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An Elastic-Plastic Model for the Non-Linear Mechanical Behaviour of Rubber-Toughened Adhesives

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SUMMARY

Details are presented of a proposed model for describing the non-linear, stress/strain curves of rubber-toughened adhesives. Following an initial elastic response at low strains, it is assumed that the material undergoes plastic deformation which, for dilatational stress states, is enhanced by the generation and growth of cavities within the rubber particles. The model is based on modifications to Gurson’s theory of plasticity in porous materials. Parameters are included that allow for the effect of pressure on the yield stress of matrix material between the cavities and for the influence of void interactions on matrix shear banding. Account is also taken of the change in matrix composition, and hence the matrix yield stress, during void nucleation. The nucleation is assumed to occur over a critical range of volumetric strain $\varepsilon_V$ and to involve the replacement of rubber particles by an equal volume of effective cavities. Two different nucleation functions have been investigated to describe the dependence of the effective void fraction on $\varepsilon_V$.

General relations are presented that govern the elastic, yield and flow behaviour under multiaxial stress states. Derived equations are also given in an Appendix for uniaxial compression, shear, uniaxial tension and butt-joint tension respectively. These equations were used in determining required material parameters and subsequently predicting the behaviour in compression, tension and butt-tension from shear hardening data. The predicted and observed behaviour show quite good agreement, indicating that the model may be applied with some confidence to the multiaxial stress states in finite element calculations.
1 INTRODUCTION

Elastic-plastic models were originally developed to describe the stress/strain behaviour of metals but they have been applied with some success to plastics, adhesives and other materials. The models assume that the onset of non-linearity in a stress/strain curve arises from plastic deformation and occurs at a stress level regarded as the first yield stress. Subsequent increases of stress with strain are then attributed to strain hardening. Although this approach neglects the viscoelastic effects observed with plastics and adhesives, the associated rate-dependence of the stress/strain curves can be incorporated into the models. Furthermore, elastic-plastic models have been proposed for porous materials. With some modification, these should be applicable to rubber-toughened adhesives, and allow for the effects of rubber particle cavitation on the yield and plastic deformation for stress states having a dilatational component.

Following the work of McClintock\(^1\) and Berg\(^2\), Gurson\(^3\) developed a yield stress criterion and flow rule for ductile materials containing a given volume fraction of voids. The materials were modelled as a single spherical or cylindrical void in a shell of matrix material of similar geometry, and yielding of the matrix was assumed to be governed by the pressure-independent von-Mises criterion. The results of this analysis illustrated the effect of voids in reducing the yield stress and the role of hydrostatic (dilatational) stress in enhancing this reduction and promoting void growth. For metallic materials, Chu and Needleman\(^4\) introduced a void nucleation criterion into the Gurson model to allow for the generation of new voids during the deformation. This was assumed to involve the cracking or debonding of rigid particle inclusions determined by the plastic strain in the matrix. Subsequently Tvergaard\(^5\) proposed the inclusion of parameters in the yield criterion to account for the effects of void interactions on the localised shear/dilatational banding in the matrix. With the above modifications, and using a method for integrating the elastoplastic equations developed by Aravas\(^6\), the Gurson model has been implemented into an ABAQUS finite element package\(^7\) for the stress-analysis of metallic components.

For rubber-toughened adhesives, the yield behaviour under dilatational stress states is similarly affected by the generation of voids in the dispersed rubber particles. The cavitation produces an increase in stress concentration in the glassy polymer matrix and enables the material to dilate.
through the growth of voids which is made possible through matrix shear yielding. Cavitated rubber particles can also promote multiple crazing in the matrix for those polymers prone to craze formation under dilatational stress states. Bucknall and coworkers\textsuperscript{8-10} have discussed these effects and proposed that rubber particle cavitation occurs at a critical volumetric strain such that the strain-energy released is sufficient to increase the surface area of the void and biaxially stretch the surrounding layers of rubber. On this basis, the critical volumetric strain was shown to increase with decreasing radius of the rubber particle and with increasing rubber modulus and surface energy. Lazzeri and Bucknall\textsuperscript{9} also proposed that yielding in a cavitated polymer could be described by Gurson’s yield criterion after incorporating a term that allowed for the pressure dependence of the yield stress for the (uncavitated) matrix polymer.

In this report two further extensions to Gurson’s model are proposed, appropriate to its application to rubber-toughened adhesives. These involve (a) the introduction of a void nucleation function relating the void fraction to the applied volumetric strain and (b) the derivation of a relation for the matrix shear yield stress that allows for the changing composition of the matrix during the rubber-particle cavitation. Methods are discussed for deriving the model parameters using data obtained for a rubber-toughened epoxy resin from tests in shear and under uniaxial tension and compression. The validity of the model is assessed through its ability to describe the stress/strain behaviour for each of the above three stress states and in butt-tension tests on an adhesive joint.

Standard notation is used throughout the report. Boldface symbols denote tensors and the summation convention applies to repeated subscripts on the tensor components. Double dots are used to indicate the following products

\[
\mathbf{A} : \mathbf{B} = A_{ij} B_{ij}
\]

\[
\mathbf{C} : \mathbf{B} = C_{ijkl} B_{kl} \quad (i, j, k, l = 1, 2, 3)
\]

where \(\mathbf{A}\) and \(\mathbf{B}\) represent second order tensors and \(\mathbf{C}\) denotes a fourth order tensor.
2 CONSTITUTIVE RELATIONS FOR GENERAL STRESS STATES

2.1 ADDITIVITY OF ELASTIC AND PLASTIC STRAINS

A basic assumption underlying the elastic-plastic models is that a given increment of (true) strain \( \varepsilon \) can be expressed as the sum of an elastic (recoverable) component \( \varepsilon^e \) and a plastic (non-recoverable) component \( \varepsilon^p \), i.e.

\[
\varepsilon = \varepsilon^e + \varepsilon^p
\]  

(1)

2.2 MULTIAXIAL ELASTICITY EQUATION

Assuming that the elastic strains are small and that the elastic behaviour is linear then the (true) stress \( s \) is related to \( e^e \) by

\[
s = D^e : e^e
\]  

(2)

where \( D^e \) is the fourth-order elasticity tensor. For isotropic elasticity the elements of \( D^e \) are given by

\[
D_{ijkl} = \frac{E}{(1 + \nu_e)} \delta_{ik} \delta_{jl} + \frac{E\nu_e}{(1 + \nu_e)(1 - 2\nu_e)} \delta_{ij} \delta_{kl}
\]  

(3)

where \( E \) and \( \nu_e \) are the elastic Young’s modulus and the elastic Poisson’s ratio, respectively, and \( \delta_{mn} \) (\( m,n = i,j,k,l \)) is the Kronecker delta.
2.3 THE YIELD CRITERION

