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# The Effect of Temperature on the Strength of Adhesively-Bonded Composite-Aluminium Joints\*

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Different materials have different coefficients of thermal expansion, which is a measure of the change in length for a given change in temperature. When different materials are combined structurally, as in a bonded joint, a temperature change leads to stresses being set up. These stresses are present even in an unloaded joint which has been cured at say 150°C and cooled to room temperature. Further stresses result from operations at even lower temperatures.

In addition to temperature-induced stresses, account also has to be taken of changes in adhesive properties. Low temperatures cause the adhesive to become more brittle (reduced strain to failure), while high temperatures cause the adhesive to become more ductile, but make it less strong and more liable to creep.

Theoretical predictions are made of the strength of a series of aluminium/CFRP joints using three different adhesives at 20°C and -55°C. Various failure criteria are used to show good correlation with experimental results.

KEY WORDS adhesive failure criteria; CFRP; composites; effective modulus method; strength predictions; temperature stresses; aluminium; fracture mechanics; continuum failure; structural adhesives.

#### 1. INTRODUCTION

Adams and Mallick<sup>1</sup> produced a model for predicting the stresses and strains in an adhesive lap joint which gave excellent agreement with finite element analyses. The model was based on a series of algebraic equations which allowed for bending, shearing and stretching of both the adherend and the adhesive. A numerical solution was used, owing to the complexity of the equations involved. The solution was based on an equilibrium finite element approach using a method suggested by Allman.<sup>2</sup> The method can also be applied to cases in which the adhesive has elastoplastic stress-strain properties.

The accuracy of the Adams and Mallick<sup>1</sup> stress analysis was judged by correlat-

<sup>\*</sup>Presented at the Fifteenth Annual Meeting of The Adhesion Society, Inc., Hilton Head Island, South Carolina, U.S.A., February 17–19, 1992. One of a Collection of papers honoring A. J. Kinloch, the recipient in February 1992 of *The Adhesion Society Award for Excellence in Adhesion Science, Sponsored by 3M*.

ing results with a full finite element analysis. Here, a more thorough validation is used by comparing predicted joint strengths with experimental measurements. To this end, single lap joints between CFRP/Aluminium adherends, bonded by three mechanically-contrasting adhesives, were tested to failure at two temperatures,  $+20^{\circ}$ C and  $-55^{\circ}$ C. The joint materials and temperatures are typical for aircraft structures.

The analytical theory<sup>1</sup> predicts stresses and strains, not joint strengths. A failure criterion is necessary to interpret these distributions for strength prediction. Unfortunately, there is no universal criterion which is applicable to all possible modes of failure. Instead, there exist several criteria; those which can be applied to the present theory are described in the next section. Strengths are then predicted according to each criterion and the results compared with measured values. From this study, conclusions are drawn regarding the accuracy of the method in predicting the various modes of failure.

#### 2. FAILURE CRITERIA

There are, generally speaking, two approaches to predicting failure in a structure. One method assumes that the material contains some inherent flaws or cracks and that failure occurs when they propagate. Such methods make use of fracture mechanics (FM) to predict crack initiation and thus failure. The alternative is to use continuum mechanics and to predict that failure will occur when some limiting local stress or strain is reached. In this work, the continuum mechanics approach is used, but it is appropriate to say something first about fracture mechanics.

#### **Fracture Mechanics**

There are two serious drawbacks in applying FM to joints. First, FM was initially developed to predict failure in brittle materials and later extended by Hutchinson<sup>3</sup> and by Rice and Rosengren<sup>4</sup> to elasto-plastic materials. In joints, the crack is considered to be at or near the adhesive-adherend interface. For brittle (and elastic) adhesives it is possible to apply FM to calculate the stress intensity (or energy) at the crack due to a load and then to relate this to a material fracture quantity. However, when the adhesive is elasto-plastic, the basic assumption of FM is undermined since the plastic region in front of the crack is not in a continuum but at, or very close to, a bi-material interface. Therefore, it is not *rigorously possible* to relate the work done by a given load to crack propagation. However, the adherend is often more than 20 times as stiff as the adhesive, so that much of the plastic deformation will be in the adhesive. This would explain the success Chen<sup>5</sup> and Groth,<sup>6</sup> among others, have had in predicting joint failure for elasto-plastic materials using FM.

The other drawback of FM, of greater concern here, is the need to consider a crack in the stress analysis. Since there is no such provision in the present model, an alternative approach is sought. In any case, the adhesive layer is usually an uncracked continuum. Continuum failure criteria, which are failure surfaces in three-dimensional stress or strain space, are an alternative.

#### **Continuum Failure Criteria**

In an adhesive joint, there are three distinct failure modes. Failure may occur at the interface (adhesive failure) or within the adhesive layer (cohesive failure). Alternatively, the adherend itself may fail. However, if surfaces are properly prepared, then adhesive failure will be seldom.<sup>7</sup> Adherend failure is also rare unless the material is a laminate, in which case\* transverse failure is possible. Therefore, in the present work, cohesive failure and laminate failure criteria only are considered.

The purpose of a failure criterion is to predict from the behaviour of materials in a simple tensile (or shear or compression) test when failure will occur under any condition of applied stress. By far the most debatable quantity obtained from the tensile test is failure. While maximum values of stress, strain or energy determined from the test give a good indication for brittle materials, there is a lack of understanding as to what these values mean in ductile materials. Some of the difficulty is due to a lack of knowledge of stress distribution in front of cracks and inclusions within the material. For this reason, there exist a number of failure criteria. The following criteria have been applied to the present theory:

(a) Stress—The three-dimensional state of stress at a point can be resolved not only to give the maximum principal or shear stress but also the strain energy or distortion energy. When considering yield in polymers, a modified von Mises criterion must be used (see Appendix I). This has been extended to give the following failure criterion:

$$\sigma_{\rm max} = (3J_2)^{1/2} \tag{1}$$

where  $\sigma_{max}$  is the maximum stress given by the uniaxial test,

$$J_2 = (1/2)\{(\sigma_x - J_1/3)^2 + (\sigma_y - J_1/3)^2 + (\sigma_z - J_1/3)^2 + (\tau_{xy})^2 + (\tau_{yz})^2 + (\tau_{yz})^2\}, \text{ and}$$
  
$$J_1 = \sigma_x + \sigma_y + \sigma_z$$

(b) **Strain**—Strain, too, can be resolved to give either maximum vectorial values or energy at a point. Since the von Mises criterion already gives an indication of the energy, it was decided to use a maximum principal tensile strain criterion:

$$\epsilon_{\max} = (\epsilon_x + \epsilon_y)/2 + [(\epsilon_x - \epsilon_y)^2/4 + \gamma^2/4]^{1/2}$$
<sup>(2)</sup>

where  $\epsilon_{max}$  is the maximum strain as determined from the uniaxial test.

(c) Plastic strain or work—In the incremental initial stress method employed here for elasto-plastic analysis, uniaxial plastic strain and work vectors are calculated. These may be related to the maximum uniaxial plastic strain and the area under the stress versus plastic strain curve, respectively. These quantities are illustrated graphically in Figure 1.

There should be no difference between these two criteria since a value of plastic strain corresponds to a unique amount of plastic work. Any disparity

<sup>\*</sup>In this paper, the transverse stresses in the laminate which may lead to failure are in the throughthickness direction. Where the term *transverse stress* is used, this means a stress normal to the fibre direction and acting through the thickness, rather than across the width of the laminate.



FIGURE 1 Derivation of the stress versus plastic strain curve from a uniaxial tensile stress-strain curve.

indicates an error in the numerical solution, probably caused by taking too large an increment of load.

(d) Maximum Stress Theory—Experimental observations suggest that failure in unidirectional composite adherends is due to through-thickness tensile stress. It was, therefore, decided to use the normal tensile stress at the interface in the Maximum Stress Failure Criterion for the composite adherend, and to assume that the through-thickness tensile ultimate strength of the laminate is equal to the transverse tensile strength of the material.

#### **Effective Modulus Method**

For their elasto-plastic solutions, Hart-Smith<sup>8</sup> and Grant and Taig<sup>9</sup> suggest a shear strain to failure criterion. As Hart-Smith states, this is, in fact, a maximum shear strain energy criterion. Other yield (and failure) criteria, such as von Mises, are also based on shear, or distortion, energy. If a maximum shear energy failure criterion is valid, then it is now shown that it is possible to estimate the strength of joints, even for those with elasto-plastic adhesives, by simple closed form solutions.

First, it is necessary to make a few definitions. In a three-dimensional principal stress system, the total strain energy, U, and shear strain energy,  $U^*$ , are defined as:

$$U = [\sigma_1^2 + \sigma_2^2 + \sigma_3^2 - 2\nu(\sigma_1\sigma_2 + \sigma_2\sigma_2 + \sigma_3\sigma_1)]/2E$$
(3)

$$U^* = [\sigma_1^2 + \sigma_2^2 + \sigma_3^2 - \nu(\sigma_1\sigma_2 + \sigma_2\sigma_3 + \sigma_3\sigma_1)]/6G$$
(4)

In a uniaxial test,  $\sigma_1 = \sigma$ ,  $\sigma_2 = 0$ ,  $\sigma_3 = 0$ , so

$$U = \frac{\sigma^2}{2E}$$
(5)

$$\mathbf{U}^* = \frac{\sigma^2}{6\mathbf{G}} \tag{6}$$

Eliminating  $\sigma$  from eqns (5) and (6) gives the following relationship between the two energies in a tensile test:

$$U^* = U\frac{2(1+\nu)}{3}$$
(7)

The total strain energy, U, in a tensile test is simply the area under the stress-strain curve. It should be noted that eqn (7) is only approximate since the Poisson's ratio, assumed constant here, will vary and approach 0.5 when the material is plastic.

Consider now the behaviour of a material in shear (Fig. 2). Then, for a given shear strain  $\gamma_1$ , there can only be one corresponding value of shear stress,  $\tau_1$ . But  $\gamma_1$  also describes a unique shear strain energy, U<sup>\*</sup><sub>1</sub>. Thus, the maximum value of shear strain,  $\gamma_{max}$ , directly corresponds to the maximum value of shear strain energy.

The problem now reduces to one of predicting the shear strain within the adhesive accurately. If the material is ductile, then an elasto-plastic analysis is usually necessary. However, elastic solutions can, by suitable modification, be used to estimate the shear strains, even when the adhesive is plastic.

This is possible because the shape of the shear strain distributions in a lap joint



FIGURE 2 A shear stress-strain curve.



FIGURE 3 (a) Theoretical adhesives curves and the resulting single lap joint shear (b) stress and (c) strain distributions for a load of 8.5 kN.

is the same whether the adhesive stress-strain curve is elastic or elasto-plastic. Figure 3a shows three theoretical adhesive tensile stress-strain curves which describe an equal shear strain energy to failure. The shear stress and strain distributions, obtained from a finite element analysis of an aluminium-aluminium single lap joint, 12.7 mm long, 25.4. mm wide and using 1.62 mm thick adherends, are shown in Figures 3b and 3c.

It can be seen that while the maximum shear stress is dependent on the curve used, the *maximum* shear strains are all within 5% of each other. This result suggests that the maximum shear strain in a joint is dependent not on the shape of the stress-strain curve, but on the total shear strain energy described. Significantly, one of the adhesive stress-strain curves, A (Fig. 3a), is linear. Thus, maximum shear strains can be estimated with an *elastic* analysis using a linear curve which describes the same shear strain energy to failure,  $U^*$ , as the true tensile stress-strain curve.

The gradient of this linear curve, termed the Effective Young's Modulus ( $E_{eff}$ ), is given by the expression (see Fig. 4):

$$E_{\rm eff} = \frac{2U^*}{\epsilon_{\rm max}^2} \tag{8}$$

The Effective Shear Modulus  $(G_{eff})$  is given by the usual relationship:

$$G_{\rm eff} = \frac{E_{\rm eff}}{2(1+\nu)} \tag{9}$$

When employing the Effective Modulus Method (EMM),  $\gamma_{max}$  is to correspond to the maximum shear strain energy, U<sup>\*</sup>, then

 $\gamma_{max}^2 = \frac{2U^*}{E_{eff}}$ 

 $E_{eff} = \frac{2U^{\star}}{\epsilon_{max}^{2}}$ 

FIGURE 4 Definition of the Effective Young's Modulus.

(10)



FIGURE 5 (a) Stress-strain curves for theoretical adhesives A and B and resulting joint shear strains for a symmetrical aluminium/aluminium lap joint: (b) adhesive A and (c) adhesive B.

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Substituting eqn (8) into eqn (10) implies the following relationship between maximum shear and tensile strains:

$$\gamma_{\max} = \epsilon_{\max} [2(1+\upsilon)]^{1/2} \tag{11}$$

Thus, for typical adhesives ( $\nu \approx 0.3$  to 0.4) maximum shear strains should be 60% to 70% greater than tensile strains. Experimental evidence suggests that shear strains are even larger than this simple equation predicts. As stated earlier, one source of error may be the assumption of a constant Poisson's ratio even when the adhesive is plastic.

In any case, if the EMM is to work for lap joints, then elastic theories ought to predict values of shear strains that are close to those obtained by the more powerful non-linear FEM. In order to judge whether this is the case, results from FEM have been compared with the EMM (applied to Allman) for two theoretical adhesive curves which represent extremes of adhesive behaviour.

Curve A (Fig. 5a) represents a stiff, low-strain-to-failure adhesive, whereas curve B represents a material with low modulus and a high strain to failure. The FEA was conducted with a large displacement solution. The results are presented in Figures 5b and 5c. The peak strains are in close agreement, in fact within 3% for adhesive A and 5% for adhesive B. The success of this approach for strength prediction has been demonstrated by Mallick and Adams for balanced double<sup>10</sup> and single<sup>11</sup> lap joints. It remains to be seen whether the EMM is applicable to the unbalanced CFRP/Aluminium joint considered here.

#### 3. ADHESIVES AND ADHERENDS USED

Three structural adhesives were used in this investigation. Their properties are given in Tables I, II and III.

MY 750/HY 956 (Ciba) is a strong adhesive, typical of unmodified epoxies, and has a limited ductility even at 20°C. It was mixed 100:25 by weight.

MY 750/2005B (Ciba) is a moderately strong but reasonably ductile epoxy adhesive, mixed 100:50 by weight.

VOX 501 (Permabond) has a low modulus and high ductility at 20°C, but its ductility is very low at  $-55^{\circ}$ C, being slightly better than one, but slightly worse than the other, epoxy.

The aluminium alloy was a high-strength, unclad material to BS L164.

TABLE IL 164 aluminium alloy mechanical and physical properties(No significant difference between $+20^{\circ}C$ and $-55^{\circ}C$ )							
Young's modulus E (GPa)	Shear modulus G (GPa)	Proof stress (MPa)	Tensile strength (MPa)	Expansion coefficient (C <sup>-1</sup> )			
75	29	280	560	22×10 <sup>-6</sup>			

TABLE IIMechanical and physical properties of unidirectional CFRP using XAS fibres in 914 epoxy resin<br/>(No significant difference between +20°C and -55°C)

Long.	Trans.	Shear	Poisson	Poisson	Trans.	Expansion
modulus	modulus	modulus	ratio	ratio	strength	coefficient
E <sub>x</sub> (GPa)	E <sub>y</sub> (GPa)	G (GPa)	v <sub>xy</sub>	v <sub>yz</sub>	(MPa)	(C <sup>-1</sup> )
130	8.9	4.7	0.3	0.02	45	$7.5 \times 10^{-6}$

 TABLE III

 Mechanical and physical properties of the three adhesives used

Adhesive Type	MY 750/HY 956		MY 750/2005B		VOX 501	
Temperature (C)	+ 20	- 55	+ 20	- 55	+ 20	- 55
Young's Modulus (GPa)	3.18	5.62	2.48	4.6	0.49	0.9
Shear Modulus (GPa)	1.15	2.0	0.92	1.68	0.18	0.33
Maximum Stress (MPa)	86.2	45.2	52.0	62.4	23.1	15
Maximum Strain (%)	6.4	0.85	12.0	1.3	57	1.1
Maximum Plastic Strain (%)	3.8	_	9.9	_	52.3	_
Expansion Coeff. $(C^{-1})$	$56.7 \times 10^{-6}$	$56.7 \times 10^{-6}$	$54.6 \times 10^{-6}$	54.6×10 <sup>-6</sup>	$60.0  imes 10^{-6}$	$60.0 \times 10^{-6}$

The carbon fibre composite adherends were supplied ready-made by RAE (Farnborough). Courtaulds XAS fibre was impregnated with Ciba 914 epoxy resin to form Fibredux 914-CXAS 10K-5-34% prepreg. This was used in an autoclave with a ramp rate of 7°C/min to 175°C. At 80°C, a pressure of 340 MPa was applied. The material was held for 1 hour at 175°C and was post cured for 4 hours at 190°C.

The composite adherends were prepared prior to bonding by rubbing with fine emery paper until they passed the "water break" test. The aluminium was degreased and etched.

#### 4. FAILURE ANALYSIS

Single lap joints between aluminium and CFRP adherends (Fig. 6) were tested to failure in a Zwick screw-driven universal testing machine at  $+20^{\circ}$ C and  $-55^{\circ}$ C. Joints were tested at various overlap lengths and for three mechanically-contrasting adhesives cured at  $+60^{\circ}$ C. The adherend and adhesive material properties at these temperatures are given in Tables I, II and III. In the analysis, thermal stresses were considered by setting  $\Delta T =$ Cure (°C) – