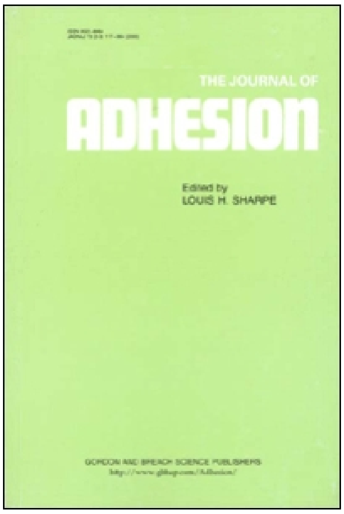

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High Resolution Optical Interference Investigation of Swelling Due to Water Uptake by Model Adhesive Joints

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Fully cured resin adhesives swell when they absorb water. By manufacturing a model butt joint between a thin, and therefore flexible, microscope cover slip as one adherend and a massive, and therefore rigid, slab of glass as the other adherend, development of the swelling can be observed and measured by generating Moiré images from photographs of the pattern of optical interference fringes formed in the gap between an optical flat and the free surface of the cover slip.

The swelling is strongly inhomogeneous and this inhomogeneity gives rise to a distribution of stress normal to the joint which is compressive near both the rim and centre, and tensile within an annular region located between the rim and centre. It has been demonstrated that swelling stresses large enough to cause fracture of the cover slip can be developed. Regularly spaced radial perturbations in the boundary between unswollen and swollen adhesive have been observed in a film which contains a carrier cloth.

INTRODUCTION

Water uptake by epoxy resin based materials is accommodated by swelling. For all cases, other than that which corresponds to a uniform distribution of diffused water, the swelling is inhomogeneous and causes the material to be self-stressed. In geometries such as that provided by an adhesive joint, where most of the resin is shielded from direct access to the outside environment, the swelling, and hence the stress field, is strongly inhomogeneous. The purposes of this research are to demonstrate the nature of this swelling inhomogeneity and to measure its magnitude for joints manufactured using a range of commercial adhesives.

EXPERIMENTAL

The swelling inhomogeneity can be conveniently observed and monitored by manufacturing a specimen (shown schematically in Figure 1) consisting of a thoroughly cleaned glass cover slip bonded to a thoroughly cleaned rigid block of glass, mounted so that the free surface of the cover slip is in close proximity to an optical flat. With monochromatic light, a complex pattern of interference fringes (Fizeau fringes) is formed in the space between cover slip and optical flat. These fringes precisely reflect the topology of the thin cover slip and therefore of the underlying resin; they are the contours of the glass cover slip, the contour level with respect to the reference flat differing by $\lambda/2$ from one dark or bright fringe to the next. Should the shape of the cover slip change, as happens when the resin swells during water uptake, the pattern of interference fringes changes. Whilst direct observation of this

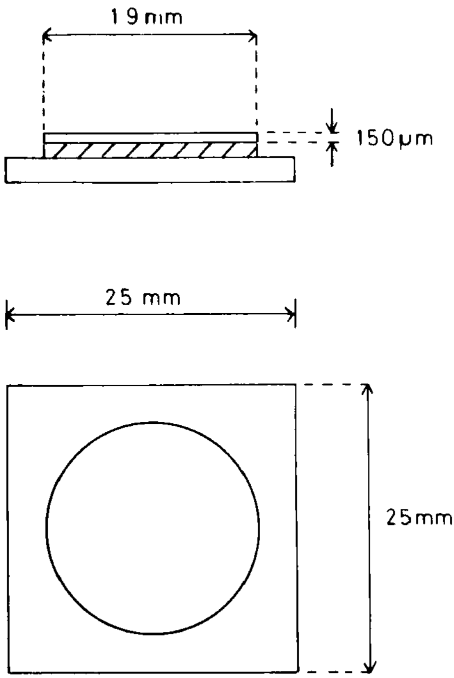


FIGURE 1 Schematic diagram showing the glass test specimen.

interference pattern can be used to measure increments of swelling, the swelling geometry can be obtained directly by superimposing on the pattern photographed before swelling, successive images of the pattern photographed during swelling. Superimposition produces Moiré fringes, the development of which faithfully follows any changes in shape of the cover slip.