The proposed modification to Gurson’s yield function is as follows

$$\Phi = \frac{\sigma_e^2}{\sigma_M^2} - (q_1 f)^2 + 2q_1 f \cosh \left( \frac{3\sigma_k}{2\sigma_M} \right) - \left( 1 - \frac{\mu \sigma_k}{\sigma_M} \right)^2 = 0 \quad (4)$$

where $\Phi$ is defined such that whenever $\Phi < 0$ the response is purely elastic. In this equation the effective (Mises) stress $\sigma_e$ and the mean normal stress $\sigma_k$ are given by

$$\sigma_e = \sqrt{3} J_{2D}^{\sigma} = \left( \frac{3}{2} S : S \right)^{\sigma} \quad (5)$$

and

$$\sigma_k = \frac{J_1}{3} = \frac{1}{3} s : I \quad (6)$$

where $J_{2D}$ is the second invariant of the deviatoric stress tensor,

$$S = s - \sigma_k I \quad (7)$$

$J_1$ is the first invariant of $s$ and $I$ is the second order identity tensor. The yield stress $\sigma_M$ in equation (4) is equal to $\sqrt{3} \sigma_S$ for the matrix material between voids, $\sigma_S$ being the (microscopic) shear yield stress. The effective void fraction is denoted by $f$ and $q_1$ is a parameter proposed by Tvergaard$^5$ to account for the effect of void interactions on the shear/dilatational banding in the matrix (the parameters $q_2$ and $q_3$, also introduced by Tvergaard, have been eliminated by taking $q_2 = 1$ and $q_3 = q_1^2$). The parameter $\mu$ was introduced into Gurson’s yield function by Lazzeri and Bucknall$^9$ to allow for the dependence of yield stress on the mean normal stress for the unvoided polymer ($f = 0$).
2.4 THE FLOW RULE

According to the elastic-plastic models, increments of plastic strain are given by the flow rule,

\[ \text{de}^p = d\lambda \frac{\partial g}{\partial s} \]  \hspace{1cm} (8)

where \( g \) is the plastic potential and \( \lambda \) is a scalar multiplier. In the case of “associated” flow, the plastic flow direction is along the outward normal to the yield surface and the yield function \( \Phi \) is employed as the plastic potential. Assuming that associated flow occurs with the modified Gurson model we thus write

\[ \text{de}^p = d\lambda \frac{\partial \Phi}{\partial s} \]  \hspace{1cm} (9)

where from equation (4)

\[ \frac{\partial \Phi}{\partial s} = \frac{3S}{\sigma_M^2} + q \frac{f}{\sigma_M} \sinh \left( \frac{s : I}{2\sigma_M} \right) + \frac{2\mu I}{3\sigma_M} \left( 1 - \frac{\mu s : I}{3\sigma_M} \right) \]  \hspace{1cm} (10)

A relation for \( \lambda \) is obtained by equating the plastic work done in the voided material under the given stress state to the equivalent plastic work for the matrix material, i.e.

\[ s : \text{de}^p = (1 - f) \sigma_M \text{ de}_0^p \]  \hspace{1cm} (11)

In this equation, the equivalent matrix plastic strain is assumed to be independent of rubber content and hence of the void fraction. It is thus identified with the \( \varepsilon_0^p \) determined from shear data on the unvoided material (Section 3.2). From equations (9) and (11) we then have

\[ d\lambda = \frac{(1 - f)\sigma_M \text{ de}_0^p}{s : \partial \Phi / \partial s} \]  \hspace{1cm} (12)
where $\frac{d\Phi}{ds}$ is given by equation (10).

### 2.5 VOID NUCLEATION AND GROWTH

The change in void volume fraction during an increment of strain is partly due to the nucleation of voids and partly to their subsequent growth. We thus write,

$$df = df_{\text{nucl}} + df_{\text{gr}}$$

In this report it is assumed that cavities are generated only in the rubber particles and that each particle offers negligible resistance to deformation once a small cavity has formed. The nucleation process is then regarded as involving the progressive replacement of rubber particles by an equal volume of effective voids. It should be emphasised that the volume fraction of these effective voids can be much larger than the actual volume fraction of voids within the material and may increase from zero to a value equal to the volume fraction of rubber $v_{R0}$ in the material.

Following the work of Bucknall et al.\cite{8-10} we assume that a cavity is nucleated in a rubber particle at some critical volumetric strain in the particle that increases with the rubber shear modulus and surface energy and decreases with increasing particle diameter. For a distribution of particle sizes, the void nucleation should then occur over a range of total volumetric strain $\varepsilon_V = e : I$ related to the critical volumetric strain range for the rubber particles.

Two different void nucleation functions have so far been explored. In the first of these, the range of $\varepsilon_V$ values for nucleation follows a normal (Gaussian) distribution with mean value $\varepsilon_{NV}$ and standard deviation $S_{NV}$:

$$f_{\text{nucl}} = \frac{v_{R0}}{S_{NV} \sqrt{2\pi}} \int_0^{\varepsilon_V} \exp \left[ -\frac{1}{2} \left( \frac{\varepsilon_V - \varepsilon_{NV}}{S_{NV}} \right)^2 \right] d\varepsilon_V$$

(14)
This relation is similar to that proposed for metals\textsuperscript{4,6,7} except that \( \varepsilon_V \) is used in place of the equivalent plastic strain in the matrix. The value of \( f_{\text{nucl}} \) increases from zero to the known value of \( \nu_{R0} \). In the second function considered, \( f_{\text{nucl}} \) is zero for volumetric strains up to some initial value \( \varepsilon_{1V} \) and subsequently increases with \( \varepsilon_V \) according to a stretched-exponential function:

\[
\begin{align*}
\text{for } \varepsilon_V \leq \varepsilon_{1V} & f_{\text{nucl}} = 0 \\
\text{for } \varepsilon_V > \varepsilon_{1V} & f_{\text{nucl}} = \nu_{R0} \left[ 1 - \exp \left( - \left( \frac{\varepsilon_V - \varepsilon_{1V}}{\varepsilon_{2V}} \right)^{\beta_V} \right) \right]
\end{align*}
\]  

(15a) (15b)

In equation (15b) the parameters \( \varepsilon_{2V} \) and \( \beta_V \) (0<\( \beta_V \)<1) determine the location and breadth of the volumetric strain range over which void nucleation occurs.

The expansion of voids following their nucleation occurs through plastic deformation within the matrix. Assuming that this is dominated by shear, and that volume changes within the matrix material are negligible, then the increase in effective void volume fraction due to growth is related to the total plastic volume change by

\[
\text{df}_{gr} = (1-f) \text{ de}^p \cdot I
\]

(16)

2.6 EVOLUTION OF THE MATRIX COMPOSITION AND YIELD STRESS \( \sigma_M \)

As discussed in Section 2.5, the nucleation of voids is considered to involve the replacement of rubber particles with cavities having the same effective volumes as the original particles. The volume fraction \( \nu_{R0}^* \) of rubber particles in the “matrix” surrounding the voids will therefore decrease as the nucleation proceeds. If \( V, V_R \) and \( V_V \) are the instantaneous values of the total volume, unvoided rubber volume and effective void volume, respectively, during the nucleation then

\[
V_R^* = \frac{V_R}{V - V_V} = \frac{V_{R0} - V_V}{V - V_V} = \frac{V_{R0} - f_{\text{nucl}}}{1 - f_{\text{nucl}}}
\]

(17)
where $V_{R_0}$ and $v_{R_0}$ are the initial volume and volume fraction, respectively, of rubber particles.

