The three-fibre method for measuring glass fibre to resin bond strength

P. Järvelä, K.W. Laitinen, J. Purola and P. Törmälä

This paper presents a method developed to measure the bond strength of a fibre/resin bond. Equipment has been developed to enable the test to be carried out in a normal tensile testing machine, and the results to be followed by scanning electron microscopy. Bond strengths obtained by this method are in agreement with earlier results. There are, however, some problems related to the method, including reproducibility of the test, determination of the degree of polymerization of the resin and the use of very thin fibres. However, the method has been proved a very quick and relatively accurate indicator of the strengths of fibre/resin bonds.

Key words: adhesion tests; tensile testing; microscopic analysis; glass fibres; adhesives; 3-fibre method.

The tensile strength of a fibre-reinforced composite is mainly determined from the properties of the finished material[1-3]. This method is advantageous in that the results can be directly incorporated into practical designs; on the other hand, the results are dependent on the manufacturing process used.

The results of adhesion strength tests between matrices and fibres have recently become available. These tests can be divided into two groups: one in which the test material used is of massive size, *eg* boards, cylinders and rods[4-7], and the second in which the tests are performed on single fibres[8-9]. The first method has several weaknesses if the fibre/resin bond is considered. Unhomogeneity (*ie* porosity) in the resin layer and its thickness variations, are the main causes of error.

This study presents the development of testing equipment[8] used for determining the strength of single fibre/resin bonds at the Institute of Materials Science of the Tampere University of Technology. It also presents some of the initial results obtained with different combinations of materials.

The equipment was so designed that the bond fracture process could be followed during the tensile test with either an optical or scanning electron microscope. The results could also be stored on videotape for further examination.

The method

The method, named the 3-fibre method, is shown schematically in Fig. 1. The vertical fibre is pulled up from the drop of resin between it and the horizontal fibres whilst the movement of the horizontal fibres in the direction of the pull is prevented. This principle is very simple and can be applied to different fibre/resin bonds. However there are two possible limiting factors, these are:

- that the tensile strength of the vertical fibre is too small (the vertical fibre fractures in pull),
- that the viscosity of the matrix is too small (the resin does not form a drop at the point where the fibres meet).
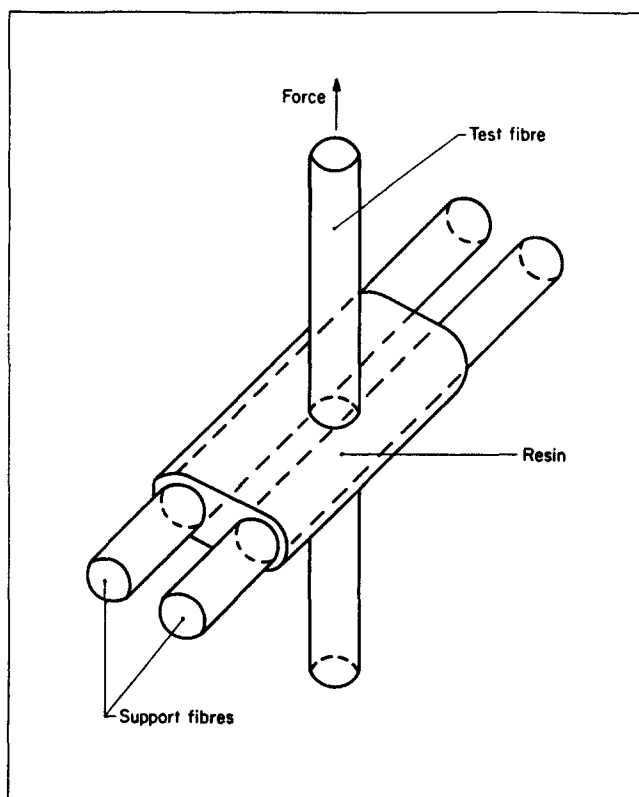

Fig. 1   Schematic representation of the 3-fibre method, used to determine fibre/resin bond strength

The 3-fibre method has the following requirements (shown schematically in Fig. 2):

1) The vertical fibre must have freedom to move vertically, thus allowing the precise placement of the resin drop between the horizontal fibres. The strength of the bond between the vertical fibre and the upper grip of the rig must be greater than tensile strength of the fibre.

2) The horizontal fibres must be positioned so that they are parallel to each other and can be moved independently of the vertical fibre. In addition, the horizontal fibres must be so supported during the tensile test that they cannot move or bend towards the direction of the pull on the vertical fibre.

3) The equipment must allow the bond to be moved to different conditions for polymerization (thermal or UV-polymerization). The bond must be removable from the rig intact, *eg* for microscopic examination.

4) The rig must be so designed that the bond can be tested in a tensile testing machine. This was achieved by dividing the rig into an upper and lower grip. The upper grip was linked so that the vertical fibre was always perpendicular to the plane made by the horizontal fibres, and the applied force was parallel to the vertical fibre. The upper and lower grips also detached without friction.

