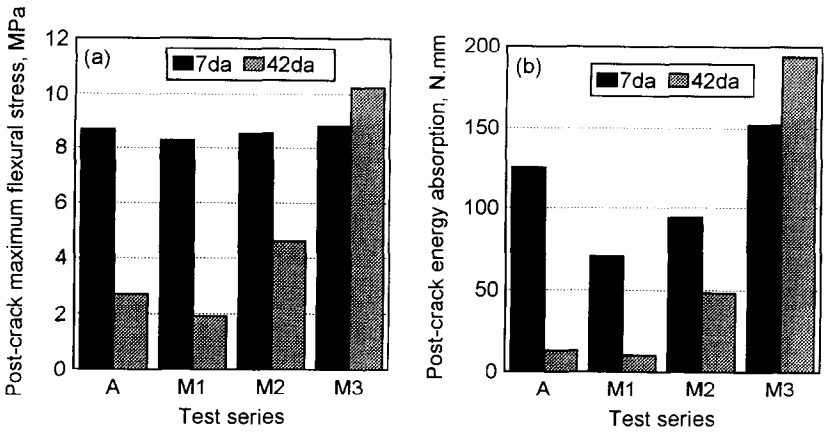

FIG. 3.
Effect of ageing and matrix modifications on (*a*) flexural strength and (*b*) flexural toughness.

interfaces can be obtained by testing individually the outer filaments (i.e., the fibres at the perimeter of the interface with the matrix) and the inner filaments within the strand/bundle, respectively. A very thin (approximately 6–8 μm in diameter) and relatively flat tungsten indenter tip and a maximum load of 100 mN were used to avoid damage/splitting of the filament during the push-in test. A series of calibrations were carried out to eliminate the possible penetration of the indenter into the sliding fibre and machine compliance contributions from the Load-Depth curve.

Results and Discussions

Flexural Properties

The flexural test results for the composite specimens with different matrix modifications and ageing periods are presented in Figure 3. Two principal parameters, namely post-crack maximum flexural stress and post-crack energy absorption, were calculated from the load-deflection curves of the four-point bending test (11) and were used to serve as an estimate of the flexural strength and toughness of the composites, respectively.

For the reference composite (series A), results in Figures 3a and 3b show that the post-crack flexural strength and toughness decrease considerably with accelerated ageing. The specimen retained roughly one third of the initial post-crack flexural strength and lost almost all the initial improvement in toughness after 42 days of the accelerated ageing. The results in Figure 3 indicated that the matrix modifications by the acrylic polymer (series M1) or by the microsilica (series M2) were not very effective in improving the ageing performance of the GRC, and at best could only slow down the ageing process. The matrix modification by metakaolinite (series M3), however, did significantly improve the properties of the aged composites. It appeared that the post-crack flexural strength and toughness of the composite still remained at a high level after substantial periods of ageing.

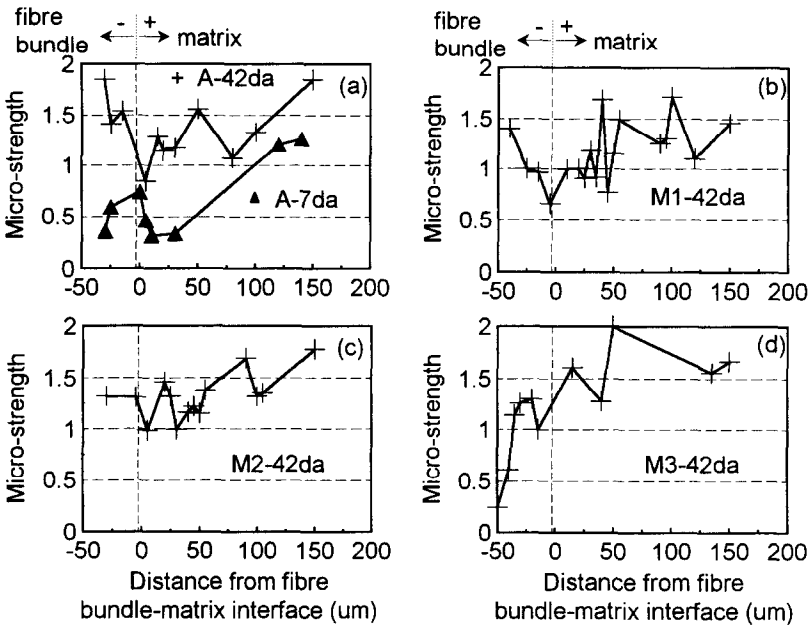

FIG. 4.
Microstrength distribution patterns in the interfacial area and within the fibre strand for specimens of (*a*) A-7da and A-42da, (*b*) M1-42da, (*c*) M2-42da, and (*d*) M3-42da.

Microstrength Distribution Patterns Around and Within Glass Fibre Strands

The changes of interfacial microstructure in the composite due to ageing and the matrix modifications were assessed by the microindentation testing in the fibre-matrix interfacial zone and within the fibre strand. Results are presented as the micro-strength distribution graphs in Figure 4. The negative distance values in the graphs indicate the test points that fell within the overall cross section of the fibre strand. With the test parameters chosen, a single indentation within the fibre bundle usually affected an area containing around 15–20 filaments (or approximately 40 × 60 μm^2). The results are averages of two to six test values from the indentation points that have the same distance from the fibre-matrix interface.