The decrease in $v_R^*$ can have a marked influence on the increase in matrix yield stress $\sigma_M$ during the nucleation. In modelling this effect, we let $\sigma_0$ and $\sigma_{01}$ denote the values of $\sqrt{3}\sigma_S$ for the unvoided material with and without rubber particles, respectively, where $\sigma_S$ is the shear yield stress. Available evidence suggests that $\sigma_0$ and $\sigma_{01}$ are related by

$$\sigma_0 = \sigma_{01} \left(1 - k v_{R_0}^*\right)$$  \hspace{1cm} (18)

where $k$ is a constant. Since $\sigma_M$ is the value of $\sqrt{3}\sigma_S$ for the matrix material with rubber volume fraction $v_R^*$ we write

$$\sigma_M = \sigma_{01} \left(1 - k v_R^*\right) = \sigma_{01} \left[1 - k \left(\frac{v_{R_0} - f_{\text{nucl}}}{1 - f_{\text{nucl}}}\right)\right]$$  \hspace{1cm} (19)

Using equation (18) we then obtain

$$\sigma_M = \frac{\sigma_0 (\varepsilon_0^p)}{1 - k v_{R_0}} \left[1 - k \left(\frac{v_{R_0} - f_{\text{nucl}}}{1 - f_{\text{nucl}}}\right)\right]$$  \hspace{1cm} (20)

In equation (20) the plastic strain $\varepsilon_0^p$ equals $\gamma_0^p / \sqrt{3}$ where $\gamma_0^p$ is the engineering shear strain for the unvoided rubber-toughened adhesive. The variation of $\sigma_0$ with $\varepsilon_0^p$ can be determined from shear tests on the unvoided material and represents the basic hardening data required in applications of the model. For several adhesives and plastics this variation can be modelled by the stretched-exponential function

$$\sigma_0 = \sigma_{0U} + (\sigma_{0R} - \sigma_{0U}) \left\{1 - \exp\left[-\left(\frac{\varepsilon_0^p}{\varepsilon_{0U}}\right)^{\beta_0}\right]\right\} + g_0 \varepsilon_0^p$$  \hspace{1cm} (21)
where $\sigma_{0U}$, $\sigma_{0R}$, $\varepsilon_{0S}$, $\beta_0$ ($0 < \beta_0 \leq 1$) and $g_0$ are parameters characterising the non-linear behaviour in shear.

3 DETERMINATION OF THE MODEL PARAMETERS AND YIELD SURFACES

In this section we illustrate methods for evaluating the model parameters through analyses of stress/strain data for a commercial Epoxy adhesive containing 13.7% rubber ($v_{R0} = 0.137$) and 16% talc filler. The data were obtained at 23 °C using uniaxial tension, shear and uniaxial compression tests, respectively. Details of these methods have been reported previously\textsuperscript{12,13}. The strain rates employed in the tensile and compressive tests were close to the effective shear strain-rate ($\dot{\gamma} / \sqrt{3}$) of 0.0024 s\textsuperscript{-1}.

3.1 ELASTIC MODULUS AND POISSON’S RATIO

Values of $E$ and $\nu_e$ were calculated from tensile stress/strain data obtained in the linear, low-strain region, employing contact extensometers for the accurate measurement of both longitudinal and lateral strains. The derived $E$ and $\nu_e$ values are included in Table 1.

3.2 DETERMINATION OF $k$

It follows from equation (18) that $k$ could be determined from measurements of $\sigma_0$ and $\sigma_{01}$, the effective stresses ($\sqrt{3}\sigma_s$) from shear data for the rubber-toughened and untoughened materials, respectively. However, attempts to measure $\sigma_{01}$ for specimens of the adhesive obtained without rubber were unsuccessful owing to premature failure in a region of stress concentration within the Arcan shear specimens. A value for $k$ was thus derived from compression tests assuming that $\mu$ is independent of rubber content. With this assumption we obtain from (18)
\[ k = \frac{1}{v_{R0}} \left[ 1 - \frac{\sigma_0}{\sigma_{01}} \right] = \frac{1}{v_{R0}} \left[ 1 - \frac{\sigma_{Ct}}{\sigma_{Cu}} \right] \]  

(22)

where \( \sigma_{Ct} \) and \( \sigma_{Cu} \) are compressive yield stresses for the toughened and untoughened adhesive respectively.

Figure 1 shows plots of true stress \( \sigma_C \) versus true strain \( \varepsilon_C \) for a toughened and an untoughened specimen of the Epoxy adhesive. A peak is observed in the stress/strain curve for the untoughened material associated with a decrease in stress at higher strains. On the basis of thermal analysis studies, such strain softening has been attributed to stress-induced structural changes (deageing) within the material\(^4\). The magnitude of the peak thus depends on the thermal history of the material but specimens with different heat treatments generally exhibit the same plateau or flow stress following the strain-softening effect. We thus equate \( \sigma_{Cu} \) to the flow stress (108 MPa) for \( \varepsilon_C = 0.1 \), as indicated in Figure 1, and take \( \sigma_{Ct} \) as the yield stress (84 MPa) observed at the same strain for the toughened material (for which the deageing effect is absent). With \( \sigma_{Ct}/\sigma_{Cu} = 0.78 \) equation (22) then gives a \( k \) value of 1.6.

3.3 DETERMINATION OF \( \mu \)

On the basis of the yield criterion (4), a value for \( \mu \) is best determined from measurements of the yield stresses under two different stress states for which no cavities are nucleated \((f=0)\). A combination of compression and shear tests was used for this purpose, the \( \mu \) value being calculated using

\[ \mu = 3 \left( 1 - \frac{\sigma_0}{\sigma_C} \right) \]  

(23)

which follows from equation (A5) in the Appendix. In applying this relation, a value of \( \sigma_0 \) (73 MPa) was employed, corresponding to an effective plastic strain \( \varepsilon_0^p = 0.03 \) at which void nucleation was considered complete. The value of \( \sigma_C \) should be determined at a plastic strain \( \varepsilon_C^p \) corresponding to
the effective plastic strain $\varepsilon_{0}^{p}$. In this context the relation between the increments of plastic strain $d\varepsilon_{c}^{p}$ and $d\varepsilon_{0}^{p}$ follows from the flow rule (9) together with the plastic work expression (11) and is given by equation (A9) of the Appendix. Noting that $d\varepsilon_{c}^{p} = 0$ when $d\varepsilon_{0}^{p} = 0$ and assuming that $\mu$ is constant then the integration of (A9) gives

$$\frac{\varepsilon_{c}^{p}}{\varepsilon_{0}^{p}} = \frac{3 - \mu}{3}$$

(24)

Since $\mu$ values usually lie in the range 0-0.4, it was assumed that $\varepsilon_{c}^{p} = \varepsilon_{0}^{p} = 0.03$ to determine an approximate value of $\sigma_{C}$ and hence $\mu$. From the approximate $\mu$ value we obtained a revised value of $\varepsilon_{c}^{p} = 0.027$ (using (24)) and corrected values of $\sigma_{C} = 81$ MPa and $\mu = 0.297$ (using (23)). Figure 2 illustrates the equivalent coordinates ($\sigma_{C}$, $\varepsilon_{c}^{p}$) and ($\sigma_{0}$, $\varepsilon_{0}^{p}$) on the respective curves resulting from the above calculations.