Experimental

Testing equipment and sample preparation

Fig. 3 shows a cross-section of the testing equipment used and Fig. 4 shows the equipment installed to the tensile testing machine. To speed up the testing time, twenty rigs were used, thus enabling the testing of bonds in series.

The samples were prepared from fibres which had been stored in a dark, dust-free atmosphere. The drop of resin was applied to the vertical fibre and was then moved into the same plane as the horizontal fibres by adjusting the upper grip of the rig. The gap between the horizontal
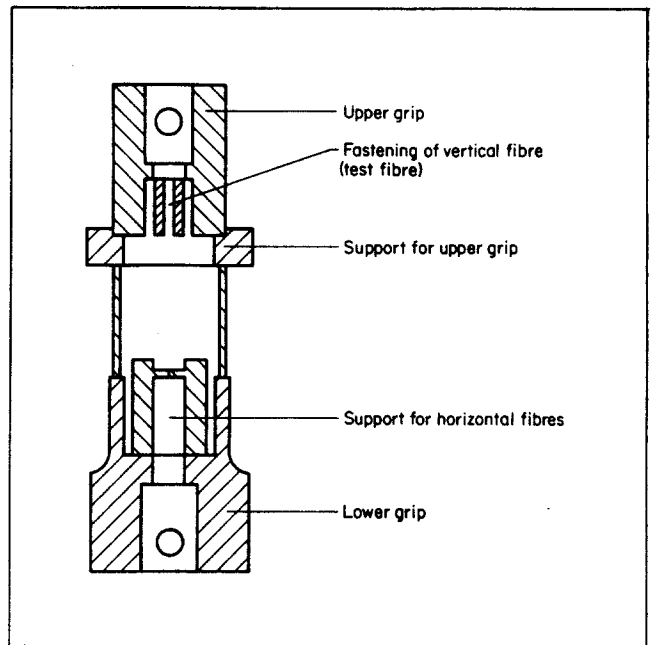

Fig. 3   Cross-section of equipment used for preparation and testing of the 3-fibre bond

fibres was then reduced, thereby bringing the horizontal fibres into contact with the resin drop. It was ensured that the fibres did not touch one another and that the fibres were not touched at the point of the bond during bond preparation.

The 3-fibre samples prepared by the method above were then studied for their load/displacement behaviour on a JJ T5003 or Instron 1115-TT-DM-C tensile testing machine.

The microscopic examination of the bond and its fracture in a tensile test was carried out using an ISI 40 scanning electron microscope (SEM). Extra equipment consisted of anticontamination equipment enabling the scrutiny of uncoated nonconductive samples; a very sensitive ISI Robinson detector (for identifying backscattered electrons); video equipment; and a small tensile testing machine to fit into the goniometer of the microscope, and the specimen chamber, to enable the microscopic tensile testing of the bond.
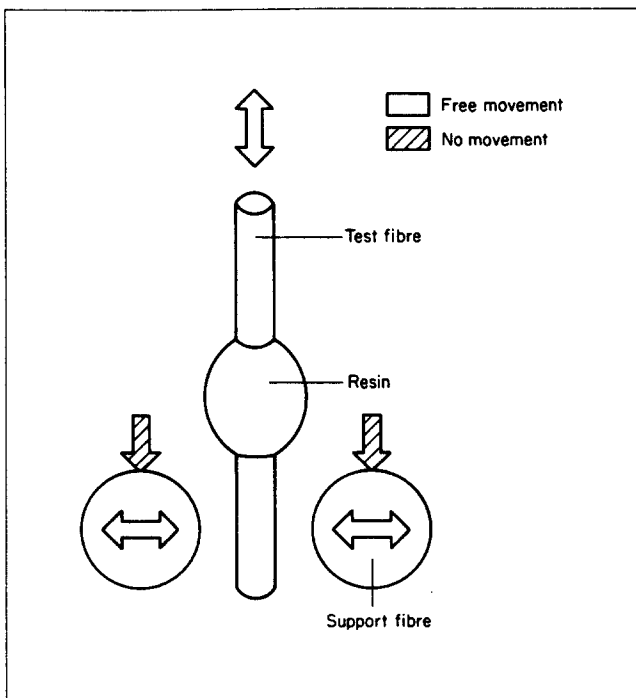