As shown in Figure 4, the microstrength distribution exhibited a "trough" at a small positive distance from the fibre-matrix interface, indicating a weaker/softer area for the reference composite (series A). A comparison of the graphs for A-42da and A-7da in Figure 4a shows that the microstrength values at the fibre-matrix interfacial zone (10–50 μm) increased considerably with ageing. This change is well in agreement with the phenomenon observed in other fibre-cement and aggregate-cement interfaces (15–16).

It is interesting to note that the most significant change in the microstrength distribution pattern due to ageing happened within the fibre strand (i.e., −50–0 μm). For the younger specimen (A-7da) the microstrength value was at its lowest near the centre core of the fibre bundle and increased gradually toward the bundle-matrix interface. This trend was reversed for the aged composite; the graph of A-42da showed that the highest value of microstrength occurred at the centre core of the fibre bundle and decreased toward the bundle-matrix

interface. This change of microstrength distribution within the fibre strand/bundle indicated that the microstructural characteristics in the interstitial spaces and fibre-fibre bond changed greatly during the ageing process.

To understand the reason for such changes and to gain further insight into the ageing mechanism from the results, a discussion of the implications and factors involved in the determination of microstrength values within the fibre bundle is necessary. First, by definition the microstrength value is very similar to the Vickers microhardness number in conventional microhardness testing. Thus, like other indentation tests, the result of a microstrength test indicates the resistance against plastic deformation of the test area. For the test carried out in the interfacial area of the GRC composites, the porosity, composition, and hardness/strength of the different materials in the test area determine the microstrength values. The interfacial bond (adhesional or frictional) that resists the relative displacement between fibre and matrix or different fibres also contributes to the microstrength values.

It is reasonable to believe that no significant changes in hardness of the glass filament occurred due to the ageing. This is because the hardness in the axial direction of the glass filament is not much affected by small local defects (or notches) on the fibre surface, although the tensile strength is. Furthermore, the glass fibre itself is much harder than the cement paste, and when the test point moves gradually from the bundle-matrix interface toward the centre of the bundle, more internal filaments of the bundle will be hit by the indentation, thus contributing to the microstrength value measured. This means that if the microstructure were the same at the fibre-matrix interface as at the fibre-fibre interface within the fibre strand/bundle, the microstrength should have the highest value at the centre of the bundle and decrease toward the bundle-matrix interface. Thus, the microstrength distribution patterns similar to that of the younger composite (A-7da) would indicate that the microstructure and interfacial bond were weaker within the fibre bundle than at the bundle-matrix interface. The change of the microstrength distribution pattern within the fibre strand due to the ageing discussed above, therefore, indicated that the porous and weaker microstructure within the fibre bundle that existed in the early stage of ageing had become dense and strong in the later stage of the ageing, possibly due to the interstitial spaces being filled up or replaced by denser and stronger cement hydration products.

Similar discussions could be easily applied to the different cases of matrix modifications in Figures 4b to 4d. Graphs in Figures 4b and 4c show that the matrix modification by the acrylic polymer (M1-42da) or microsilica (M2-42da) had little effect on the overall microstrength distribution pattern of the aged composites. The matrix modification by the metakaolinite (M3-42da), however, appeared to have changed substantially the microstrength distribution pattern of the aged composite. In particular, the microstrength value at the centre core of the fibre strand was very low and increased towards the bundle-matrix interface, i.e., a trend similar to that of A-7da, although microstrength in the fibre bundle-matrix interfacial zone was relatively high compared to that in the reference composite.

Results in Figure 5 show relative values of the microstrength at the centre core of the fibre strands for the different specimens. As shown in Figure 5 for the younger composites (A-7da and M1-7da), the microstrength values at the centre core of the fibre strand were less than 30% that of the aged composite (A-42da). The matrix modifications by microsilica (M2-42da) or by acrylic polymer (M1-42da) only modestly reduced or delayed the increase of the microstrength at the centre of the strand/bundle. However, for the matrix modifications by metakaolinite (M3-42da) the microstrength at the centre of the fibre strand remained very low

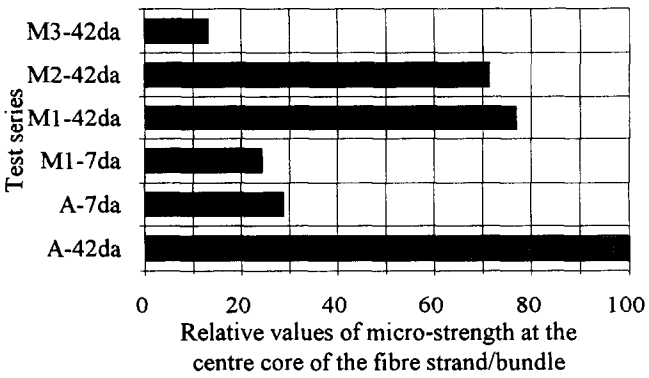

FIG. 5.
Comparison of microstrength values at the centre core of the fibre strands/bundle.