For regions of plastic strain over which $\sigma_{C}$ and $\sigma_{0}$ exhibit large variations the above procedure may be repeated iteratively to maximise the accuracy of the calculated $\mu$. Alternatively, with the aid of a polynomial fit to the $\sigma_{C}$ versus $\varepsilon_{c}^{p}$ or the $\sigma_{0}$ versus $\varepsilon_{0}^{p}$ data, an accurate $\mu$ value could be calculated from the $\sigma_{C}$ and $\sigma_{0}$ values found to satisfy the condition $\sigma_{C} \varepsilon_{c}^{p} = \sigma_{0} \varepsilon_{0}^{p}$. This relation follows from (23) and (24).

3.4 DETERMINATION OF $q_{1}$

The void interaction parameter $q_{1}$ was evaluated from tensile and shear data using the derived $k$ and $\mu$ values and the known $\nu_{R0}$. The calculations involved solving the yield criterion for tensile loading (equation (A23) of the Appendix) to obtain $q_{1}f$ from the measured $\sigma_{T}$ and $\sigma_{M}$. The $\sigma_{T}$ and $\sigma_{M}$ values were those corresponding to an effective plastic strain $\varepsilon_{0}^{p} = 0.03$ for which void nucleation is essentially complete and negligible void growth has occurred. Under these conditions $f = f_{\text{nucl}} = \nu_{R0}$ and (A23) becomes a quadratic equation in $q_{1}$ with known constant coefficients.
Using the experimental value of $\sigma_0 = 73$ MPa, obtained for $\epsilon_0^p = 0.03$, equation (20) with $f_{\text{nucl}} = v_{R0} = 0.137$ gives $\sigma_M = \sigma_0/(1-kv_{R0}) = 93.5$ MPa. An approximate $\sigma_T$ value was first determined from the tensile data assuming that $\epsilon_T^p = \epsilon_0^p$ and an approximate $q$ derived from $\sigma_M$ and $\sigma_T$ using equation (A23). A revised $\epsilon_T^p$ value was then obtained from the numerical integration of increments $d\epsilon_T^p$ using the method outlined in Appendix B.1 with the approximate $q_1$ value and other derived parameters. The calculations gave $\epsilon_T^p = 0.039$ for $\epsilon_0^p = 0.03$ (see Fig 2) from which we obtained $\sigma_T = 58.4$ MPa. The solution of (A23) then gave $q_1 = 1.95$.

### 3.5 PARAMETERS OF THE VOID NUCLEATION FUNCTIONS

The yield criterion (A23) and derived $\mu$ and $q_1$ values were used to evaluate the parameters in the void nucleation functions (14) and (15b) and for assessing the relative accuracy of the respective functions. Values of $\sigma_T$ were first calculated from the given $\sigma_M$ and $f$ (by solving (A23) using a Newton-Raphson method) and compared with the measured $\sigma_T$ over a range of strains for which void growth was negligible ($0 < f \leq f_{\text{nucl}} \leq v_{R0}$). Parameters in the function for $f_{\text{nucl}}$ were then varied iteratively to achieve optimum agreement with the $\sigma_T$ data.

It was assumed in these calculations that $\epsilon_0^p \approx \epsilon_T^p$ and experimental values of $\epsilon_V = \epsilon_T(1-2\nu)$ were related to the measured $\epsilon_T^p$, and hence $\epsilon_0^p$, by the coefficients of a polynomial function. Using the Gaussian function with selected values of $S_{NV}$ and $\epsilon_{NV}$, $f_{\text{nucl}}$ was calculated for each $\epsilon_V$, and $\sigma_M$ determined from $f_{\text{nucl}}$ and $\sigma_0$ at the corresponding $\epsilon_0^p$. The parameters $S_{NV}$ and $\epsilon_{NV}$ were then varied iteratively until the values of $\sigma_T(\epsilon_T^p)$ calculated from $\sigma_M$ and $f_{\text{nucl}}$ agreed with the observed $\sigma_T(\epsilon_T^p)$. A similar procedure was used to estimate values of $\epsilon_1V$, $\epsilon_2V$ and $\beta_V$ in the stretched-exponential function (15).

Table 1 includes the values obtained for each parameter and Figure 3 compares the variation of $f_{\text{nucl}}$ with $\epsilon_V$ for the two functions. The increase in $f_{\text{nucl}}$ is seen to be initially more rapid and subsequently more gradual for the stretched-exponential than for the Gaussian function.
3.6 PARAMETERS OF THE HARDENING FUNCTION $\sigma_0(\epsilon_0^p)$

An iterative procedure was employed to determine values of the parameters in equation (21) giving a best fit to the experimental plot of $\sigma_0$ versus $\epsilon_0^p$. These values are included in Table 1 and the fit to the data is illustrated in Figure 2.

<table>
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<th>Parameter</th>
<th>Value 1</th>
<th>Value 2</th>
<th>Value 3</th>
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<td>$\beta_Y$</td>
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3.7 CONSTRUCTION OF YIELD SURFACES

After substituting the values of $\nu_{R0}$, $k$, $\mu$ and $q_1$ into the criterion (4), the variation of $\sigma_e$ with $\sigma_k$ at yield can be calculated for different values of $\epsilon_0^p$ and $f$. Plots of $\sigma_e$ against $\sigma_k$ represent a cross-section to the yield surface and are shown in Figure 4 for the selected $\epsilon_0^p$ of 0.03. The experimental stresses used in deriving the yield parameters are shown for the case of uniaxial compression ($\sigma_k = -\sigma_e/3 = -\sigma_C/3$), shear ($\sigma_k = 0, \sigma_e = \sigma_0$) and uniaxial tension ($\sigma_k = \sigma_e/3 = \sigma_T/3$).

Compressive stresses are shown for the untoughened and toughened adhesives, as is the stress $\sigma_0$ obtained from the shear data for the toughened material and the derived stress $\sigma_{01} = \sigma_0/(1-k\nu_{R0})$ for the untoughened material. The straight line joining the compressive and shear data points for the toughened material is of slope $\mu$ and corresponds to equation (4) with $f=0$ and $\nu_{R0} = 0.137$. The parallel line for the untoughened material is also governed by equation (4), but with $f=0$ and $\nu_{R0} = 0$. 

Table 1 – Values of Parameters for a Rubber-toughened Epoxy Adhesive at 23 °C and 0.0024 s⁻¹
The tensile stress for the toughened adhesive lies on the curve calculated from (4) with $f = v_{\text{R0}} = 0.137$ and thus $q_1 f = 0.267$. This curve meets the shear axis ($\sigma_0 = 0$) at a stress equal to $\sigma_0 (1-q_1 f)$, which follows from equation (A13) of the Appendix when $f = v_{\text{R0}}$. The data point included in Figure 4 from a butt-joint tension test was derived from numerical calculations discussed in section 4.

4 COMPARISON OF PREDICTED AND OBSERVED BEHAVIOUR

Figure 5 compares the experimental stress/strain data under uniaxial compression for the rubber-toughened Epoxy with a curve calculated from the $\sigma_0$ versus $\epsilon_0^p$ data and the derived value of $\mu$. Stresses $\sigma_C$ were determined using equation (A5) and strains $\epsilon_C$ were calculated using

$$\epsilon_C = \epsilon_C^p + \frac{\sigma_C}{E} = \left( \frac{3-\mu}{3} \right) \epsilon_0^p + \frac{\sigma_C}{E}$$

which follows from the integrated forms of (A1) and (A9). The good overall agreement reflects the equality of $E$ values under tension and compression and the fact that $\mu$ is essentially independent of strain.