Fig. 2   Basic requirements of 3-fibre method concerning possibilities of movement during preparation and testing of the bond
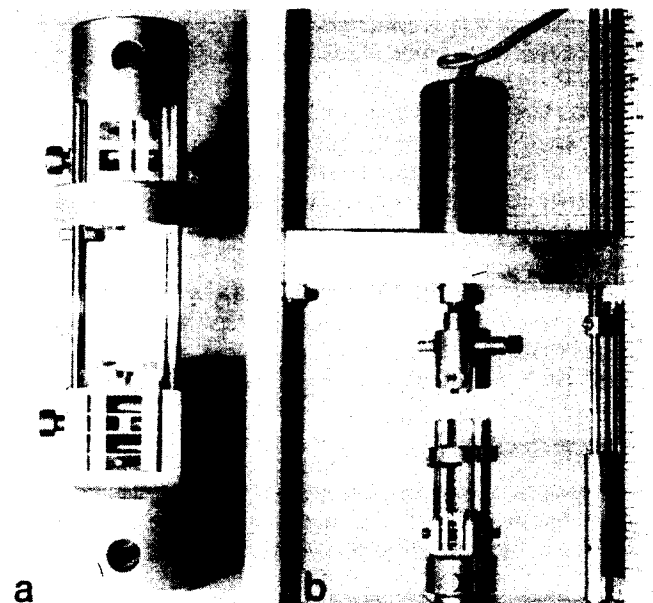

Fig. 4   Photograph of equipment for testing of the 3-fibre bond: (a) the tensile testing rig itself; (b) rig installed in the tensile testing machine (JJ T5003)

The bonds examined by SEM were prepared in the rig shown in Fig. 4. Fig. 5 shows the tensile testing equipment used in the microscope. The broken line in Fig. 5a shows the area which can be followed during the test on the SEM.

Materials

The study concentrated on type A glass fibres. The fibres were manufactured by drawing from liquid glass without any surface treatment. All tests were carried out with horizontal fibres of approximately 160 μm in diameter, the diameter of the vertical fibres was about 80 μm.

The bonding material consisted of different resins, which can be divided into commercial and non-commerical adhesives. Commercial adhesives consisted of some thermoset- and thermoplastic type materials.

Results

The 3-fibre method studies the pull resisting strength of the bond when the vertical fibre is pulled at constant speed. In practice the breaking of the system can occur either by the fracture of the vertical fibre or by the bond breaking.

This test considered the fracture of the vertical fibre undesirable because it indicates that the bond is stronger than the tensile strength of the vertical fibre, which is then the measured value. This occurrence can be partly eliminated by manufacturing stronger fibres from the material to be tested. Because the strength of the fibre/resin boundary

a

b

Fig. 5 Pull-grip system of the tensile testing machine built into the ISI 40 SEM for testing 3-fibre bonds: (a) schematic view of grips; (b) grips fastened to tensile testing machine
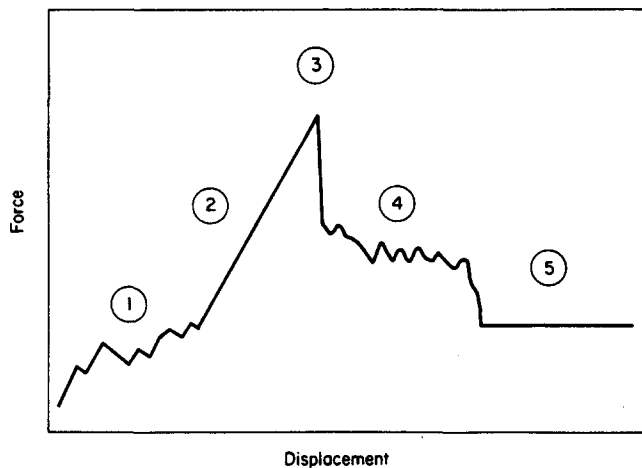

Displacement

Fig. 6 Force acting on the bond during pull as a function of time in testing the 3-fibre bond. (1) the slack disappears from testing equipment; (2) the deformation of the bond for a thermoset plastic; (3) the bond fractures; (4) friction occurs between fibre and resin; (5) the vertical fibre is completely free of the resin

depends, in part, on the shape and composition of the fibre surface, the method used to manufacture the fibre does not have an effect on the test data.

The 3-fibre method does not allow the precise measurement of the bond deformation because the whole grip system goes through a series of small strains in the direction of the pull. Fig. 6 shows schematically the change of force as a function of time in a pull of constant speed with a brittle thermoset type resin. The following stages can be noted in the curve:

1) the bond, in the grips, moves to the correct position and the gaps close,
2) the bond deforms plastically,
3) the bond fractures,
4) friction occurs between the separated vertical fibre and matrix,
5) the vertical fibre is completely free of the bond.

The measuring process involves the evaluation of the shear strength between matrix and fibre. The shear strength (T) is calculated using Equation (1) below, where F_{max} is the largest recorded value of the force resisting the pull, and d and D are defined in Fig. 7.

$$T = F_{max}/\pi dD \qquad (1)$$

This study does not take into account the ability of the resin to wet the vertical fibre. An improvement in wetting would probably increase the strength due to a larger boundary surface. However, as the increase in strength is not necessarily directly proportional to the increase in boundary surface, the effect of wetting is not calculated into the test results.

Several parameters were varied to derive information about the sensitivity scatter and usefulness of the method. The effect of fibre thickness (ie the effect of drop size), the pull-out speed and human factors on the measured strength values are discussed below.