(15–25%) even after substantial periods of ageing, suggesting a softer or flexible fibre core being retained due to such a modification.

Comparing the results of the microstrength in Figures 4 and 5 with those of the four-point bending tests in Figure 3, it is interesting to find that the trend of the change of mechanical performance (strength and toughness) of the composites due to ageing and the different matrix modifications can be closely related to and is well in agreement with the relative magnitudes of the microstrength at the centres of their fibre strands/bundles. The embrittlement of the composite was accompanied by a substantial increase of the microstrength value at the centres of the fibre strands. This implies that the fibre-fibre interface within the fibre bundle in GRC composites plays a much more important role than has been previously assumed in controlling the performance of the aged GRC.

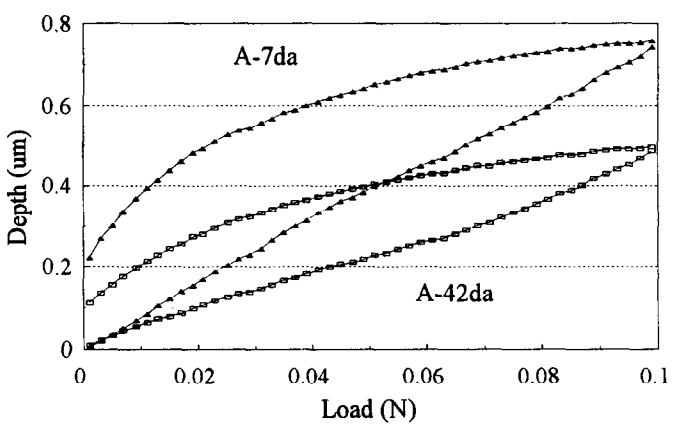

FIG. 6.
Typical Load-Depth curve of the push-in test on individual filaments. (Sample curve compiled from the outer filaments at the edge/perimeter of a strand/bundle.)

TABLE 2
Results of the Single Fibre Push-in Test

Location of the filament tested	A-7da			A-42da		
	T (nm)	P (nm)	Wp ($\times 10^{-9}$ J)	T (nm)	P (nm)	Wp ($\times 10^{-9}$ J)
Outer individual filament separated from a strand	295	159	7.8	261	177	6.1
Outer filament at the edge of a strand	718	466	21.0	472	347	15.0
Inner filament at a distance of 15–20 μm from matrix	692	587	27.2	634	528	16.2
Inner filament at the centre of a loosely packed strand	841	789	43.8	684	552	24.3
Inner filament at the centre of a tightly packed strand	1856	1688	80.1	1645	1502	50.6

Values are averages from the testing of 2–6 filaments.

Fibre Push-in Test to Assess the Bond Properties

Preliminary push-in tests on selected individual glass filaments were carried out in order to obtain quantitative information about the bond properties at the fibre-matrix interface and at the fibre-fibre interface within the bundle. Typical Load-Depth curves obtained from the fibre push-in test on the reference specimen (A-7da and A-42da) are presented in Figure 6. Results of the push-in test on selected fibres at different locations of the fibre strand/bundle are given in Table 2.

The results in Figure 6 and Table 2 clearly demonstrate that the resistance to fibre sliding increased significantly due to ageing. The results in Table 2 also indicate that a wide range of bond properties existed in different locations of a fibre strand/bundle. The resistance to sliding was much higher at the outer filaments than at the inner central filaments. Furthermore, the results appear to suggest that the tightness of the filament packing within a strand/bundle plays a very important role in controlling the bond properties of the inner fibres and the ageing performance of the composites. This finding is in agreement with previous SEM observations (17) that loosely packed strand usually showed a complete fibre fracture upon failure whereas the densely packed strand showed a telescopic or partial fracture/partial pullout failure in a tensile testing. The loosely packed strand/bundle was also found to become embrittled sooner than one that was tightly packed during the ageing.

Conclusions

A new technique based on an unique microindentation apparatus was developed and successfully used in this study to carry out microstrength testing in the interfacial area and push-in tests on selected individual filaments. The microstrength test proved to be a very useful method for the assessment of microstructure and interfaces in aged GRC. Particularly, the microstrength distribution pattern within the fibre strand/bundle can provide overall quantitative information about the behaviour of the reinforcement and was found to be

strongly related to the ageing performance of the GRC composites. The test results showed that the embrittlement of the aged GRC was closely associated with a substantial increase of the microstrength value at the centre core of the fibre bundle during the ageing process.

The fibre push-in test, for the first time, allow the bond properties at both the matrix-fibre interface and the fibre-fibre interface within the fibre strand/bundle in an aged GRC to be determined. The preliminary results revealed that a wide range of bond properties existed in the glass fibre strand. The resistance to fibre sliding was much greater at the outer filaments than at the inner central filaments of a fibre strand/bundle.

Acknowledgments

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