In Figure 6 the observed stress/strain curve under uniaxial tension is compared with curves calculated using the Gaussian and stretched-exponential void nucleation functions respectively. Since the data cover a fairly small strain range, it was assumed in the calculations that contributions from void growth to $f$ could be neglected. The equations of Appendix A.3, with the values of parameters in Table 1, were then solved numerically using the procedures outlined in Appendix B.1. Each void nucleation function gives a calculated stress/strain curve close to the experimental curve. However, the curve predicted using the Gaussian function exhibits a shoulder at strains around 0.02-0.04 which is not found experimentally, the overall shape of the tensile curve being more accurately modelled using the stretched-exponential function. This difference reflects the relative sharpness of the Gaussian function in the volumetric strain range 0.01-0.02 (Fig 3).
The observed and predicted variations with $\varepsilon_T$ of the Poisson’s ratio ($\nu = -\varepsilon_{\text{lat},T}/\varepsilon_T$) and the derived volumetric strain ($\varepsilon_V = \varepsilon_T(1-2\nu)$) are shown in Figure 7 and 8 respectively. The initial small increase in $\nu$ is accompanied by an increase in $\varepsilon_V$. This is attributed partly to an elastic expansion in the linear range and partly to yielding of the uncavitated material associated with the positive term in $\mu$ in equations (A24) and (A25). For tensile strains above 0.02 ($\varepsilon_V>0.005$), $\nu$ exhibits a gradual decrease, reflecting a more rapid increase in $\varepsilon_V$ with $\varepsilon_T$. This effect is ascribed to void nucleation which occurs over the strain range 0.02-0.05. The predicted rate of decrease of $\nu$ is somewhat smaller than that observed. This may partly be due to inherent inaccuracies in the Poisson’s ratio measurement but could also arise from the neglect of void growth contributions to $f$ or from the assumption of associated flow in the use of equation (9).

Figure 9 compares the measured curve of $\sigma_{11}$ versus $\varepsilon_{11}$ from the butt-joint tension test with predicted curves using the Gaussian and stretched-exponential void nucleation functions respectively. The initial slopes of the predicted curves agree well with the observed slope and a discrepancy of about 10% between the predicted and observed plateau yield stresses could be reduced by including the void growth contributions to $f$ in the calculated curves. Figure 9 also shows the calculated values of lateral stress $\sigma_{22}$ as a function of $\varepsilon_{11}$. From the plateau levels of $\sigma_{11}$ and $\sigma_{22}$ we calculate $\sigma_e = \sigma_{11}-\sigma_{22} \approx 34$ MPa and $\sigma_k = (\sigma_{11} + 2\sigma_{22})/3 \approx 45$ MPa. These values were used in locating the butt-joint tension point on the yield surface (Fig 4).
5 CONCLUSIONS

• For the rubber-toughened Epoxy adhesive, the proposed model gives satisfactory predictions of the stress/strain curves under compression, tension and butt-joint tension, using hardening data $\sigma_0(\varepsilon_0^p)$ obtained from shear tests.

• The predicted trends in Poisson’s ratio with tensile strain are also consistent with observations. More accurate Poisson’s ratio measurements would be required to test whether the assumption of associated flow underlying equation (9) is accurately valid.

• More accurate predictions of the behaviour at higher strains should include contributions from void growth (equation (16)) to the calculated $f$ values and consideration of possible effects of orientation hardening on the calculated stresses in uniaxial tension.

• The model could be applied with confidence in finite element calculations of component behaviour under multiaxial stress states. However, further modifications to the model are required for those toughened materials in which cavitation also occurs in the matrix phase.

6 ACKNOWLEDGEMENT

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APPENDIX A - DERIVED EQUATIONS FOR SELECTED STRESS STATES

Relevant relations are given here for compression, shear, uniaxial tension and butt-joint tension tests respectively. These equations follow from the general relations of Section 2 and provide a basis for the determination of model parameters and for interrelating or predicting the stress/strain behaviour under different stress states.

A.1 UNIAXIAL COMPRESSION

With this mode of deformation, a single negative stress $\sigma_C = -\sigma_{11}$ acts along the long direction of a rod which we denote as direction 1. The longitudinal and lateral strain increments are each decomposed into elastic and plastic components according to equation (1)

$$d\varepsilon_C = -d\varepsilon_{11} = d\varepsilon_C^e + d\varepsilon_C^p$$  \hspace{1cm} (A1)

$$d\varepsilon_{\text{lat},C} = d\varepsilon_{22} = d\varepsilon_{33} = d\varepsilon_{\text{lat},C}^e + d\varepsilon_{\text{lat},C}^p$$  \hspace{1cm} (A2)

The elasticity relations then follow from (2) and (3) as

$$\sigma_C = E\varepsilon_C^e$$  \hspace{1cm} (A3)

and

$$\varepsilon_{\text{lat},C}^e = v_e \varepsilon_C^e$$  \hspace{1cm} (A4)
Since volumetric strains are negative under compression, the value of $f_{\text{nucl}}$ (equations (14) and (15)), and hence $f$, is zero and it follows from (20) that $\sigma_M = \sigma_0$. We also obtain from equations (5)-(7) that $\sigma_e = \sigma_C$ and that $\sigma_k = -\sigma_C/3$. The yield criterion (equation (4)) then reduces to

$$\frac{\sigma_C}{\sigma_0} = \frac{3}{3-\mu} \quad \text{(A5)}$$

The increments of plastic strain are obtained from (9) and (10) as

$$d\varepsilon_C^p = d\lambda \left[ \frac{2\sigma_C}{\sigma_0^2} - \frac{2\mu}{3\sigma_0} \left( 1 + \frac{\mu \sigma_C}{3\sigma_0} \right) \right] \quad \text{(A6)}$$

and

$$d\varepsilon_{\text{lat.c}}^p = d\lambda \left[ \frac{\sigma_C}{\sigma_0} + \frac{2\mu}{3\sigma_0} \left( 1 + \frac{\mu \sigma_C}{3\sigma_0} \right) \right] \quad \text{(A7)}$$

where $d\lambda$ is given by (equations (10) and (12))

$$d\lambda = \frac{\sigma_0 d\varepsilon_0^p}{2 \left( \frac{\sigma_C^2}{\sigma_0^2} - \frac{2\mu \sigma_C}{3\sigma_0} \left( 1 + \frac{\mu \sigma_C}{3\sigma_0} \right) \right)} \quad \text{(A8)}$$

Substituting (A8) into (A6) and using (A5) we obtain

$$d\varepsilon_C^p = \frac{\sigma_0}{\sigma_C} d\varepsilon_0^p = \left( \frac{3-\mu}{3} \right) d\varepsilon_0^p \quad \text{(A9)}$$
and the increment of plastic Poisson’s ratio, obtained as the ratio of (A7) to (A6), will have values 
$\nu_p \geq 0.5$ (since $\mu \geq 0$) corresponding to a material expansion.