The effect of fibre thickness

If the value measured during the test is really proportional to shear strength, then it must be independent of the area of the boundary surface. Fig. 8 shows the variation of bond strength with different thickness ratios ($a = D/d$) for two different fibre types. The differences in the results were not very large, which suggests that the fibre thickness can be chosen quite freely.
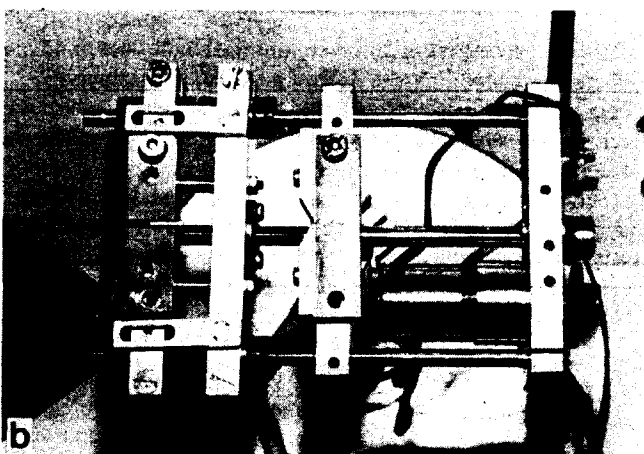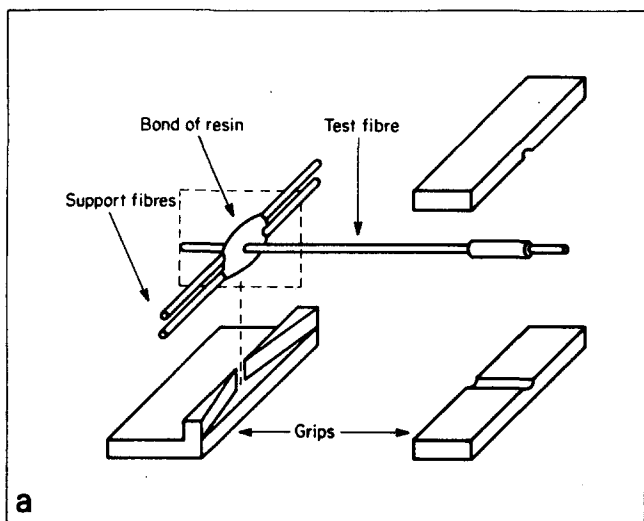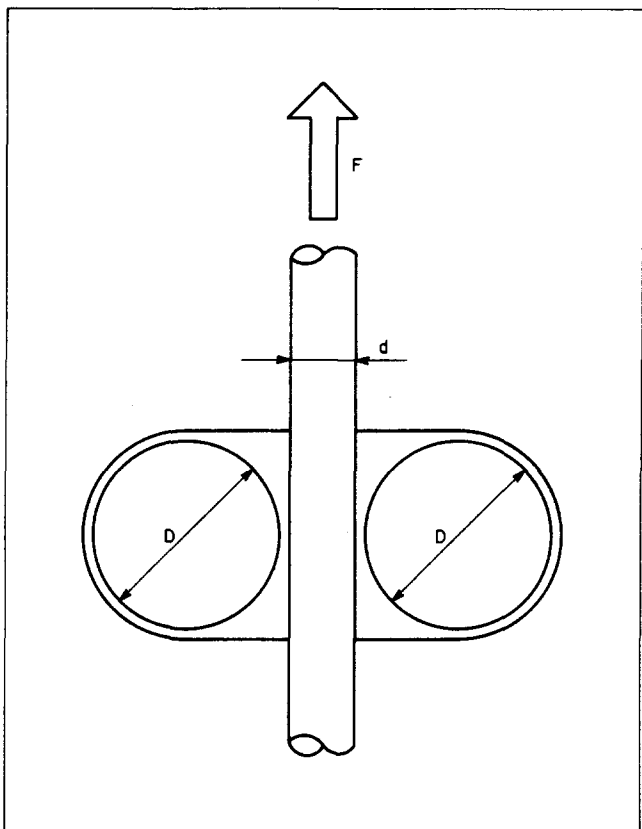
Fig. 7   Definition of values used to measure the shear strength of fibre/resin boundary surface in the 3-fibre method. This method does not account for the wetting between fibre and resin
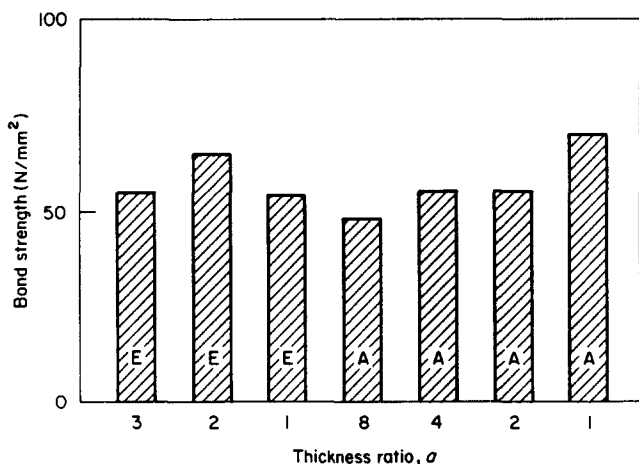

