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Measurement of the stress field created within the resin between fibres in a composite material during cooling from the cure temperature

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In a long uniaxial glass fibre/epoxy resin composite that is constrained by a rigid outer shell not to change its overall external dimensions, the transverse components of principal stress generated during cooling from the temperature of cure have been measured at various distances from the specimen ends. The radial (compressive) and hoop (tensile) components of stress in the resin between four fibres whose axes define the edge of a prism with square section are in excess of 6 and 100 MPa, respectively. In an identical specimen, 4 days of exposure to distilled water at 80° C was found to give rise to a tensile hoop stress of magnitude sufficient to cause fracture of the rigid outer shell.

1. Introduction

The resins used as matrix materials in composite materials are usually cured at temperatures close to their glass transition temperatures. Consequently it is generally believed that the resin shrinkage which accompanies curing is completely relieved by viscoelastic flow of the resin itself. Differential thermal contraction between fibre and matrix materials during cooling from the cure temperature is not accommodated by resin flow and the state of self-stress attributable to this source has been calculated for simple fibre geometries by Adams and co-workers [1, 2]. In glass fibre/epoxy resin systems, it is predicted that the radial principal stress is tensile along the line of centres between adjacent fibres and compressive elsewhere, and that its magnitude at the fibre/resin interface is in the range 100 to − 80 MPa.

The aim of the present research is to measure two of the principal components of the self-stress field of a glass fibre/epoxy resin composite in the as-cured state.

2. Method

In order to reduce the problem to one of two-dimensional elasticity, thin transverse sections were cut from a long unidirectional composite. Accommodation of shrinkage by way of rigid body displacements in the transverse plane were prevented by casting the composite inside a thick walled glass tube. The unrelaxed radial and hoop principal stresses within any slice subsequently removed from the specimen were then amenable to measurement by the oblique incident photoelastic method described by Durelli and Riley [3]. The two-dimensional stress-optic law for a beam of light that enters the surface of a thin transverse section at normal incidence is

$$R = \frac{Ct}{\lambda}(\sigma_1 - \sigma_2) \qquad (1)$$

where σ_1 and σ_2 are the principal stresses in the transverse plane, λ is the wavelength of the light, C is the stress-optical coefficient and t is the thickness of the transverse section. In terms of the fringe order n, Equation 1 becomes

$$n = \frac{Ct}{\lambda}(\sigma_1 - \sigma_2) = \frac{2Ct}{\lambda}\tau_{max}. \qquad (2)$$

τ_{max} = maximum shear stress in the transverse plane. Equation 2 can also be written as

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$$\tau_{max} = n\frac{\lambda}{2Ct} = nF \quad (3)$$

where

$$F = \frac{\lambda}{2Ct} \quad (4)$$

is the model stress fringe value. The model stress fringe value is the change in maximum shear stress necessary to produce a change of unity in the fringe order. The quantity

$$Ft = \frac{\lambda}{2C} = f \quad (5)$$

is independent of the thickness of the model and is called the material stress fringe value. f is the change in maximum shear stress required to produce a change of unity in fringe order in a model of unit thickness. Substituting Equation 5 into Equation 2,

$$n = \frac{\tau_{max}t}{f} = \frac{(\sigma_1 - \sigma_2)t}{2f}.$$

Let the specimen be rotated about principal axis 1, through an angle θ. The path length (t_0) of the light becomes

$$t_0 = \frac{t}{\cos\theta} \quad (6)$$

and the principal stresses transform to

$$\sigma_1; \quad \sigma_2 \cos^2\theta. \quad (7)$$

Let

$$a_1 = \frac{\sigma_1 t}{2f}; \quad a_2 = \frac{\sigma_2 t}{2f}.$$

The corresponding fringe orders identified by $n_0^{(1)}$ are

$$n_0^{(1)} = \frac{(\sigma_1 - \sigma_2\cos^2\theta)t}{2f\cos\theta} = \frac{(a_1 - a_2\cos^2\theta)}{\cos\theta} \quad (8)$$