A.2 SHEAR

Here we consider the variation of shear stress $\sigma_S = \sigma_{ij} = \sigma_{ji}$ with engineering shear strain $\gamma = \gamma_{ij}$ in a shear test where $i,j = 1,2 \text{ or } 3 \text{ and } i \neq j$. Noting that $\gamma_{ij}$ is related to the tensor shear strains $\varepsilon_{ij}$ and $\varepsilon_{ji}$ by

$$\gamma_{ij} = \varepsilon_{ij} + \varepsilon_{ji} = 2\varepsilon_{ij} \quad (A10)$$

it follows from (1) that increments $d\gamma$ can be expressed as the sum of elastic and plastic components

$$d\gamma = d\gamma^e + d\gamma^p \quad (A11)$$

From equations (2) and (3) the elasticity relation becomes

$$\sigma_S = \frac{E}{2(1+\nu_e)} \gamma^e = G\gamma^e \quad (A12)$$

where $G$ is the elastic shear modulus.

Since the volumetric strain is zero during shear deformations, no voids should be nucleated in the rubber particles. However, it is informative to consider the yield behaviour in shear tests carried out on specimens previously voided under tension as well as on the original unvoided material.
Equations (5)-(7) give $\sigma_e = (3(\sigma_{ij}^2 + \sigma_{ji}^2)/2)^{1/2} = \sqrt{3} \sigma_s$ and $\sigma_k = 0$ and from the yield criterion (4) we have

$$\sqrt{3} \sigma_s = \sigma_M (1 - q_1 f) \tag{A13}$$

for the prevoided material. For the unvoided plastic ($f=0$, $\sigma_M = \sigma_0$) this equation reduces to

$$\sqrt{3} \sigma_s = \sigma_0 \tag{A14}$$

Noting that $S_{ij} = \sigma_{ij} = \sigma_s$ then the increment of plastic shear strain for the prevoided material becomes (equations (9), (10) and (A13))

$$d\gamma^p = d\varepsilon_{ij}^p + d\varepsilon_{ji}^p = d\lambda \left( \frac{2\sqrt{3}(1-q_1 f)}{\sigma_M} \right) \tag{A15}$$

where, using (10), (11) and (A13)

$$d\lambda = \frac{(1-f)\sigma_M d\varepsilon_0^p}{2(1-q_1 f)^2} \tag{A16}$$

Substitution of (A16) into (A15) then gives

$$d\gamma^p = \frac{(1-f)\sqrt{3} d\varepsilon_0^p}{(1-q_1 f)} \tag{A17}$$
which reduces to

\[ d\gamma^p = \sqrt{3} \, d\varepsilon^p_0 \quad (A18) \]

for the unvoided material.

A.3 UNIAXIAL TENSION

The uniaxial tension of a long, thin strip is characterised by a single non-zero stress component \( \sigma_T = \sigma_{11} \) acting parallel to the length direction 1. The additivity of elastic and plastic strain components (equation (1)) applies in both longitudinal and lateral directions,

\[ d\varepsilon_T = d\varepsilon_{11} = d\varepsilon^e_T + d\varepsilon^p_T \quad (A19) \]
\[ d\varepsilon_{\text{lat},T} = d\varepsilon_{22} = d\varepsilon_{33} = d\varepsilon^e_{\text{lat},T} + d\varepsilon^p_{\text{lat},T} \quad (A20) \]

and from equations (2) and (3) the elasticity relations become

\[ \sigma_T = E \varepsilon^e_T \quad (A21) \]
\[ \varepsilon^e_{\text{lat},T} = -v \varepsilon^e_T \quad (A22) \]

From equations (5)-(7) it follows that \( \sigma_e = \sigma_T \) and that \( \sigma_k = \sigma_T/3 \). The yield stress criterion (4) then becomes
\[ \frac{\sigma_T^2}{\sigma_M^2} - (q_1 f)^2 + 2q_1 f \cosh \left( \frac{\sigma_T}{2\sigma_M} \right) - \left( 1 - \frac{\mu \sigma_T}{3\sigma_M} \right)^2 = 0 \]  
(A23)

Equations (9) and (10) now give for the increments of plastic strain (using \( S_{11} = 2\sigma_T/3 \) and \( S_{22} = -\sigma_T/3 \)),

\[ d\varepsilon_T^p = d\lambda \left[ \frac{2\sigma_T}{\sigma_M^2} + q_1 f \sinh \left( \frac{\sigma_T}{2\sigma_M} \right) + \frac{2\mu}{3\sigma_M} \left( 1 - \frac{\mu \sigma_T}{3\sigma_M} \right) \right] \]  
(A24)

and

\[ d\varepsilon_{int,T}^p = d\lambda \left[ -\frac{\sigma_T}{\sigma_M^2} + q_1 f \sinh \left( \frac{\sigma_T}{2\sigma_M} \right) + \frac{2\mu}{3\sigma_M} \left( 1 - \frac{\mu \sigma_T}{3\sigma_M} \right) \right] \]  
(A25)

where, from equations (10) and (12)

\[ d\lambda = \frac{(1-f)\sigma_M d\varepsilon_0^p}{2 \frac{\sigma_T^2}{\sigma_M^2} + q_1 f \frac{\sigma_T}{\sigma_M} \sinh \left( \frac{\sigma_T}{2\sigma_M} \right) + \frac{2\mu}{3} \frac{\sigma_T}{\sigma_M} \left( 1 - \frac{\mu \sigma_T}{3\sigma_M} \right) + \frac{2\mu}{3} \frac{\sigma_T}{\sigma_M} \left( 1 - \frac{\mu \sigma_T}{3\sigma_M} \right) \]  
(A26)

Substituting (A26) into (A24) we obtain,

\[ d\varepsilon_T^p = (1-f) \frac{\sigma_M}{\sigma_T} d\varepsilon_0^p \]  
(A27)
From equations (A24) and (A25) the increment of Poisson’s ratio \( \Delta \nu_p = -\left( \frac{\partial \nu_{T}}{\partial \nu_{T,\text{lat}}} \right) \) will have values \( \Delta \nu_p \leq 0.5 \) representing a material expansion that increases with increasing \( f \) and \( \mu \).

Regarding the void nucleation functions (14) and (15) we note that the volumetric strain \( \epsilon_V \) under uniaxial tension is

\[
\epsilon_V = \epsilon_T + 2\epsilon_{\text{lat},T} = \epsilon_T (1-2\nu)
\]  

(A28)

where \( \nu \) is the total Poisson’s ratio. Similarly the relation (16) for void growth becomes

\[
\frac{df}{gr} = (1-f) (\partial \epsilon_T + 2\partial \epsilon_{\text{lat},T})
\]

\[
= (1-f) \partial \epsilon_T (1-2\Delta \nu_p)
\]  

(A29)

A.4 BUTT-JOINT TENSION

With the butt-tension test, a load is applied normal to the wide faces (i.e. in the through-thickness direction 1) of a wide, thin sheet or joint. The specimen geometry serves to prevent contractions occurring in the lateral (in-plane) 2 and 3 directions. Ignoring stress distributions around the edges of the joint, a stress \( \sigma_{11} \) and strain \( \epsilon_{11} \) are produced in the through-thickness direction, stresses \( \sigma_{22} = \sigma_{33} \) are set up in the lateral directions and the net lateral strains (\( \epsilon_{22} = \epsilon_{33} \)) are zero. The additivity of elastic and plastic strain components may thus be written

\[
\partial \epsilon_{11} = \partial \epsilon_{11}^e + \partial \epsilon_{11}^p
\]  