Fig. 8   Effect of thickness ratio on bond strength in 3-fibre method for a Loctite glass adhesive matrix and fibres of E- and A-glass

Therefore, the thickness of the fibres to be subsequently examined was chosen as about 80 µm. Fibres in this size range can be drawn successfully from different glass types and experiment had shown that there were few test failures with this fibre diameter.

The effect of pull-out speed

The speed of the pull might be expected to have some effect on the results, mainly because with greater speeds the bond does not have time to find its correct position (machined metal surfaces are not smooth compared with a glass fibre). On the other hand, the pull-out speed may affect the fracture mechanism of the viscoelastic resin.

The effect of the pull-out speed was examined by using urea-formaldehyde resin as the matrix and pull-out speeds of 0.05–1.0 mm/min. The results are presented in Fig. 9.
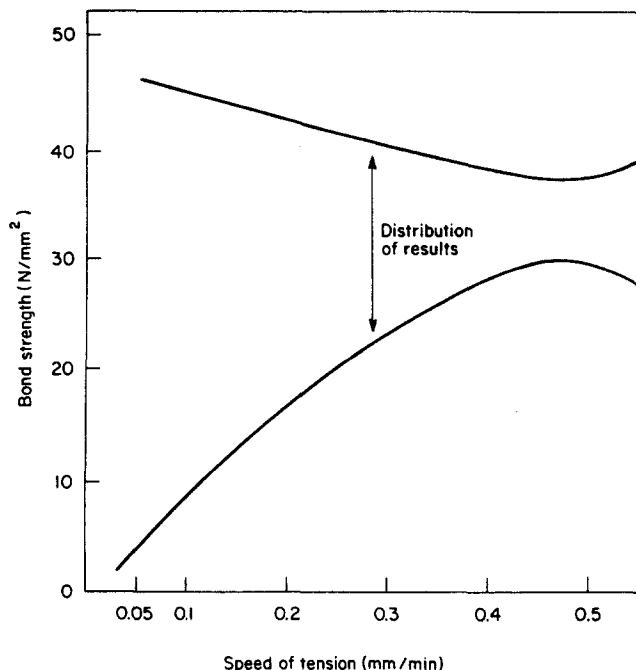

Fig. 9   Effect of pull-out speed (speed of tension) on the bond strength of thermoset type urea-formaldehyde resin with a vertical fibre of A-glass

The results show that the maximum shear strength is almost independent of pull-out speed (or speed of tension, see top line in Fig. 9). The smallest scatter in results was obtained with a pull-out speed of 0.5 mm/min, and scatter increased when pull-out speed was varied in either direction. At speeds greater than 0.5 mm/min, breaking of the vertical fibre occurred more frequently, which limits the method. This may be explained if there are also bending and shear stresses in the vertical fibre as well as tensile stresses. The large scatter in the results with low pull-out speeds is probably affected by small defects in the bond, where the relatively long time between the incremental pulls allows fracture propagation to occur.

The effect of human factors

In tests where the main part of the work is exact handwork, different persons can very often obtain distinctively different results. Fig. 10 shows the test results of three different persons using three different resins. It can be seen that the maximum values and scatter are approximately the same.

Variation of bond strength with resin type

The bond strengths of some commercial adhesives and glass fibres determined by the 3-fibre method, are shown in Fig. 11. The adhesives evaluated were some glass adhesives, general glues and special adhesives. Details of the adhesives used are given in Table 1.

The bond strengths of the various adhesive types vary quite widely; for example, the actual glass adhesives had very large differences in shear strengths and also in fracture mechanisms. These differences are hard to explain in detail because the exact composition of the adhesives is unknown.