Moiré fringes are generated when patterns with nearly identical periodic structures are made to overlap. In this instance, photographic images of the interference pattern form the periodic structures and differences in the pattern, seen on comparing the image photographed before swelling with images taken during swelling, give rise to the Moiré effect. In Figure 2, the Moiré fringes are the circumferential lines running around the rim of the specimen and are superimposed upon the interference pattern. (It should be noted that the broad, dark, almost parallel, bands running across the specimen are due to tilting with respect to the optical flat). As water diffuses into the specimen, new interference fringes are generated at the edge and migrate towards the centre, each fringe representing the locus of points of constant displacement normal to the joint. Similarly in the Moiré pattern, a new Moiré fringe is generated and migrates in from the edge of the specimen whenever a new interference fringe is generated. Thus successive Moiré fringes map out of the loci of positions which differ in displacement normal to the joint by half a wavelength. Hence the number of Moiré fringes N to have passed a given point is simply related to the normal displacement of the joint w by:

$$w = \frac{N\lambda}{2\mu}$$

where λ = wavelength of light

μ = refractive index of the immersion liquid

Specimens usually took the form of a circular glass coverslip bonded to a rigid block of glass. However, some specimens were manufactured using cover slips which were both square in outline and of different thickness. This was done in order to investigate the effects of varying the mechanical constraint attributable to cover slip geometry. The optical arrangement is shown in Figure 3. Light of wavelength 546.1 nm from a mercury vapour discharge lamp passes through an interference filter, (the interference filter has a half-bandwidth of approximately 7 nm which effectively suppresses all unwanted lines), a collimating lens, a half-silvered mirror, and continues through the optical flat and towards the specimen. Use of an optical flat with a high refractive index ensured a reasonably high reflection coefficient at the glass water interface thereby permitting interference fringes of reasonable contrast to be obtained. By having the optical flat

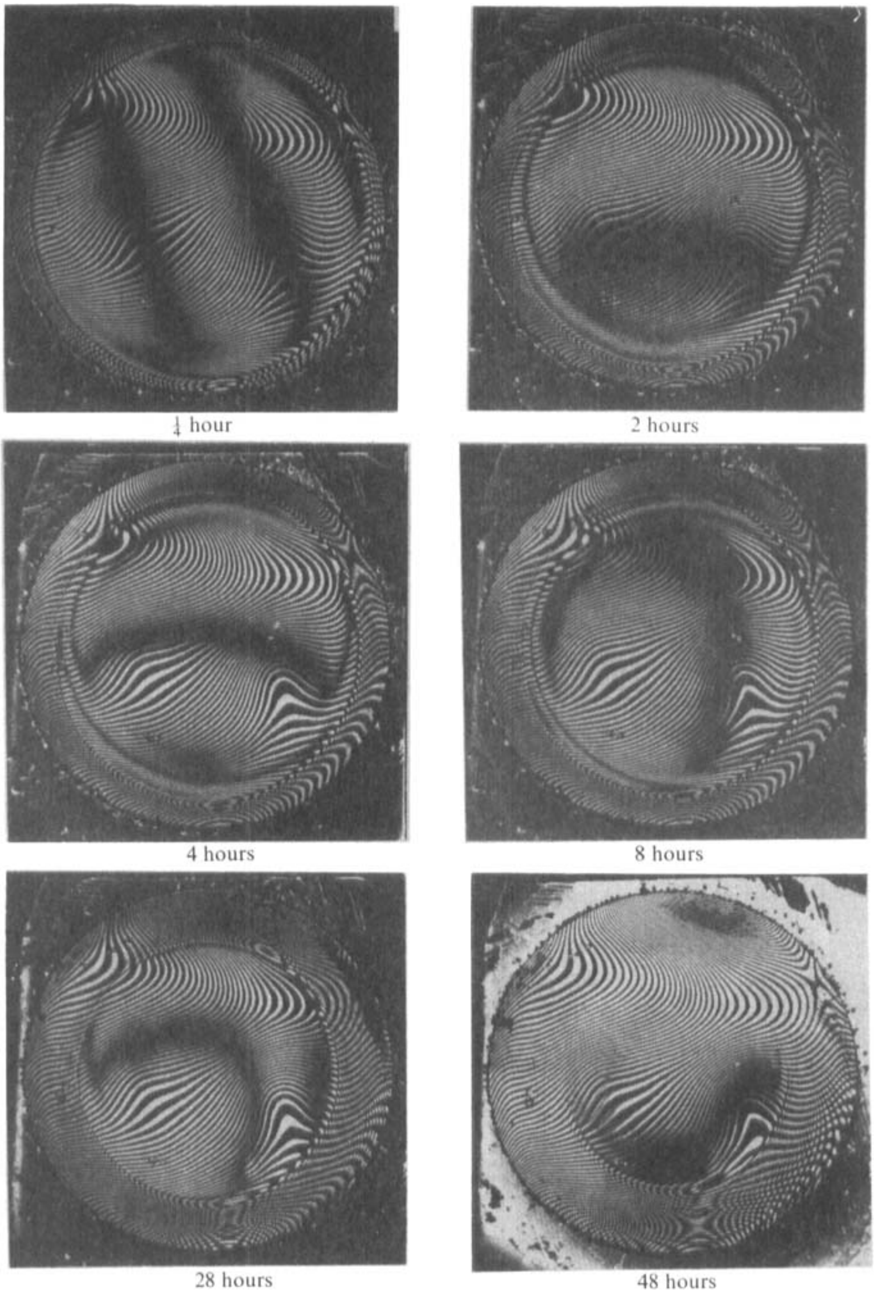


FIGURE 2 Moiré pattern for FM1000 immersed in distilled water at 62.5°C showing the ingress of swelling during water uptake.