$$a_1 = \frac{\cos\theta(n_0^{(1)} - n\cos\theta)}{\sin^2\theta}$$

$$a_2 = \frac{n_0^{(1)}\cos\theta - n}{\sin^2\theta}.$$

Therefore

$$\sigma_1 = \frac{\cos\theta(n_0^{(1)} - n\cos\theta)2f}{t\sin^2\theta}$$

$$\sigma_2 = \frac{(n_0^{(1)}\cos\theta - n)2f}{t\sin^2\theta}. \quad (9)$$

3. Specimen preparation and examination

Model epoxy resin unidirectional composites containing 0.5 mm diameter ZrO_2 glass filaments have been cast inside glass tubes whose dimensions are 5 mm o.d. and 3 mm i.d. Prior to casting, all the glass components were thoroughly cleaned using the following procedure: (1) Ion-bombardment; (2) De-oxidation by exposing for 10 to 15 min to a mixture consisting of 170 ml HNO_3: 30 ml HF: 800 ml distilled water; (3) Tap-water spraying, 10 to 15 min; (4) Deionised water spraying, 2 min; (5) Drying in hot air, minimum of 30 min and (6) Cooling to room temperature, maximum of 120 min. Small bundles of fibres, usually about 10 or 12, were gathered together and inserted into the glass tube. In the early stages of the investigation no attention was paid to the packing arrangement, the fibres usually adopting an irregular array. Epoxy resin (Araldite MY753 manufactured by Ciba–Geigy (UK) Ltd) was then driven into the specimen until all the air, including all visible bubbles, had been displaced. The resin was gelled at room temperature for 2 days and cured at 60° C for 6 h in accordance with the manufacturer's recommendations.

1 mm thick transverse sections were removed from the specimen at intervals along its length using a high speed diamond impregnated annular saw. In order to render the sections optically transparent and avoid corrections for refraction when examined by oblique incidence, it was necessary to coat the as-cut surfaces lightly with immersion oil.

Fig. 1 is a photograph of one such section examined with a Nikon Apophot 180-00-103 polarizing microscope. Fig. 2 is an enlargement of a small region taken from another section of the same specimen and shows the complexity of the stress birefringence created during cooling from the cure temperature.

The range of stress birefringence resolved in these figures is large, there being several fringe orders present. In an attempt to reduce the complexity of the stress pattern and to render it amenable to comparisons with theoretical investigations, specimens with parallel fibres arranged in a periodic array were prepared. Individual 5 cm fibre lengths were carefully laid up outside of the tube in a rectangular array and their ends locked into position with quick-setting epoxy resin. The fibre bundle so obtained was then cleaned in the normal way and inserted into the glass tube with the ends outside of the tube. Epoxy resin was then driven into the tube and allowed to gel, the fibre ends were cut off and the tube cut into two equal