The effect of polymerization temperature and time

The practicality of the 3-fibre method in the optimization of polymerization time and temperature for a thermoset resin was demonstrated by measuring the development of the urea-formaldehyde resin/glass fibre bond strength as a function of these parameters.
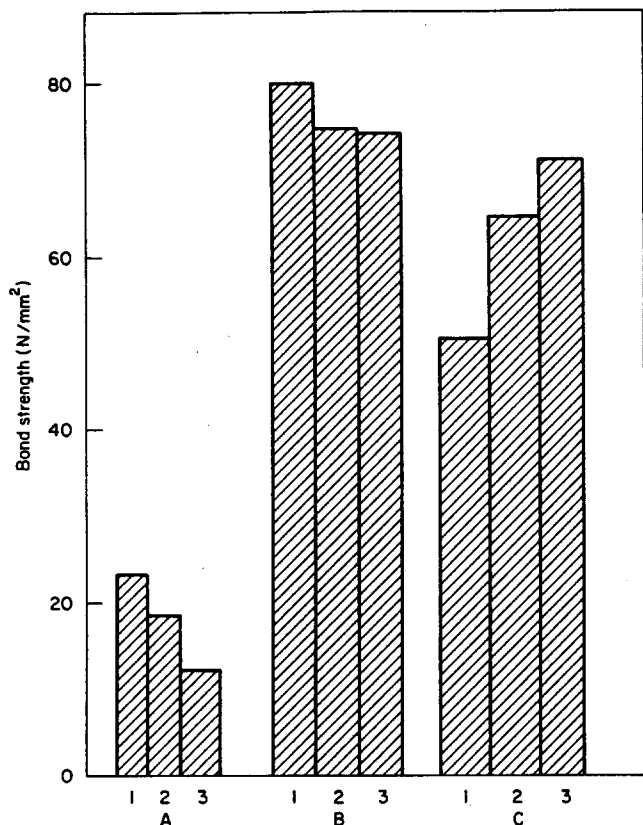
Fig. 10   Effect of human factors on bond strength obtained by the 3-fibre method. Results of three different persons with the same materials and conditions (A, B and C different resins, 1, 2 and 3, different persons)

Table 1.   Commercial adhesives and their polymerization conditions

Adhesive*	Polymerization conditions†
Scotch 3M glass adhesive 3 M/France	24 h at RT
Tammer F3/phenol formaldehyde resin/Sarvis Oy/Finland	24 h at RT
Super adhesive/cyanoacrylate/Plastic Padding/Sweden	24 h at RT
Glass adhesive/Loctite	1 h in UV-radiation
Super Epoxy/epoxy resin/Plastic Padding/Sweden	24 h at RT
Multiband/Loctite	24 h at RT

*Name, resin type, manufacturer, country
†RT, room temperature. All resins post-cured for 24 h at 50–60°C

The results are given in Fig. 12, and they clearly show that the bond strength depends on the polymerization conditions. In principle, these results were to be expected, and they are a good indication of the dependability of the method. The temperatures shown in Fig. 12 refer to the temperature of the atmosphere surrounding the sample; the exact measurement of the temperature of the bond was not possible.

Discussion

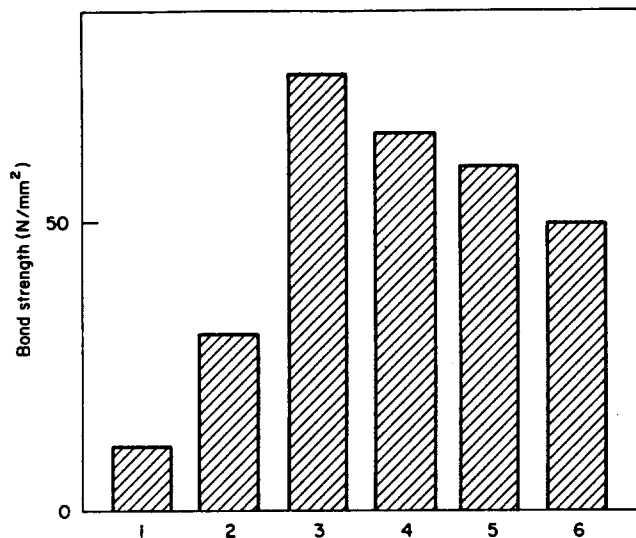The aim of this study was to determine the absolute bond

Fig. 11   Bond strengths of commercial resins in the 3-fibre test with A-glass fibre: (1) Loctite Multibond; (2) Scotch 3M glass adhesive; (3) Loctite glass adhesive (UV-cured); (4) Tammer F3 (phenol-formaldehyde resin); (5) Plastic Padding Super Adhesive; (6) Super Epoxy

strengths of various fibre/resin combinations. This paper has concentrated on examining the practicality of the 3-fibre method in determining bond strength.

Another aim of this study was to compare the absolute values of the bonds strengths with results achieved with other methods. The values received with this method are generally larger than results received in other single bond studies; indeed when compared to values obtain from real composites, the results of this study are notably larger.