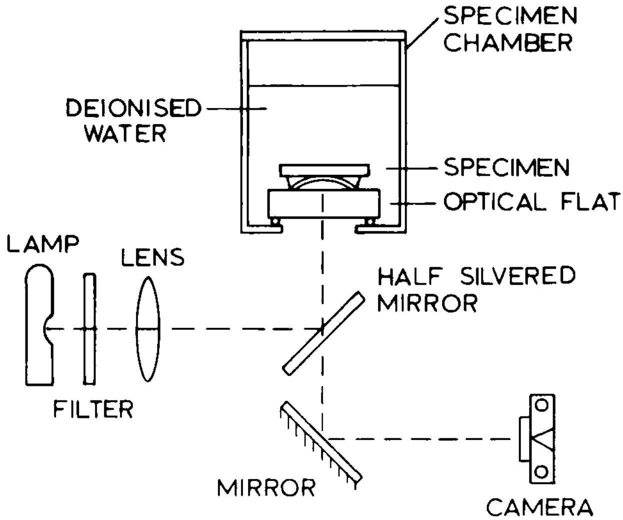


FIGURE 3 Schematic representation of the apparatus showing the optical path.

wedge-shaped it was possible to ensure that spurious reflections at the air glass interface were not coincident with the main image.

Figure 4 is a cross-section of the specimen chamber and shows the specimen mounting assembly. This arrangement permits long-term undisturbed observation of the specimen with the temperature of the specimen controlled automatically to approximately $\pm 1/10°C$ throughout the duration of a run.

JOINTS HAVING CIRCULAR GEOMETRY

Circular specimens took the form of 19 mm glass coverslips bonded with an epoxy resin to a rigid block of glass. Figure 2 is a sequence of Moiré patterns generated with a specimen manufactured using FM1000, a nylon modified epoxy film adhesive cured at 170°C in accordance with the manufacturer's recommendations (Messrs Bloomingdale Rubber Co) and tested at 62.5°C. Figure 5 shows plots of the swelling profile across a single diameter at different immersion times. The general features revealed in Figure 5 are in accord with behaviour found for a range of adhesive films,[1] *i.e.* a circumferential region of swollen resin with a water concentration that decreases from the rim of the specimen towards its centre.

The swelling is non-uniform and therefore has considerable self-stressing associated with it. There are two mechanical constraints to consider, namely the influence of swollen resin upon unswollen resin, and the influence of