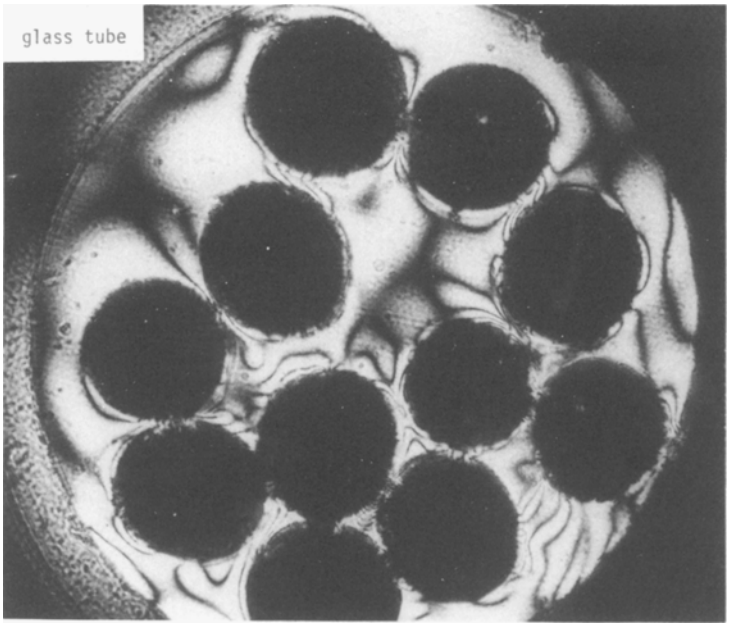


Figure 1 Photograph of a transverse section of a model unidirectional composite examined with the polarizing microscope.

lengths which were then placed in an oven to effect curing. Using this technique it was possible to obtain two nearly identical specimens, one of which could be retained as a control specimen.

4. Measurements

Fig. 3 shows photographs of 1 mm thick transverse sections cut from various positions along a 2 cm length of specimen. The fibres were arranged on a rectangular grid and photographs were taken using both plane and circularly polarized light. Although stress differences revealed by the isochromatics vary non-linearly along the length of the specimen, the general distribution of the in-plane stresses near the fibre is consistent with finite element method (fem) calculations. It is interesting to note that large stress gradients surround some fibres and fringe orders of at least 5 can be counted.

Fig. 4 shows a photograph of a 1 mm thick transverse section cut from the middle of a specimen which exhibited both well defined fringes and a large retardation. The image was taken with

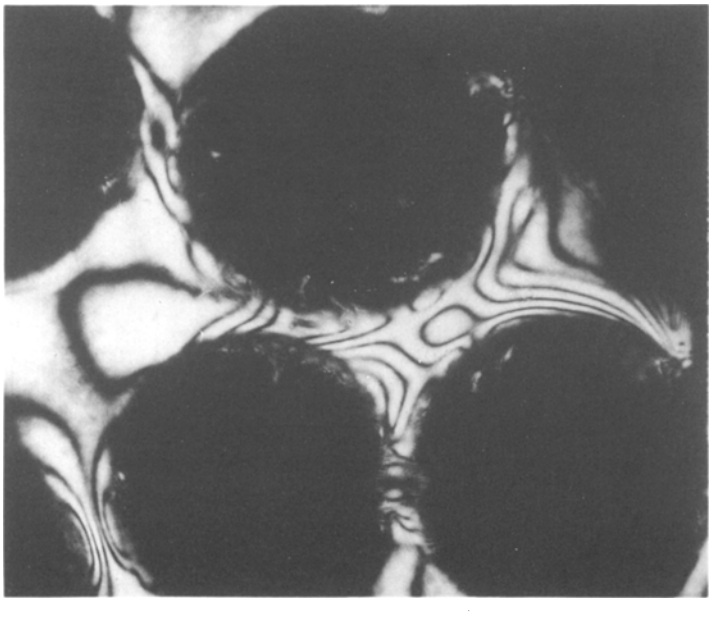


Figure 2 Photograph of a small region of a transverse section examined with polarizer and analyser crossed.