The biggest cause of uncertainty responsible for the scatter in the results was the incomplete control of dropsize and the
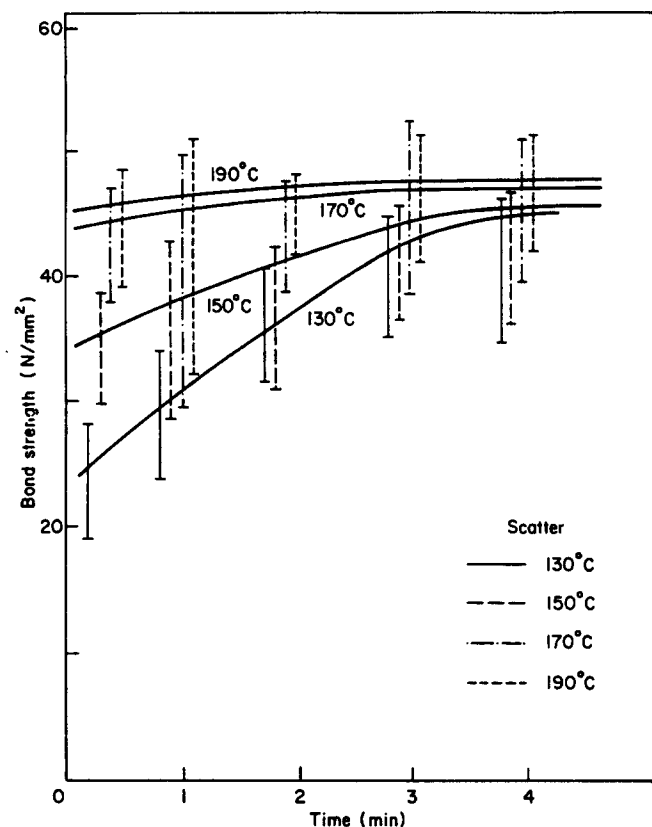

Fig. 12   Effect of polymerization time and temperature on the bond strength in the case of phenol-formaldehyde resin with a vertical fibre of A-glass
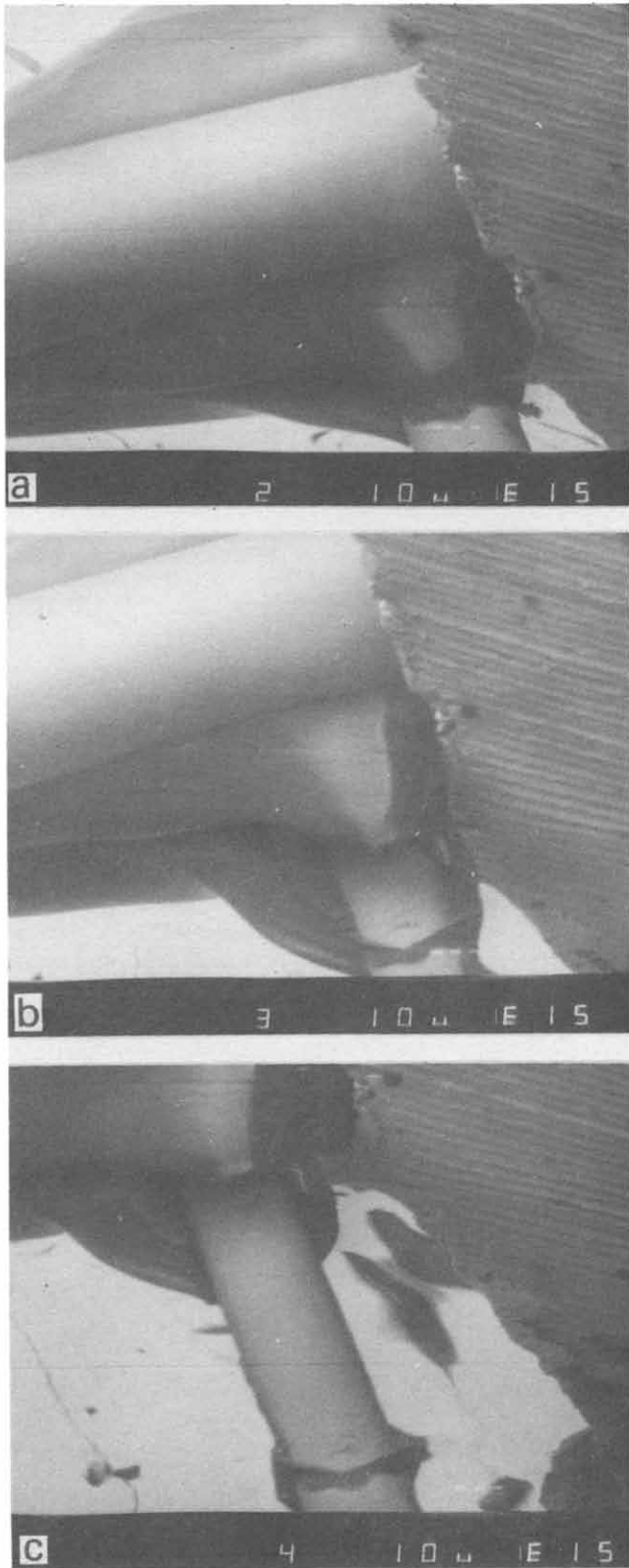
Fig. 13   Scanning electron microscope pictures of a bond in different stages of testing: (a) bond before pull; (b) bond during pull (first fractures seen in resin); (c) bond completely fractured (vertical fibre totally free)

completely prevent the making of a drop. However, due to the small drop size and the fibre thickness used in this study, it was possible to make a drop from a thermoset type polycondensate resin without the formation of porosity.