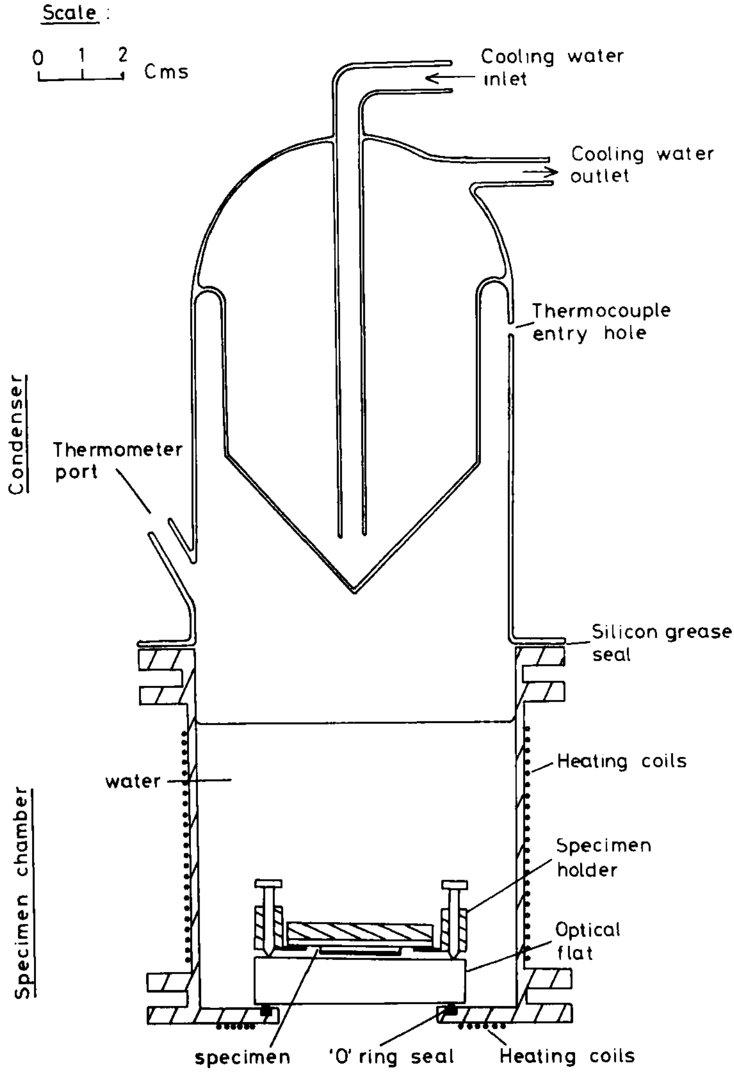


FIGURE 4 A cross-sectional drawing of the specimen chamber showing the specimen mounting assembly.

flexural rigidity of the cover slip. FM1000 is characterised by a very large hygro coefficient of expansion (approximately 12% linear expansion at saturation) and by using a thicker than normal cover slip (0.3 mm) it was possible to study an extreme case of the behaviour of a circular joint subjected to an aqueous environment.

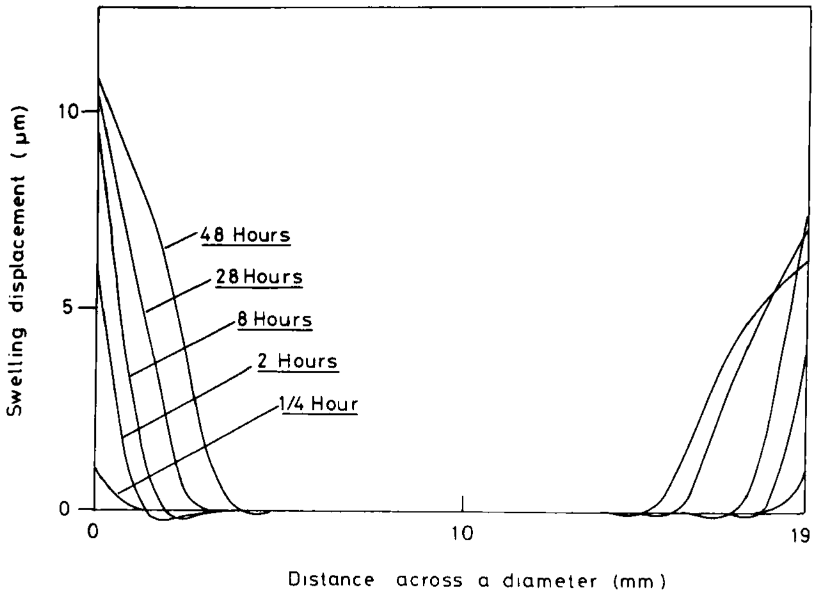


FIGURE 5 The change in adhesive film profile for the specimen from Figure 2 across one diameter as a function of exposure time.

A noteworthy feature of Figure 5 is the compression of resin immediately ahead of the swelling front. This manifests itself in Figure 2 which shows that the innermost Moiré fringes form closed loops, there being a region of minimum displacements within each loop. This observation was investigated further using a specimen manufactured using the same resin but with a thicker cover slip which had a square instead of a circular outline.

THE EFFECT OF A CARRIER CLOTH

Figure 6 is a sequence of Moiré fringes generated using a specimen made from Redux 312/5 (Manufactured by CIBA-GEIGY Ltd) and immersed in distilled water at 62°C. This is a film adhesive that is supported on a knitted nylon carrier cloth. The Moiré pattern reveals swelling which is in general accord with the pattern observed above but with the following differences:

1) The hygro coefficient of expansion for Redux 312/5 is very much smaller than for FM1000.

2) The swelling of Redux 312/5 is anomalous in that it gives rise to regular perturbations, *i.e.* waviness in the Moiré fringes.

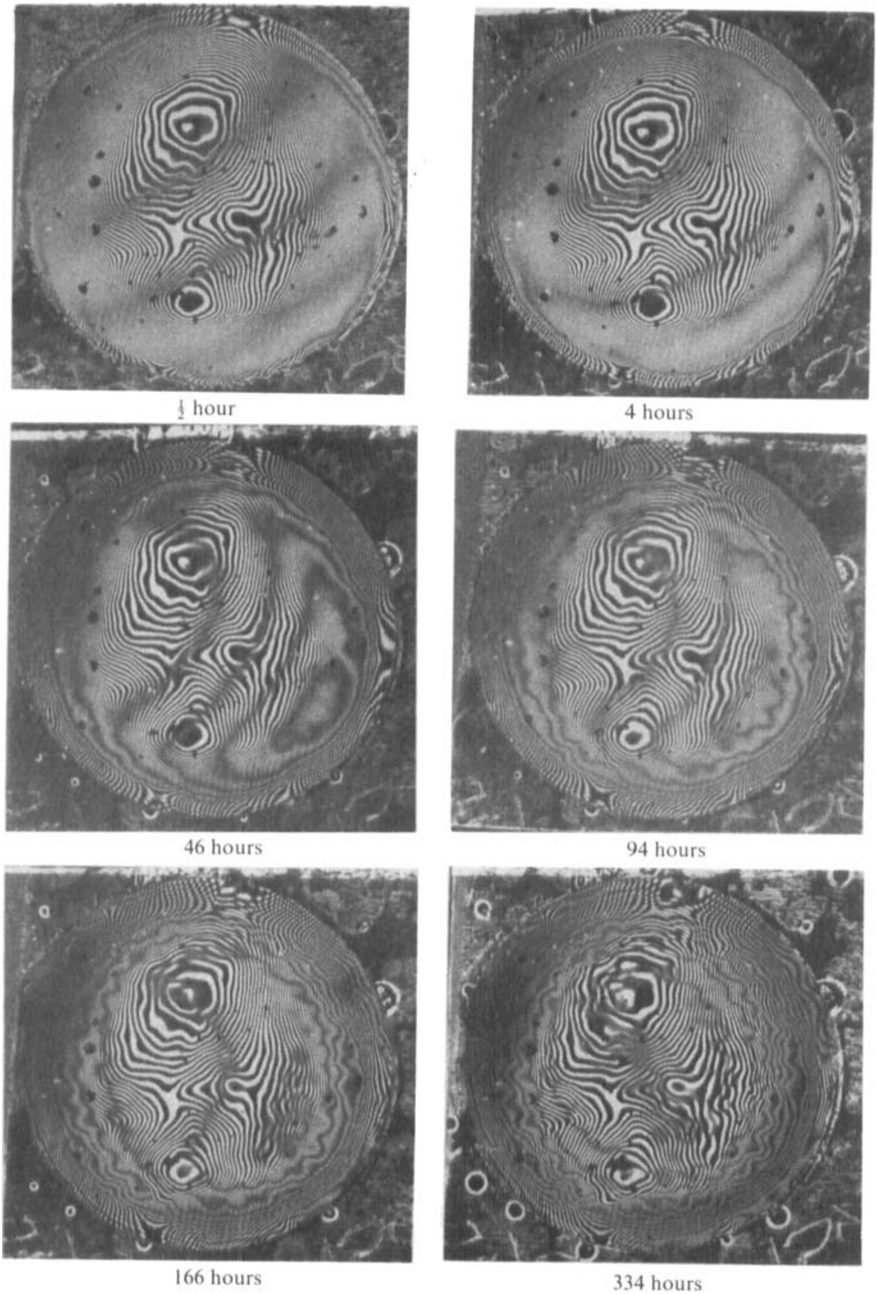


FIGURE 6 Moiré pattern for Redux 312/5 immersed in distilled water at 62°C.

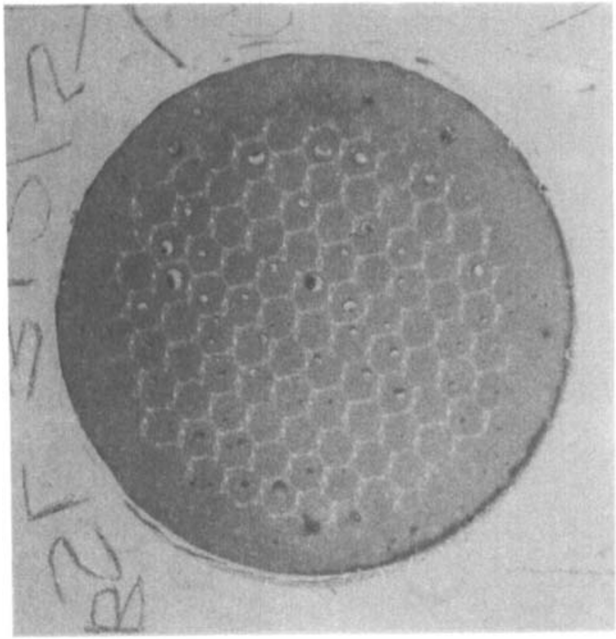


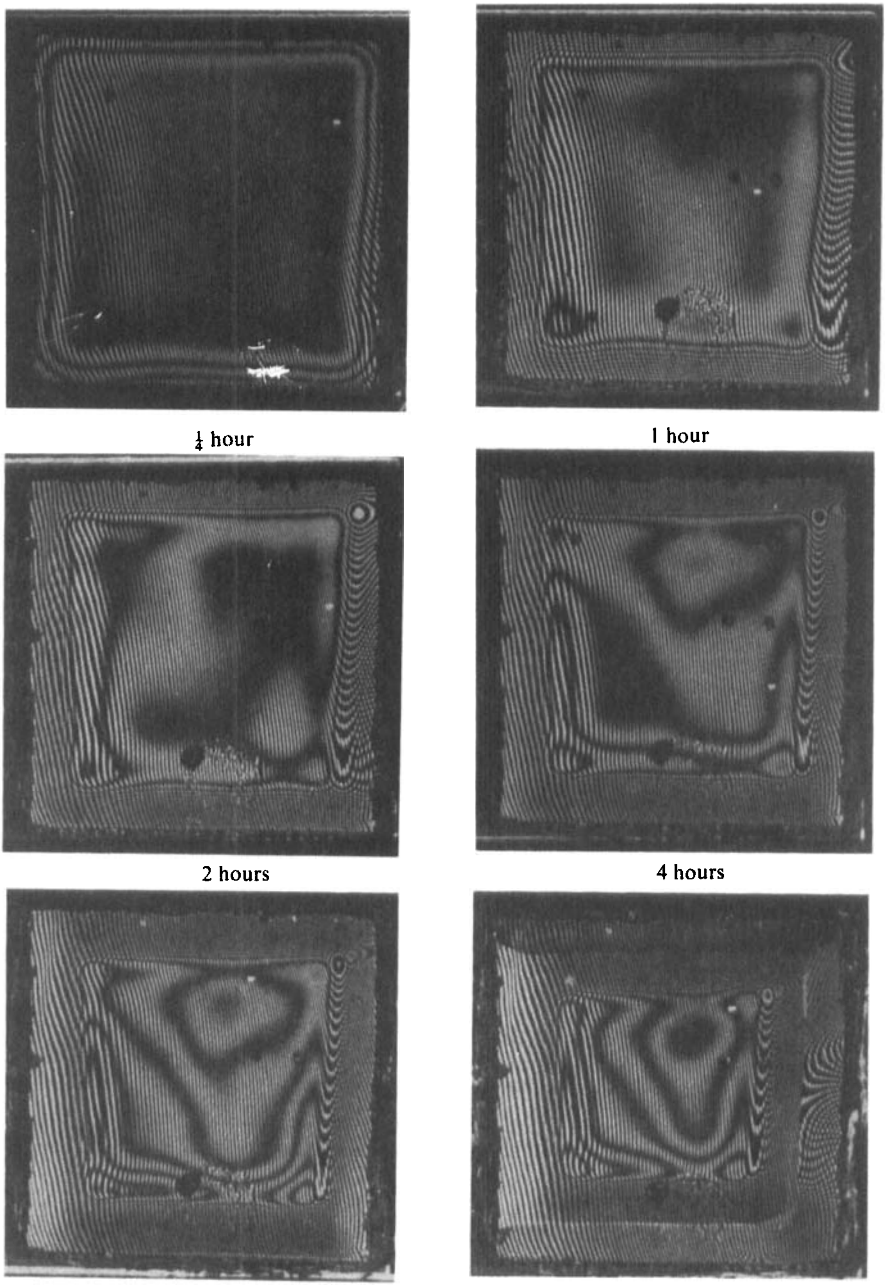
FIGURE 7 A cross-polars image of the specimen from Figure 6 showing the carrier cloth.

With respect to the second point these perturbations are particularly apparent after approximately 94 hours. Figure 7 shows the specimen viewed under crossed polars. It is evident that the wavelength of the perturbations matches the mesh size of the carrier cloth (1 mm) and, close comparison of the images established that the fingers of enhanced swelling are closely associated with individual strands in the carrier cloth. Hence it is concluded that the hygro coefficient of expansion of the nylon exceeds that of the adhesive.

JOINTS HAVING A NON-CIRCULAR EDGE

A specimen made with a square cover slip, more representative of the geometry found in technological adhesive joints was prepared for investigation. The cover slip thickness was 0.64 mm and FM1000 was used to bond it to a thick plate glass substrate. Figure 8 shows a sequence of Moiré patterns photographed at various times during the test. The specimen was immersed in distilled water at 60°C.

The square pattern of Moiré fringes, mimicking the outline of the square



¼ hour

1 hour

2 hours

4 hours

11 hours

23 hours

FIGURE 8 Moiré pattern for immersed in distilled water at 60°C.

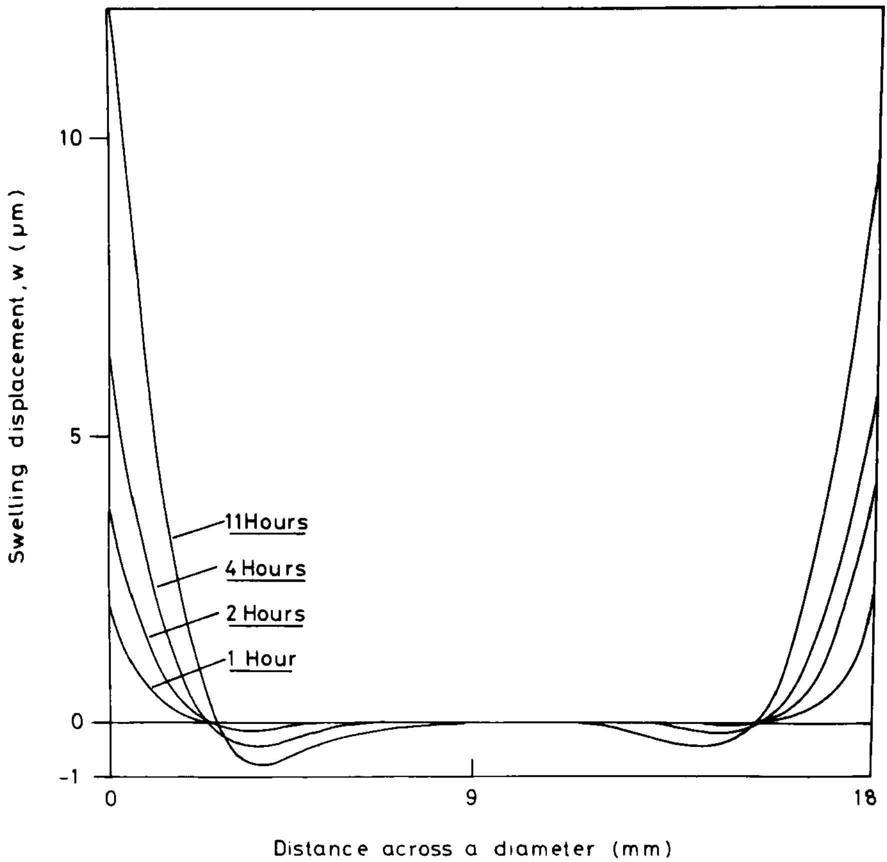


FIGURE 9 The change in adhesive film profile for the specimen from Figure 8 across one diameter as a function of exposure time.

cover slip, is typical of the pattern observed for a range of different adhesive joints with similar geometry. However, certain departures were noticed for this FM1000 joint because of the inherently large hygro coefficient of expansion and the larger than normal cover slip thickness, namely (a) the presence of a substantial region of compressed resin ahead of the swelling front and (b), fracture of the cover slip after approximately 23 hours.

The compressed internal region is noticeably more apparent here than that seen for the specimen shown in Figure 2. Thus, in Figure 8, the images obtained after 4 hours contain dark fringes towards the middle of the specimen which form large closed loops, there being regions of minima contained within these loops. Figure 9 is a plot of the swelling profile across

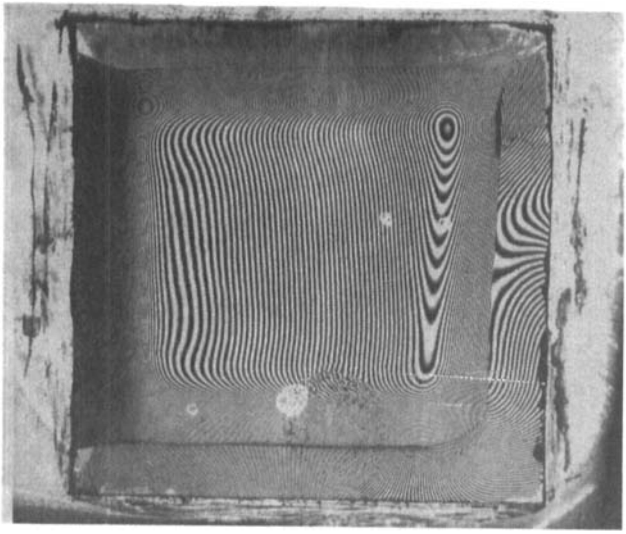


FIGURE 10 An interference pattern for the specimen from Figure 8 photographed after 23 hours and showing the fracture lines.

one diameter as indicated by the Moiré contours at different times of immersion.

The fracture of the cover slip is best resolved in Figure 10, which shows the interference pattern photographed after 23 hours' immersion. The fracture path is delineated by discontinuities in the individual interference fringes.

CALCULATION OF THE STRESS FIELD

From the behaviour of the specimen shown in Figure 8 and 10, it is evident that large stresses were generated by the resin swelling that accompanied water uptake sufficient in fact to fracture the cover slip. Using Love's[2] analysis for the displacement field in a thin shell deformed by a pressure drop across its faces, values for the normal stresses generated have been calculated as follows:

$$D\nabla^4 w = -p$$

where p = normal pressure

D = flexural rigidity, given by $D = \dfrac{2Eh^3}{3(1-\gamma^2)}$

E = Young's modulus

$2h$ = thickness of cover slip

γ = Poisson's ratio

w = axial displacement of cover slip

This equation is valid for thin plates and for normal displacements that are sufficiently small enough for lateral stretching of the plate to be negligible. To a first approximation, the cover slip deformation conforms to these criteria and results obtained predict the magnitude and positions of the normal stress for the specimen which fractured. The Moiré fringe method generates accurate data for displacement of the cover slip. Hence Love's equation was applied by graphically differentiating the displacement. The data shown in Figure 9 after 11 hours' immersion across one $\frac{1}{2}$ diameter of the specimen yielded the curves shown in Figure 11.

The second derivative is proportional to the bending stress and, as might be expected, this is a maximum at the place where the normal displacement with respect to the centre is zero.

The distribution of stress is as anticipated from the geometry of the displacement field. There is, however, one somewhat unexpected feature, namely that there exists a large tensile normal stress in a region where the normal displacement relative to that at the centre of the cover slip is zero.

DISCUSSION

When discussing the magnitude of the normal stresses indicated by this work, Sir Charles Frank drew the authors' attention to the following examples of fractures in glass caused by dimensional changes (shrinkage that accompanies curing, to be precise) in cross-linked polymers

1) The traditional method for manufacturing frosted glass was to paint plate glass with glue which, upon drying, peels and removes flakes of glass with it.

2) To comply with World War II blackout requirements, London Underground covered the windows between carriages with lace which was then coated with varnish. Holes in the fabric permitted direct contact between varnish and glass and when removed after the war these areas peeled off with flakes of glass attached, thereby leaving a permanent replica of the pattern woven into the lace.

The occurrence of swelling stresses of the order of 1 Kbar in adhesive joints between metallic components has far-reaching implications. A programme of research to measure swelling stresses in joints between microscope cover slips and massive blocks of metal (both anodised aluminium and titanium have been used) is under way and the data to date demonstrates that the

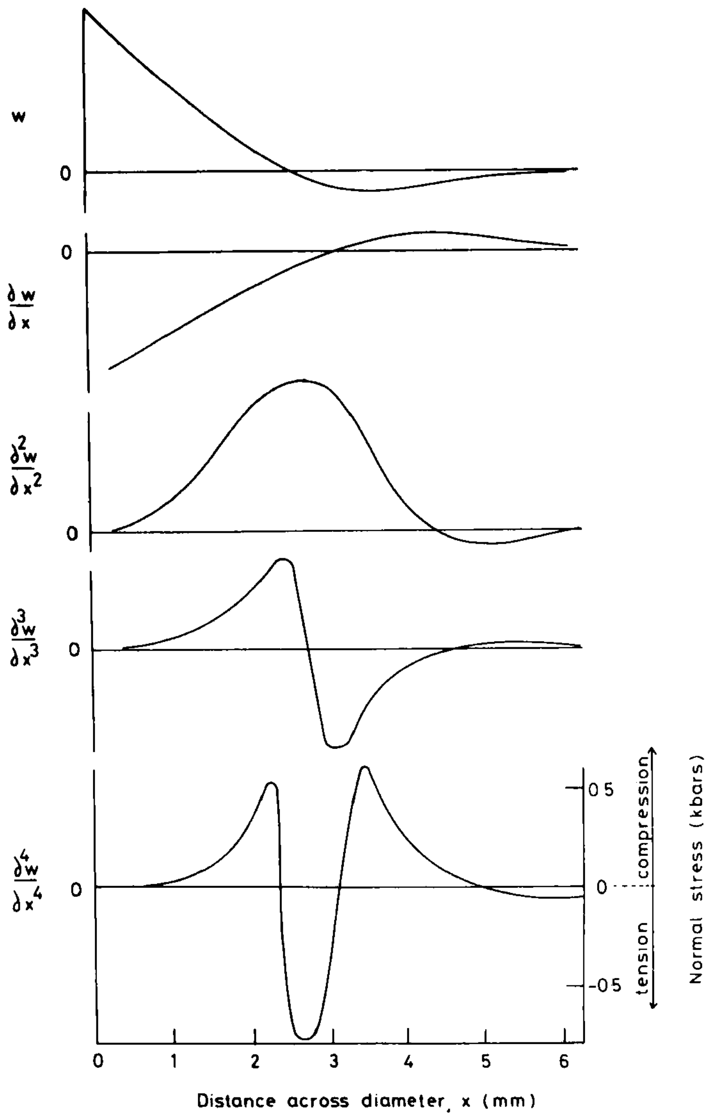


FIGURE 11 Partial derivatives of the swelling displacement with respect to distance across one half of a diameter for the specimen from Figure 8. The fourth differential shows the normal stress distribution.

phenomena reported here are features of the adhesive and not of the adherends.

Acknowledgements

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