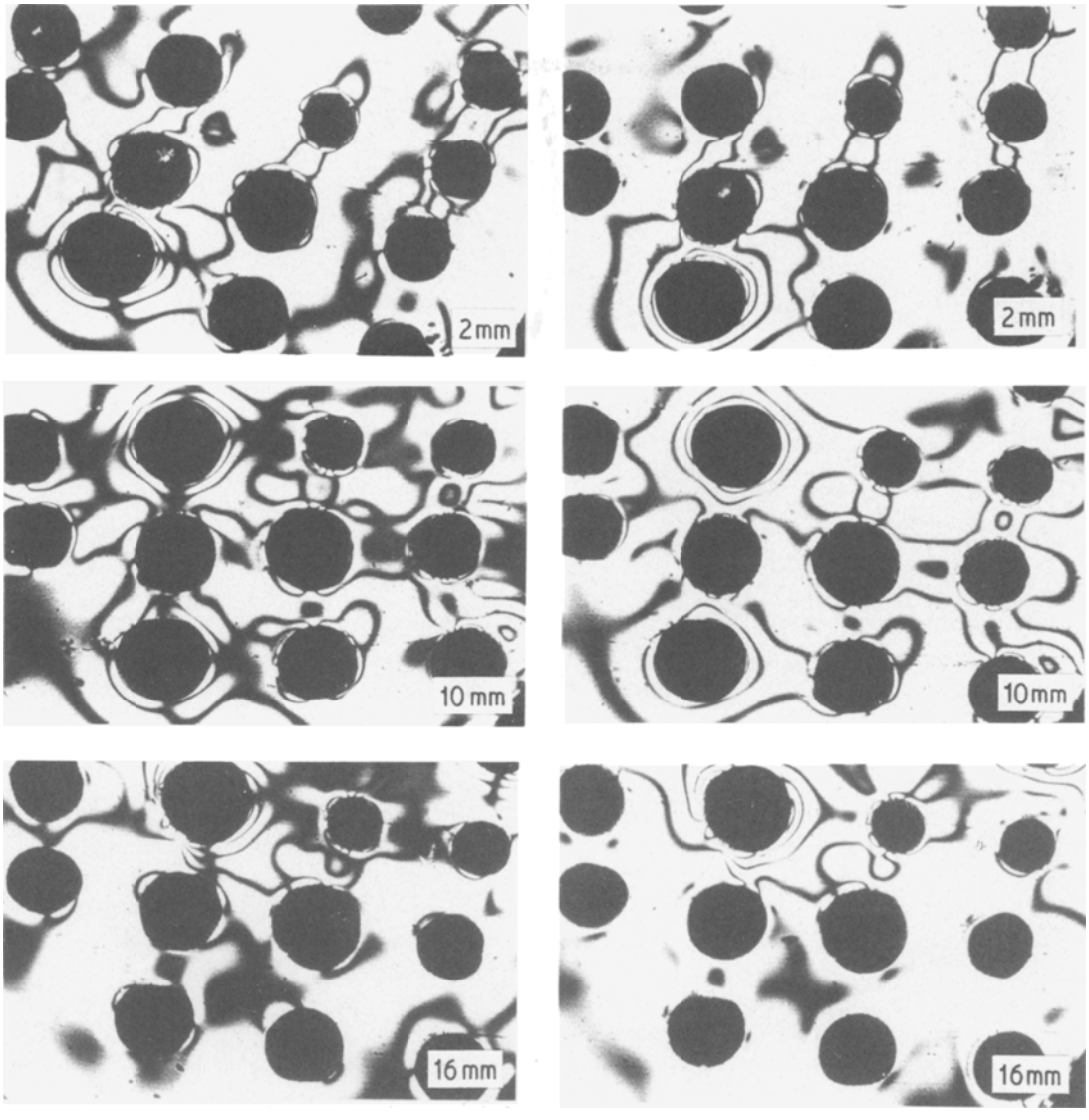


Figure 3 Photographs of transverse sections examined in both plane and circularly polarized light. The fibres are arranged in a periodic array. Left hand side are isoclinics plus isochromatics and the right hand side are isochromatics.

plane polarized light incident normally upon the specimen. Fig. 5 shows a plot of the hoop stress (σ_2) against distance from the fibre for one of the fibres seen in Fig. 4. The units of stress are λ/Ct. With $\lambda = 546\,\text{nm}$, $t = 1\,\text{mm}$, and using $C = 56.6 \times 10^{-12}\,\text{m}^2\,\text{N}^{-1}$, λ/Ct corresponds to a stress of $9.65\,\text{MN}\,\text{m}^{-2}$. The hoop principal stress at the fibre/resin interface is therefore of the order of 100 MPa falling to approximately 20 MPa at a distance of one diameter from the fibre. The radial principal stress is compressive and is approximately constant (6 MPa) with distance from the fibre.