It must be noted that the actual polymerization state of the drop cannot be measured. The thermal history of cure hardening resins, for example, cannot be defined due to the small dropsize. With some resins, there is the additional problem of evaporation of components from the resin during sample preparation. This effect becomes noteworthy when the evaporating components take part in the polymerization process.

The tensile test in the scanning electron microscope

The pull-out device and the grips developed in this study for testing bonds in the SEM were discussed above.

Fig. 13 shows a bond tested by this method in the microscope at various stages during the pull. Since this method allows the process to be recorded on videotape for later examination, a more accurate analysis of the fracture nucleation and development can be undertaken. Fig. 13a shows an intact bond before the beginning of a pull-out test, showing wetting of the fibres and the shape of the drop. Fig. 13b shows the bond just before fracture; a nucleation is formed which leaves a collar on the vertical fibre. The stage at which the fibre is completely released from the bond can be seen in Fig. 13c. Pieces of resin attached to vertical fibre, which remains between the horizontal fibres can clearly be seen. These pieces of resin are an indication of the bond strength between the fibre and resin.

A more detailed study of bond nucleation and fracture development will be published at a later date.

Conclusions

The aim of this study has been the development of a practical method for determining the absolute strengths of different fibre/resin bonds and their fracture mechanisms. The method for measuring bond strengths must be such that it enables the effect of the fibre coating, the adhesive composition and the changes in the polymerization process on the bond strength and fracture point of the bond to be determined.

This paper describes the development of the so-called 3-fibre method for determining glass fibre to resin bond strength, the sample preparation and testing equipment. The method was used to test bonds between several different glass types and many commercial and non-commercial adhesives. The results show that the recorded strength values are related to the actual strength of bond.

The results show clear correlations between different matrices, adhesion promoters and polymerization processes. All the results obtained agree with the bond strength values obtained by existing methods. The bond strength values found by this method are, however, higher than the results from other methods.

It was also noted that the results are independent of the fibre thickness in the range 20–100 μm. Thinner fibres were not used because the fibre production method employed was not capable of producing them.

Preliminary results of tensile tests carried out in a scanning electron microscope are presented towards the end of the article.

polymerization process. The standardization of the drop size was achieved manually, for a suitable method for making a standard drop with an auxiliary device was not found. The weight distribution of manually made drops in this study was ± 15%, which must be considered a good value. The small viscosity and wetting ability of some adhesives can

References

1 **Thomas, W.F.** 'The effect of fibre diameter on the strength and stiffness of glass fibre reinforced polymeric resins' *Fibre Reinforced Materials: Design and Engineering Applications* (Institute of Civil Engineers, London, 1977) pp 57–62

2 **Irion, M.N. and Adams, D.F.** 'Compression creep testing of unidirectional composite materials' *Composites* 12 No 2 (April 1981) pp 117–123

3 **Landsman, P.U. and Klunder, J.** 'Eine Prüfmethode zur Bestimmung der Glas/Harz-B ndung in Glasfaser — Kunststoffen' *Kunststoffe* 54 No 12 (1964) pp 791–794

4 **Yamaguchi, Y. and Amano, S.** 'Effect of coupling agent on the adhesive strength between glass plate and resin and on the tensile strength of F.R.P.' *SPE, Ann Tech Conf, San Francisco, CA, USA, May 1974* Paper 32 pp 452–455

5 **Laws, V.** 'Micromechanical aspects of the fibre-cement bond' *Composites* 13 No 2 (April 1982) pp 145–151

6 **De Vekey, R.C. and Majumdar, A.J.** 'Determining bond strength in fibre-reinforced composites' *Magazine of Concrete Research* 20 No 65 (December 1968) pp 229–234

7 **Gerbet, D.** 'Beitrag zur Prüfmethodik von Plastklebverbindungen' *Plaste und Kautschuk* 17 No 10 (1970) pp 753–759

8 **Andreevska, G.D. and Gorbatkina, Y.A.** 'Adhesion of polymeric binders to glass fibres' *25th Ann Tech Conf, 1970* (Reinforced Plastics/Composites Division of The Society of the Plastics Industry Inc, Section 16-F) pp 1–6

9 **Favre, J.P. and Merienne, M.-C.** 'Characterization of fibre/resin bonding in composites using a pull-out test' *Int J Adhesion and Adhesives* 1 No 6 (October 1981) pp 311–316

Authors

P. Järvelä and P. Törmälä are with the Institute of Materials Science and the Institute of Textile Technology, respectively at Tampere University of Technology, PO Box 527, SF-33101 Tampere 10, Finland. K.W. Laitinen and J. Purola are both at A. Ahlström Osakeyhtiö, Mineralfibre Industry, Research and Development, PO Box 18, SF-48601 Karhula, Finland. Inquiries about the work described in this paper should be directed initially to P. Järvelä.