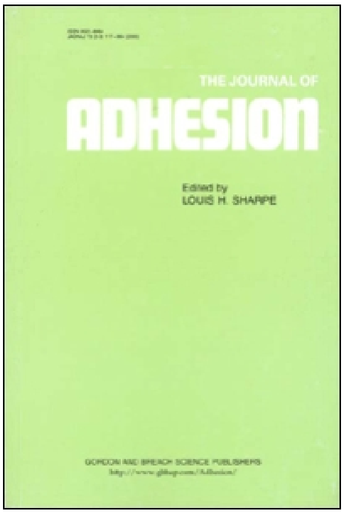

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The Deformation of the Adherends in an Adhesive Joint Undergoing Water Uptake

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The Deformation of the Adherends in an Adhesive Joint Undergoing Water Uptake

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Beam and elasticity theory have been applied to the deformation observed to occur when a model adhesive joint is exposed to water. The adhesive joint is comprised of a thin microscope cover slip/adhesive layer/rigid substrate sandwich.

The presence of damped normal displacement waves in the cover slip, predicted to exist in the joint in regions of negligible water concentration, has been confirmed by measurements on photographs of interference patterns generated with an optical interferometer.

A "theory assisted" fit for the normal displacement has led to an improved fourth derivative, and this has enabled a better estimate to be made for the distribution of normal swelling stress.

INTRODUCTION

The swelling inhomogeneity that occurs when a resin adhesive absorbs water may be conveniently demonstrated by making model joints consisting of an adhesive layer sandwiched between a rigid substrate and a flexible microscope cover slip. If such a joint is used as one of the components of an interferometer, the resulting interference pattern may be analysed to give information about the deformation of the cover slip and hence of the swelling stresses. The experimental technique, reported by Sargent and Ashbee,[1] makes use of photographs of the interference pattern to generate Moiré patterns in order precisely to follow the development of swelling in the adhesive layer.

A consequence of the inhomogeneous swelling is the development of a stress field within the adhesive. An estimate of the stresses generated normal to the joint has been made in Ref. 1 by graphically differentiating the normal displacement profiles and applying equations due to Love.[2]

The purpose of the research reported here is to make a theoretical examination of the displacement profiles previously reported and to obtain an improved method for obtaining the fourth differential of these displacement profiles with the object of more accurately calculating the resulting stress field across the adhesive joint.

The procedure adopted is to modify the equations of linear elasticity so as to include effects due to the presence of water molecules, and to apply beam theory to the cover slip in order to obtain a differential equation relating the observed displacements to the pressure acting across the surface of the cover slip. Then, by considering boundary conditions, relations can be obtained that enable a functional form for the measured displacement data to be derived.

THE DEFORMATION OF THE COVER SLIP

The equations of linear elasticity relating the stress tensor σ_{ij} to the strain tensor u_{ij}, modified to take into account the pressure due to the presence of water molecules in the resin, are of the form

$$\sigma_{ij} = 2\mu u_{ij} + \lambda\delta_{ij}u_{ii} - C(\mathbf{r})\delta_{ij}(3\lambda + 2\mu) \tag{1}$$

where μ and λ are Lamé coefficients related to Young's modulus E and Poisson's ratio ν by the two equations

$$\mu = \frac{E}{2(1+\nu)} \qquad \lambda = \frac{\nu E}{(1+\nu)(1-2\nu)}$$

and

$$C(\mathbf{r}) = \begin{pmatrix}\text{the water concentration at position}\\ \text{vector } \mathbf{r} \text{ as shown in Figure 1}\end{pmatrix} \times (\text{a material constant})$$

The boundary conditions governing the displacement vector $\mathbf{U}$ for the specimen shown in Figure 1 are

1) $U_x = 0$ at the surfaces defined by $y = 0$ and $y = L$. This is consistent with the assumption of a firm bond at the resin/glass interface.

2) $U_y = 0$ at the surface defined by $y = 0$. This is consistent with the assumption that the substrate is acting as a rigid constraint.