A specimen having a similar geometry to that reported in Fig. 3 was immersed in distilled water at 80° C for 4 days. On removal from the water, the glass tube in which the specimen was cast was observed to have cracked at each end, as shown schematically in Fig. 6. Large stresses generated during resin swelling were responsible for the cracking. The specimen was sectioned in the usual manner and examined under the polarizing microscope. The stress birefringence patterns are shown in Fig. 7. Very little stress birefringence was detected at any section along the length between the cracked ends.

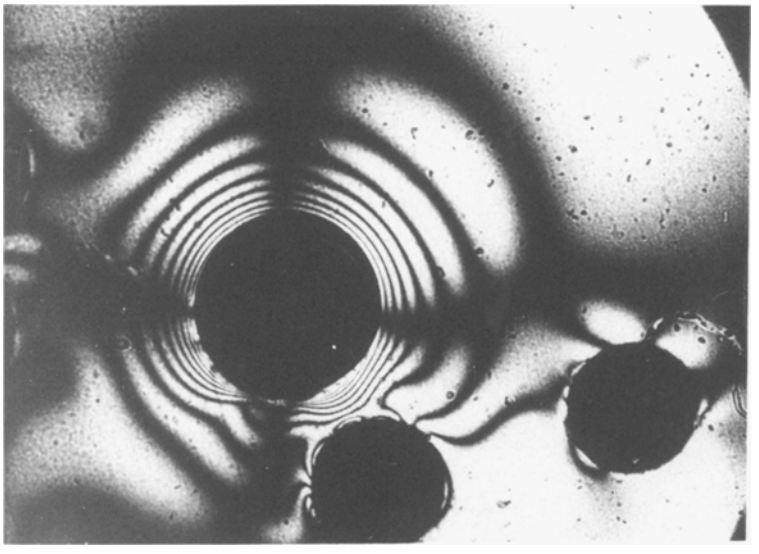


Figure 4 Photograph of a small region of a transverse section examined with polarizer and analyser crossed.

5. Discussion

Whilst the general form of the birefringence patterns indicated a relatively complex stress distribution, certain fibres exhibited very little asymmetry in the stress field. The birefringence patterns observed in Fig. 4 show a symmetrical stress field with clearly defined radial and hoop principal stresses. The hoop stress ($\sigma_{\theta\theta} = \sigma_1$) is very much larger than the radial stress ($\sigma_{rr} = \sigma_2$). The fact that σ_{rr} does not vary significantly with r means that the maximum shear stress, $(\sigma_{\theta\theta} - \sigma_{rr})/2$, rapidly increases near the resin/fibre interface, i.e. that molecular orientation, if it occurs, is expected to be localized adjacent to the fibre.

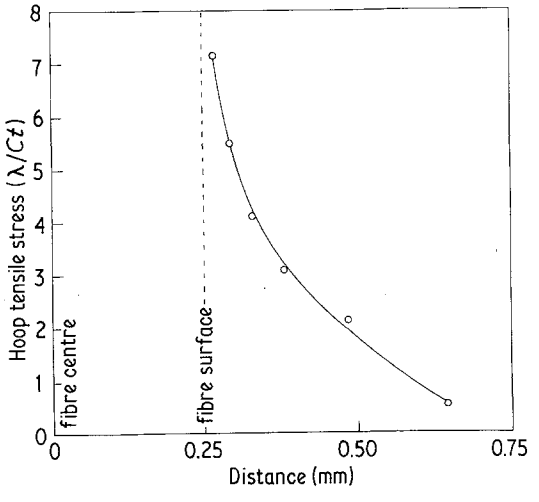


Figure 5 Plot of the hoop tensile stress against radial distance from the fibre.