(A30)

\[
\partial \epsilon_{22} = \partial \epsilon_{22}^e + \partial \epsilon_{22}^p = 0
\]  

(A31)
and from (2) and (3) we obtain the elastic equations

\[
\sigma_{11} = \frac{E(1 - \nu_e)}{(1 + \nu_e)(1 - 2\nu_e)} \varepsilon_{11}^e + \frac{2E\nu_e}{(1 + \nu_e)(1 - 2\nu_e)} \varepsilon_{22}^e \quad (A32)
\]

\[
\sigma_{22} = \frac{E\nu_e}{(1 + \nu_e)(1 - 2\nu_e)} \varepsilon_{11}^e + \frac{E}{(1 + \nu_e)(1 - 2\nu_e)} \varepsilon_{22}^e \quad (A33)
\]

It follows from (5) and (7) that \(\sigma = \sigma_{11} - \sigma_{22}\) and that \(\sigma_k = (\sigma_{11} + 2\sigma_{22})/3\) so that the yield criterion (4) becomes

\[
\left(\frac{\sigma_{11} - \sigma_{22}}{\sigma_M^2}\right)^2 - (q_1 f)^2 + 2q_1 f \cosh\left(\frac{\sigma_{11} + 2\sigma_{22}}{2\sigma_M}\right) - \left(1 - \mu \left(\frac{\sigma_{11} + 2\sigma_{22}}{3\sigma_M}\right)\right)^2 = 0 \quad (A34)
\]

Noting that \(S_{11} = 2(\sigma_{11} - \sigma_{22})/3\) and that \(S_{22} = (\sigma_{22} - \sigma_{11})/3\) equations (9) and (10) give for the plastic strain increments

\[
de_{11}^p = d\lambda \ X_1 \quad (A35)
\]

\[
de_{22}^p = d\lambda \ X_2 \quad (A36)
\]

where

\[
X_1 = \frac{2(\sigma_{11} - \sigma_{22})}{\sigma_M^2} + \frac{q_1 f}{\sigma_M} \sinh\left(\frac{\sigma_{11} + 2\sigma_{22}}{2\sigma_M}\right) + \frac{2\mu}{3\sigma_M} \left(1 - \mu \left(\frac{\sigma_{11} + 2\sigma_{22}}{3\sigma_M}\right)\right) \quad (A37)
\]

and
\[ X_2 = \frac{(\sigma_{22} - \sigma_{11})}{\sigma_M^2} + \frac{q_1 f}{\sigma_M} \sinh \left( \frac{\sigma_{11} + 2\sigma_{22}}{2\sigma_M} \right) + \frac{2\mu}{3\sigma_M} \left( 1 - \mu \left( \frac{\sigma_{11} + 2\sigma_{22}}{3\sigma_M} \right) \right) \]  

(A38)

and, from (10) and (12)

\[
d\lambda = \frac{(1-f)\sigma_{22} d\varepsilon_0}{\sigma_{11} X_1 + 2\sigma_{22} X_2}
\]

(A39)

In butt-tension tests, the volumetric strain required in calculating \( f_{\text{nucl}} \) (equations (14) and (15)) is given by

\[
\varepsilon_v = \varepsilon_{11}
\]

(A40)

and equation (16) for void growth becomes

\[
df_{gr} = (1-f) (d\varepsilon_{11}^p + 2d\varepsilon_{22}^p)
\]

(A41)
APPENDIX B - SOLUTION OF EQUATIONS FOR UNIAXIAL TENSION AND BUTT-JOINT TENSION

Here we outline analytical methods used to solve the elastic-plastic equations for the case of uniaxial tension and butt-joint tension respectively. The calculations have so far assumed that \( f = \tau_{\text{excl}} \) and are expected to apply accurately only over strain ranges for which void growth is negligible.

B.1 UNIAXIAL TENSION

Equations (A19) to (A25) are seven relations for the seven unknowns \( \varepsilon_T, \varepsilon_{\text{lat},T}, \varepsilon^e_T, \varepsilon^e_{\text{lat},T}, \varepsilon^p_T, \varepsilon^p_{\text{lat},T} \) and \( \sigma_T \). Introducing new variables \( R = \sigma_T/\sigma_M, u = \varepsilon^p_T, v = \varepsilon^p_{\text{lat},T}, z = \varepsilon^e_T \) and \( t = \varepsilon^p_0 \) the yield function (A23) becomes

\[
R^2 - (q_1 f)^2 + 2q_1 f \cosh \left( \frac{R}{2} \right) - \left( 1 - \frac{\mu R}{3} \right)^2 = 0 \tag{B1}
\]

and, using (A26), the flow rule equations (A24) and (A25) become

\[
\frac{du}{dt} = \frac{(1 - f)}{R} \tag{B2}
\]

and

\[
\frac{dv}{dt} = \frac{(1 - f)}{R} \left\{ -R + q_1 f \sinh \left( \frac{R}{2} \right) + \frac{2\mu}{3} \left( 1 - \frac{\mu R}{3} \right) \right\} \tag{B3}
\]

\[
2R + q_1 f \sinh \left( \frac{R}{2} \right) + \frac{2\mu}{3} \left( 1 - \frac{\mu R}{3} \right) \right\}
\]
with initial conditions

\[ u = v = 0 \quad \text{when} \quad t = 0 \]

Using (A22) with the integrated forms of (A19) and (A20) we obtain for the argument \( \epsilon_v \) of the voiding functions \( f = f_{\text{nuel}} \) (equations (14) and (15b))

\[ \epsilon_v = z(1 - 2\nu_e) + u + 2v \quad \text{(B4)} \]

Noting that the new variable \( R \) is given by

\[ R = \frac{\sigma_T}{\sigma_M} = \frac{Ez}{\sigma_M} \quad \text{(B5)} \]

equations (B1) to (B3) may be solved to determine the unknowns \( u, v, z \) and the remaining unknowns follow with the aid of (A19) to (A22). A numerical scheme was developed to solve the equations using MATLAB, a software package implemented on the NPL mainframe computer. Essentially equations (B1) and (B5) constitute a non-linear equation for \( z \) which was solved using a Newton-Raphson iterative procedure. This was undertaken in parallel with numerical solutions of the ordinary differential equations (B2) and (B3).
B.2 BUTT-JOINT TENSION

Noting that for this mode of deformation the total lateral strain $\varepsilon_{22}$ is zero, equations (A30) to (A36) are seven relations for the seven unknowns $\varepsilon_{11}$, $\varepsilon_{11}^e$, $\varepsilon_{11}^p$, $\varepsilon_{22}^e$, $\varepsilon_{22}^p$, $\sigma_{11}$ and $\sigma_{22}$. For convenience we introduce another set of new variables

\[
R_1 = \frac{\sigma_{11} - \sigma_{22}}{\sigma_M}, \quad R_2 = \frac{\sigma_{11} + 2\sigma_{22}}{\sigma_M}, \quad u = \varepsilon_{11}^p, \quad v = \varepsilon_{22}^p, \quad z = \varepsilon_{11}, \quad t = \varepsilon_{0}^p
\]