A suitable ansatz for $\mathbf{U}$ consistent with the above boundary conditions is

$$\mathbf{U} = (y(L-y)f(x),\ yg(x), 0) \tag{2}$$

Since the pressure on the surface of the cover slip, $P(x)$ is given by $-\sigma_{yy}\,|\, y = L$ then

$$P(x) = (3\lambda + 2\mu)C(x) - (\lambda + 2\mu)g(x) \tag{3}$$

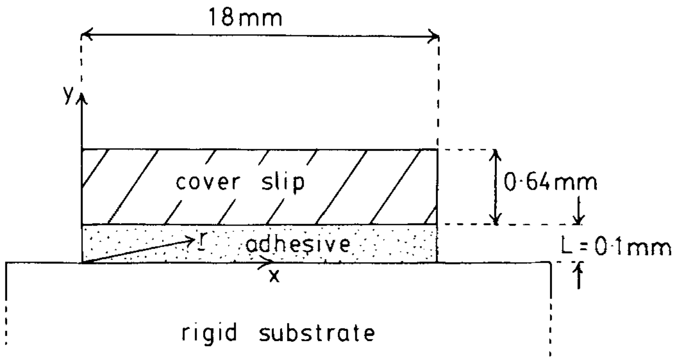

FIGURE 1 A cross-section of the specimen defining the orientation of the space axes and dimensions.

Defining the observed normal displacement of the cover slip as $W(x)$ then an additional boundary condition can be written

$$U_y = yg(x) = W(x) \quad \text{at} \quad y = L$$

hence

$$g(x) = W(x)/L$$

Substituting this in Eq. (3) gives

$$P(x) = (3\lambda + 2\mu)C(x) - (\lambda + 2\mu)W(x)/L \tag{4}$$

Application of beam theory to the cover slip gives a differential equation relating the observed displacement $W(x)$, to the pressure on the surface of the cover slip $P(x)$.

Love's result[2]† for the bending of thin plates is

$$\frac{d^2W(x)}{dx^2} = \frac{12(1-\gamma_G^2)M(x)}{E_G L_G^3}$$

where $M(x)$, E_G, γ_G and L_G are the bending moment, modulus, Poisson's ratio and thickness of the glass cover slip respectively.

Since $P(x) = d^2M(x)/dx^2$ then

$$\frac{d^4W(x)}{dx^4} = \frac{12(1-\gamma_G^2)P(x)}{E_G L_G^3}$$

† Analysis by Cottingham and Jesson[3] has shown that this equation is the first term of a series. A discussion of the errors introduced in using this result is given in the appendix.

Substitution into Eq. (4) gives

$$\frac{d^4W(x)}{dx^4} = \frac{12(1-\gamma_G^2)}{E_G L_G^3}\{(3\lambda+2\mu)C(x)-(\lambda+2\mu)W(x)/L\} \tag{5}$$

which is valid for all x except in the region near $x = 0$, due to edge effects. However, since λ and μ vary with water concentration, this equation is difficult to use in this form. Consider therefore the region of the resin where the water front has not yet penetrated, *i.e.*, the region towards the centre of the specimen. Hence $C(x) \approx 0$ and the elastic constants λ and μ assume their dry values. Equation (5) then reduces to

$$\frac{d^4W(x)}{dx^4} = \frac{-12(1-\gamma_G^2)(\gamma+2\mu)}{E_G L_G^3 L} W(x) \tag{6}$$

Noting that $W(x) \to 0$ as x is large, then the solution can be written in the form

$$W(x) = C \exp(-\phi x) \sin[\phi(x+d)] \tag{7}$$

where C is a constant

$$d = n\pi/2\phi$$

$$\phi = \frac{1}{2^{1/2}}\left(\frac{12(1-\gamma_G^2)(\gamma+2\mu)}{E_G L_G^3 L}\right)^{1/4}$$

Thus, an important consequence of the theory is the prediction of damped waves of normal displacement in the region of negligible water concentration. The predicted waves are sketched in Figure 2.

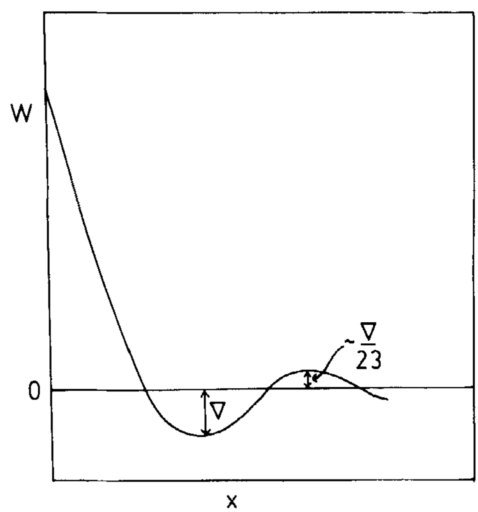

FIGURE 2 The predicted damped normal displacement waves.

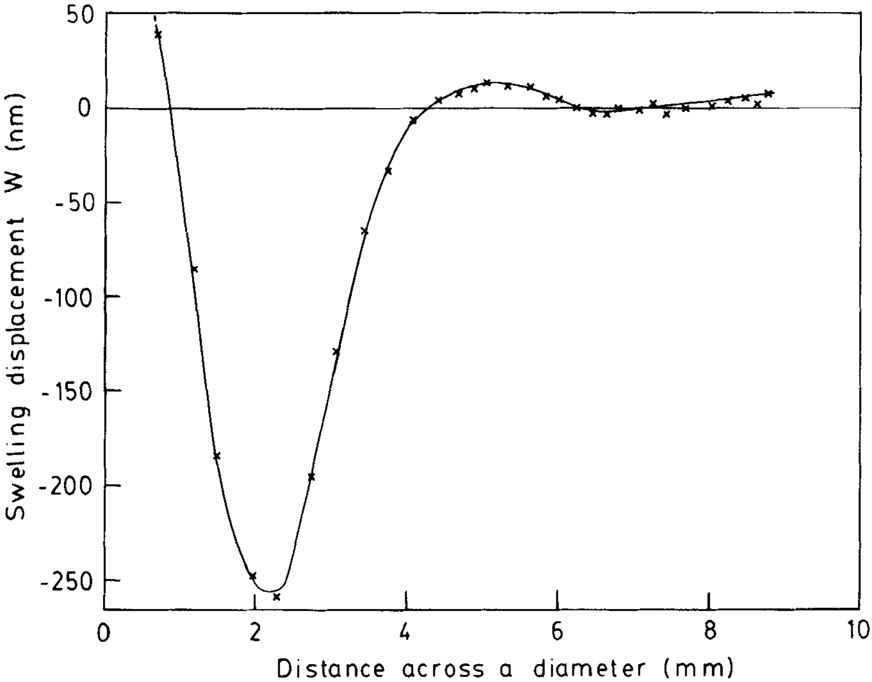

FIGURE 3 The normal displacement for the specimen from Figure 4 showing the damped normal displacement waves.

Figure 3 shows the results of a radial microdensitometer scan across the interference pattern reproduced from reference 1 in Figure 4. The specimen had undergone 4 hours exposure to distilled water at 60°C. The initial depression and first wave are clearly resolved. The height of the first peak is approximately 1/26 of the initial depression (∇), which is in good agreement with the theoretical prediction of Figure 2. It may be noted that the amplitude of the damped wave in this instance is approximately $\lambda/30$, which is below the limit of resolution directly obtainable using the Moiré technique.

A FUNCTIONAL REPRESENTATION OF THE FOURTH DERIVATIVE

To a good approximation the initial slopes of the empirical normal displacement waves may be taken as constant. Thus for small values of x, the curves may be described by the straight line

$$W(x) = -\alpha x/\beta + \alpha \quad (8)$$

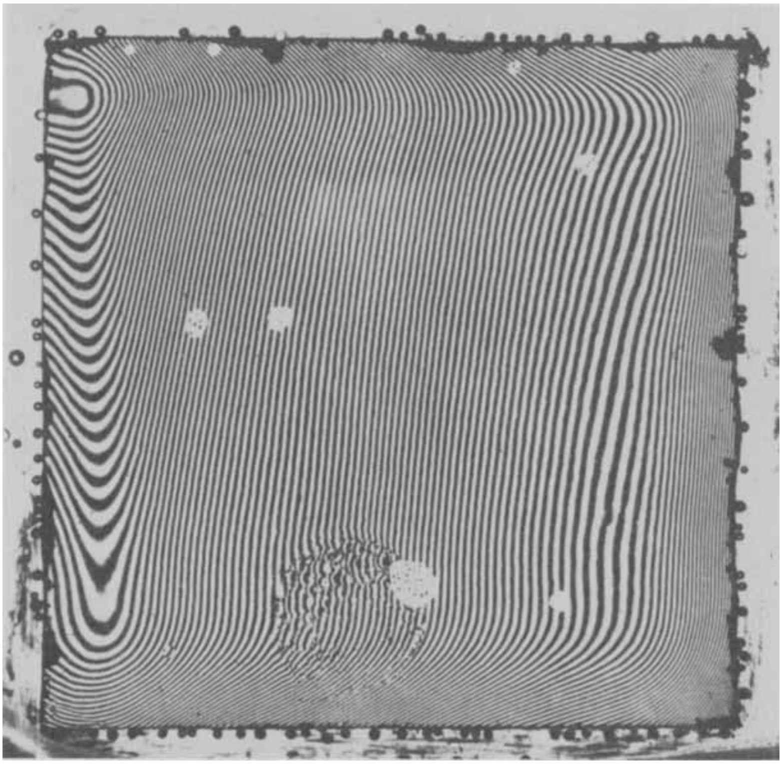

FIGURE 4 An interference pattern for a square specimen manufactured from FM1000 after 4 hours exposure to distilled water at 60°C. After Sargent and Ashbee.[1]

where α and β are intercepts on the $W(x)$ and x axes respectively.

The solution of Eq. (6) suggests that $W(x)$ is of the general form

$$W(x) = \alpha \exp(-x)(1 + ax + bx^2 + cx^3 + \cdots) \tag{9}$$

which together with the conditions that

$$\frac{d^2 W(0)}{dx^2} = 0 \quad \text{and} \quad \frac{d^3 W(0)}{dx^3} = 0$$

obtained from the application of beam theory to the cover slip, impose additional limitations on the second and third derivatives of the functional form of $W(x)$, namely that each must vanish at $x = 0$.

Defining $\chi = \alpha/\beta$ as the initial gradient of a particular displacement curve, equation 9 may be rewritten as

$$W(x) = \exp(-x)\,[\alpha+(\alpha-\chi)x+(\alpha/2-\chi)x^2+(\alpha/6-\chi/2)x^3] \qquad (10)$$

for terms up to and including x^3.

This expression represents only the first few terms of an expansion and so, to complete the fit, it is necessary to add a higher order term τx^n with $n > 3$. Equation (10) then becomes

$$W(x) = \exp(-x)\,[\alpha+(\alpha-\chi)x+(\alpha/2-\chi)x^2+(\alpha/6-\chi/2)x^3+\tau x^n] \qquad (11)$$

If a parameter x_0 is introduced as the value of x at which the normal displacement curve intercepts the x-axis, then the coefficient τ ensures that the functional representation is identically zero if

$$\tau = \frac{-W(x_0)}{x_0^n}\exp(x_0)$$

where $W(x_0)$ is expression (10) evaluated at x_0.

Estimating α, χ and x_0 from the displacement data and computing values of $W(x)$ for different values of n, it is found that the best fit is achieved for $n = 5$. The final form is therefore

$$W_F(x) = \exp(-x)\,[\alpha+(\alpha-\chi)x+(\alpha/2-\chi)x^2+(\alpha/6-\chi/2)x^3+\tau x^5] \qquad (12)$$

Equation (12) can then be differentiated four times to give the fourth derivative of displacement

$$W^{IV} = \exp(-x)\,[(4\chi-\alpha)+(3\alpha-11\chi) + (5\chi-3\alpha/2)x^2+(\alpha/2-\chi/2)x^3+\xi(x)] \qquad (13)$$

where

$$\xi(x) = -\tau(x^5-20x^4+120x^3-240x^2+120x)$$

Figure 5 shows a comparison between a plot of $W^{IV}(x)$ obtained using Eq. (13) and the graphical differentiation result employed in Ref. 1 for a time of immersion in distilled water at 60°C of 11 hours.

It has been shown that $W^{IV}(x)$ is a measure of the pressure exerted on the cover slip, thus, an interesting feature of the functional form of $W^{IV}(x)$ is the prediction of negative pressures for small x. Physically this is understood by considering the pressures exerted on the cover slip as a function of time by the waterfront as it progresses into the resin. This is shown in Figure 6. In practice, this behaviour eventually leads to debonding between the adhesive and cover slip at the edge of the specimen.

At smaller times, when there has been insufficient water uptake for the adhesive layer to saturate at the edges, we would expect the initial negative

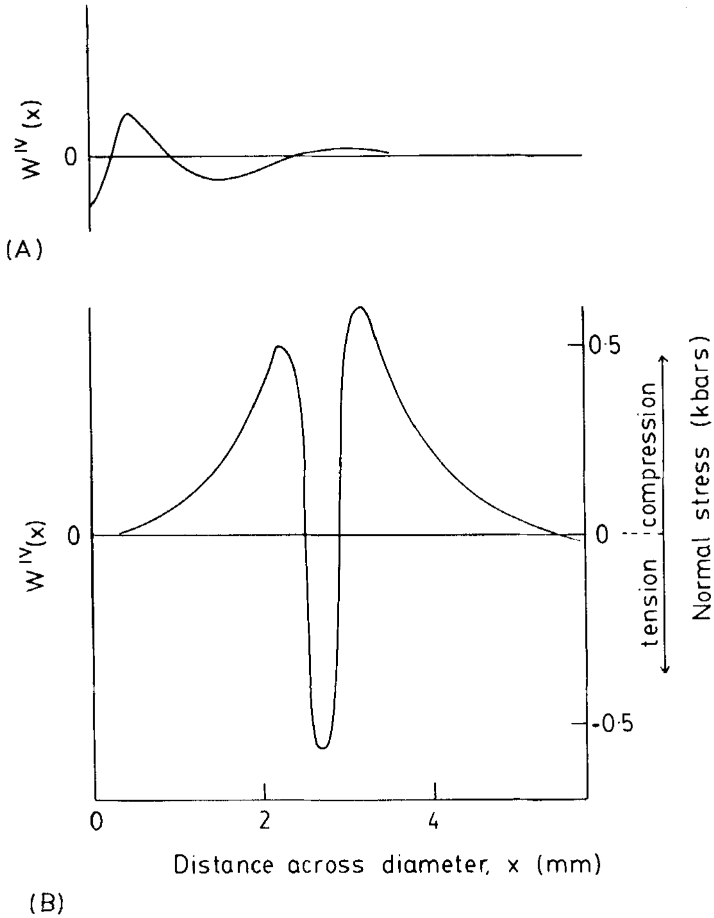

FIGURE 5a The functional form of the fourth derivative for an immersion time of 11 hours. 5b The form of $W^{IV}(x)$ obtained by graphical differentiation together with the normal stress distribution.

excursion in $W^{IV}(x)$ to vanish, which is indeed the case. This is shown in Figure 7 where the functional form of $W^{IV}(x)$ is plotted for an immersion time of 1/4 hour.

CONCLUSIONS

The application of beam and elasticity theory to the swelling of an adhesive joint consisting of a flexible cover slip/adhesive layer/rigid substrate when

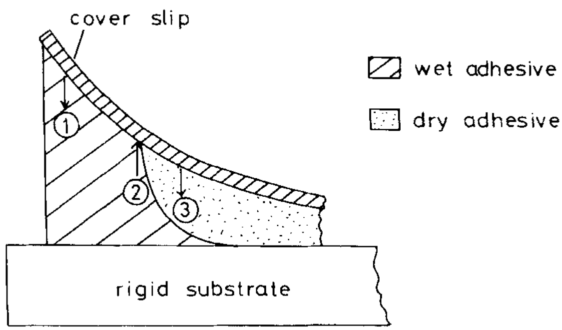

FIGURE 6 A schematic explanation for the form of the $W^{IV}(x)$ curve in Figure 5a. In regions 1 and 3 the cover slip is pulled down by dry and saturated regions of the resin. This counteracts the upward stress exerted in region 2 due to the pressure of the waterfront.

subjected to moisture absorption has resulted in the following:

1) An unexpected negative displacement of the cover slip. This is observed experimentally.

2) Damped normal displacement waves. These have also been observed in the experimental data.

3) A "theory assisted" fit for the normal displacement $W(x)$. This has facilitated the calculation of an improved fourth derivative and hence of a better estimate for the distribution of normal swelling stress.

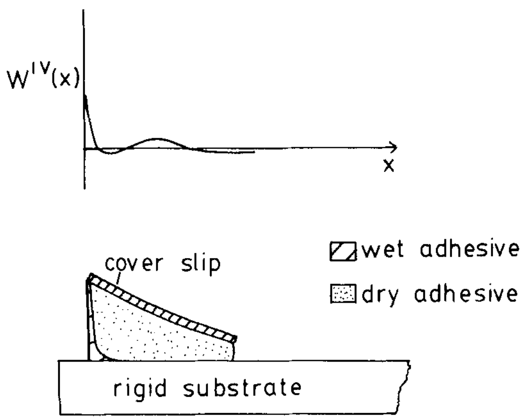

FIGURE 7 The $W^{IV}(x)$ curve for a smaller immersion time of $\frac{1}{4}$- hour together with a schematic explanation for its form.

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APPENDIX

Reference to Cottingham and Jesson[3] shows that in Fourier space

$$-A(K) = \frac{L_G^3|K|^2}{12}[\hat{O}(K)]\hat{\Pi}_2^s(K)$$

where

$$\hat{O}(K) = \frac{12}{L_G^3|K|^3}\left[\frac{(1-\exp(-2|K|L_G)-2L_G|K|\exp(-|K|L_G)}{(1+\exp(-|K|L_G))^2}\right]$$

It is shown that the Airy Stress function $A(x)$ is equivalent to the bending moment of the beam $M(x)$, and that $\hat{\Pi}_2^s(K)$ can be related to the normal displacement of the cover slip such that a modified form of Love's result taken up to 2 terms is

$$M(x) = \frac{W^{IV}(x)L_G^5 E}{60(1-v^2)} + \frac{W^{II}(x)L_G^3 E}{12(1-v^2)}$$

The equation of interest is therefore

$$\frac{d^2M(x)}{dx^2} = \frac{EL_G^3 W^{IV}(x)}{12(1-v^2)} + \frac{EL^5 W^{VI}(x)}{60(1-v^2)}$$

An indication of the error involved in the neglect of this additional term is given by the fractional change in the Airy Stress function

$$\frac{A^s(x)-A(x)}{A(x)} = \frac{L_G^2 W^{IV}(x)}{5W^{II}(x)}$$

where $A^s(x)$ is the Airy Stress function inclusive of the additional term.

From experimental data it is found that this fractional difference can be of significance. However, errors in the determination of $W^{VI}(x)$ may introduce more inaccuracy than in using Love's approximation alone.

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2. A. E. H. Love, *Treatise on the Mathematical Theory of Elasticity* (Cambridge University Press, 1959), Chap. XXII, p. 455.
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