The largest measured shear stress, 47 MPa, was at a distance of some 25 μm from the fibre/resin interface. Interpolation of the data to smaller distances suggests that the 20° C yield strength of dry resin (~ 100 MPa) would be exceeded at about 15 μm from the interface.

The resin swelling that accommodates water uptake in composite materials will depend on the concentration of diffused water and is expected to be larger near to regions of water access than when it is remote from such regions. If otherwise unconstrained, the fillet of resin between three closely packed fibres would endeavour to undergo the shape change sketched in Fig. 8b. Eventually, the swelling in resin in contact with the source of water molecules would saturate and

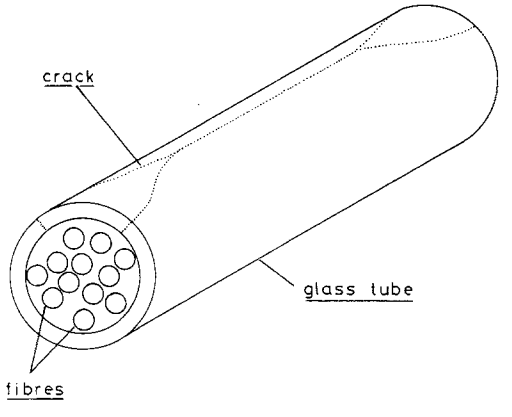
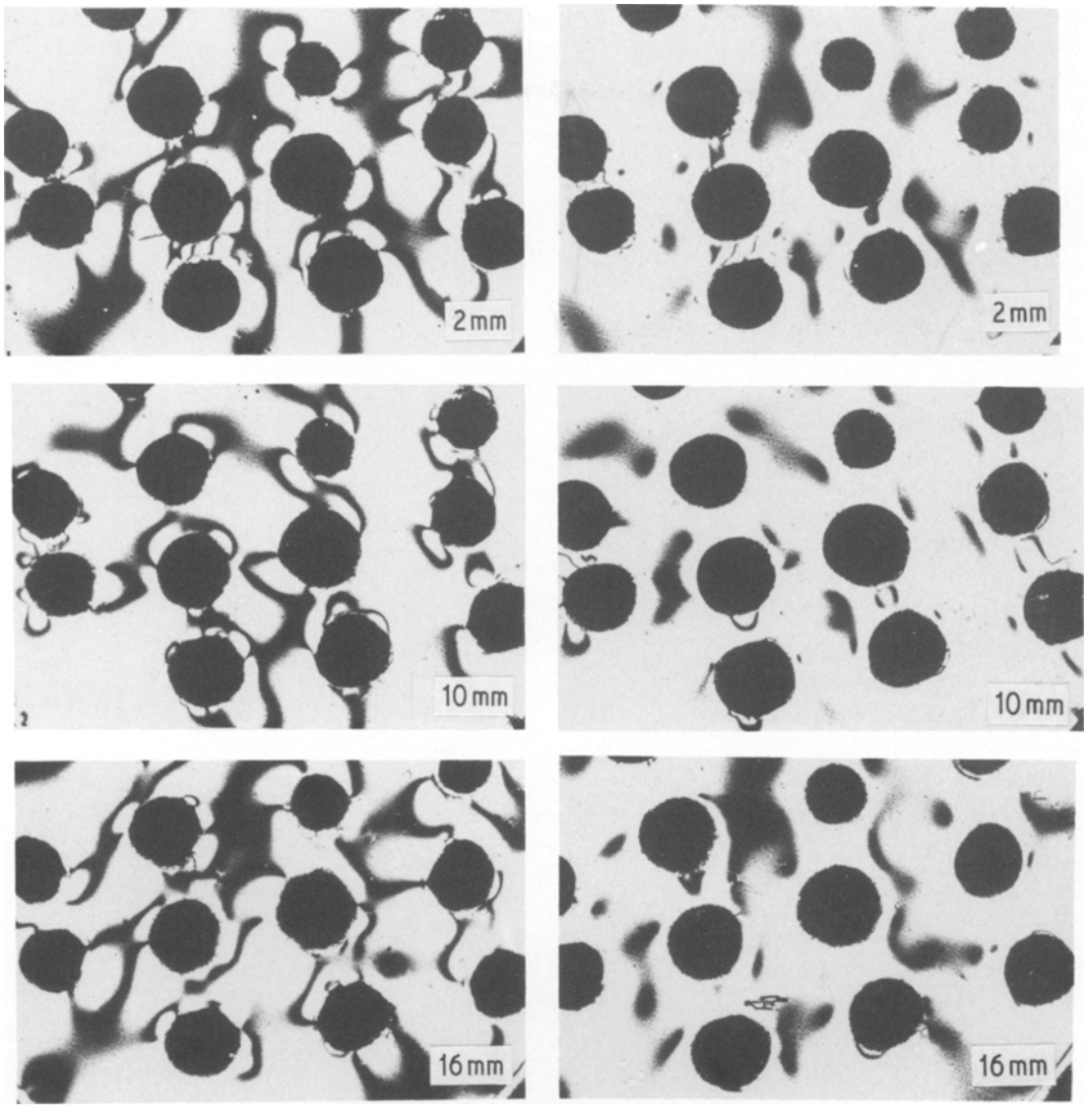


Figure 6 Schematic diagram showing the cracking of the glass tube in which the specimen was cast, after immersion in water at 80° C for 4 days.



Isoclinics and isochromatics

Isochromatics

Figure 7 Photograph of transverse sections examined in plane and circularly polarized light, after immersion in water at 80° C for 4 days. The fibres are arranged in a periodic array. Left hand side are isoclinics plus isochromatics and the right hand side are isochromatics.

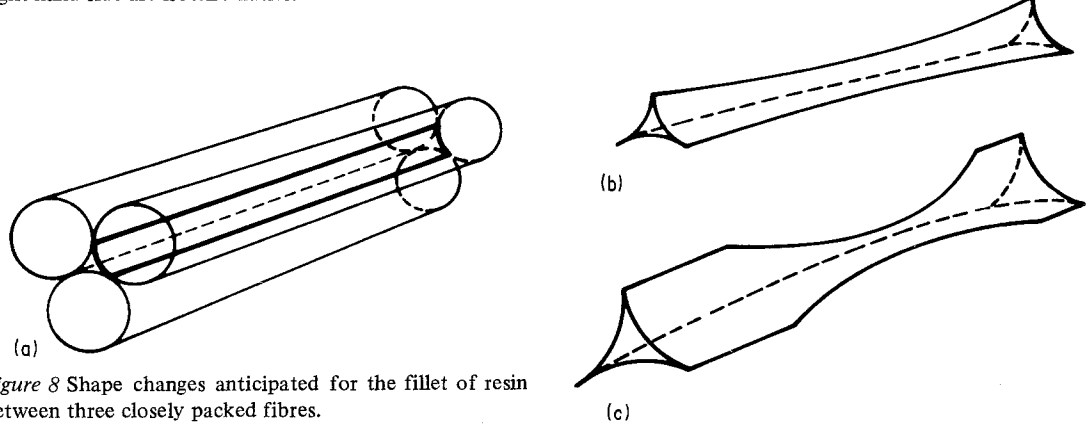


Figure 8 Shape changes anticipated for the fillet of resin between three closely packed fibres.

the regions of maximum swelling will move inwards as indicated in Fig. 8c. In the specimens studied here, the resin was not free to adopt these shape changes. The mechanical constraint offered by the glass tube surrounding the composite specimen caused the resin swelling to generate a pressure of magnitude sufficient to burst the glass tube.

Acknowledgements

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