Then the yield function (A34) becomes

\[
R_1^2 - (q_i f)^2 + 2q_i f \cosh \left( \frac{R_2}{2} \right) - \left(1 - \frac{\mu R_2}{3}\right)^2 = 0 \quad (B6)
\]

and, using (A37) to (A39), the flow rule equations (A35) and (A36) become

\[
\frac{du}{dt} = \frac{(1-f)}{2R_1^2 + R_2} \left[ 2R_1 + q_i f \sinh \left( \frac{R_2}{2} \right) + \frac{2\mu}{3} \left(1 - \frac{\mu R_2}{3}\right) \right] \quad (B7)
\]

\[
\frac{dv}{dt} = \frac{(1-f)}{2R_1^2 + R_2} \left[ -R_1 + q_i f \sinh \left( \frac{R_2}{2} \right) + \frac{2\mu}{3} \left(1 - \frac{\mu R_2}{3}\right) \right] \quad (B8)
\]
with initial conditions

\[ u = v = 0 \quad \text{when} \quad t = 0 \]

Equation (A40) now gives \( \varepsilon_V = \varepsilon_{11} = z \). From the definition of \( R_1 \) and \( R_2 \) together with (A32) and (A33) and the integrated forms of (A30) and (A31) we obtain

\[ R_1 = \frac{E}{(1 + v_e)} \frac{(z - u + v)}{\sigma_M} \]  \hspace{1cm} (B9)

\[ R_2 = \frac{E}{(1 + 2v_e)} \frac{(z - u - 2v)}{\sigma_M} \]  \hspace{1cm} (B10)

Equations (B6) to (B10) represent five equations for the five unknowns \( u, v, z, R_1 \) and \( R_2 \). The non-linear equation for \( z \), formed by substituting (B9) and (B10) into (B6), was again solved using a Newton-Raphson iterative procedure. This solution was effected in conjunction with the numerical solution of the differential equations (B7) and (B8) using a standard MATLAB routine.
FIGURE CAPTIONS

Fig 1 True stress versus true strain curves obtained under uniaxial compression for the rubber-toughened Epoxy adhesive and for the untoughened resin without rubber. Stresses $\sigma_{Ct}$ and $\sigma_{Cu}$ were used in determining the value of the parameter $k$.

Fig 2 Comparison of true stress/true plastic strain curves under compression and tension, respectively, with the effective stress ($\sigma_0$) versus effective plastic strain ($\varepsilon^p_0$) curve obtained from shear data. Equivalent coordinates ($\sigma_C$, $\varepsilon^C_\varepsilon$) and ($\sigma_0$, $\varepsilon^p_0$) were used in determining the value of $\mu$, and the coordinates ($\sigma_0$, $\varepsilon^p_0$) and ($\sigma_T$, $\varepsilon^p_T$) were employed in evaluating the parameter $q$. The continuous line through the $\sigma_0$ versus $\varepsilon^p_0$ data illustrates the fit obtained using equation (21) with the parameters in Table 1.

Fig 3 Calculated variation of $f_{\text{nucl}}$ with volumetric strain $\varepsilon_V$ giving the best fit to tensile data using the Gaussian and stretched-exponential functions respectively. Relevant parameters are included in Table 1.

Fig 4 Sections of yield surfaces represented by plots of the Mises yield stress $\sigma_e$ versus the mean normal stress $\sigma_k$. (■), derived from measured stresses under compression, shear and tension, respectively, for an effective matrix plastic strain $\varepsilon^p_0$ of 0.03 (see Fig 2), and from calculated stresses in butt-tension (see section 4). Continuous lines were calculated using the yield criterion (4) with values of $v_{R0}$, $k$, $\mu$ and $q_1$ given in Table 1.

Fig 5 Comparison of measured (○) and predicted (→) stress/strain curves under uniaxial compression.
Fig 6 Comparison of measured (■) stress/strain curves in uniaxial tension with predicted curves using the Gaussian (----) and stretched-exponential (—) void nucleation functions respectively.

Fig 7 Comparison of the measured (■) dependence of Poisson’s ratio on tensile strain with predicted variations using the Gaussian (---) and stretched-exponential (―) void nucleation functions, respectively.

Fig 8 Comparison of the measured (■) dependence of volumetric strain $\varepsilon_V = \varepsilon_T (1-2\nu)$ on tensile strain $\varepsilon_T$ with predicted variations using the Gaussian (---) and stretched-exponential (―) void nucleation functions respectively.

Fig 9 Comparison of the measured (•) and predicted curves of $\sigma_{11}$ versus $\varepsilon_{11}$ for a butt-joint tension test using the Gaussian (---) and stretched-exponential (―) void nucleation functions respectively. Also shown are the predicted variations of lateral stress $\sigma_{22}$ against $\varepsilon_{11}$. 
Fig 1  True stress versus true strain curves obtained under uniaxial compression for the rubber-toughened Epoxy adhesive and for the untoughened resin without rubber. Stresses $\sigma_{Ct}$ and $\sigma_{Cu}$ were used in determining the value of the parameter $k$.

Fig 2  Comparison of true stress/true plastic strain curves under compression and tension, respectively, with the effective stress ($\sigma_0$) versus effective plastic strain ($\varepsilon_0^p$) curve obtained from shear data. Equivalent coordinates ($\sigma_C$, $\varepsilon_C^p$) and ($\sigma_0$, $\varepsilon_0^p$) were used in determining the value of $\mu$, and the coordinates ($\sigma_T$, $\varepsilon_T^p$) and ($\sigma_0$, $\varepsilon_0^p$) were employed in evaluating the parameter $q$. The continuous line through the $\sigma_0$ versus $\varepsilon_0^p$ data illustrates the fit obtained using equation (21) with the parameters in Table 1.
Fig 3  Calculated variation of $f_{\text{nucl}}$ with volumetric strain $\varepsilon_V$ giving the best fit to tensile data using the Gaussian and stretched-exponential functions respectively. Relevant parameters are included in Table 1.

Fig 4  Sections of yield surfaces represented by plots of the Mises yield stress $\sigma_e$ versus the mean normal stress $\sigma_k$. ( ), derived from measured stresses under compression, shear and tension, respectively, for an effective matrix plastic strain $\varepsilon_0^p$ of 0.03 (see Fig 2), and from calculated stresses in butt-tension (see section 4). Continuous lines were calculated using the yield criterion (4) with values of $v_{R0}$, $k$, $\mu$ and $q_1$ given in Table 1.
Fig 5  Comparison of measured (○) and predicted (→) stress/strain curves under uniaxial compression.

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Fig 7 Comparison of the measured (■) dependence of Poisson’s ratio on tensile strain with predicted variations using the Gaussian (---) and stretched-exponential (—) void nucleation functions, respectively.

Fig 8 Comparison of the measured (■) dependence of volumetric strain $\varepsilon_V = \varepsilon_T (1-2\nu)$ on tensile strain $\varepsilon_T$ with predicted variations using the Gaussian (---) and stretched-exponential (—) void nucleation functions respectively.
Fig 9  Comparison of the measured (*) and predicted curves of $\sigma_{11}$ versus $\varepsilon_{11}$ for a butt-joint tension test using the Gaussian (---) and stretched-exponential (—) void nucleation functions respectively. Also shown are the predicted variations of lateral stress $\sigma_{22}$ against $\varepsilon_{11}$. 

Rubber-toughened Epoxy  
Butt-tension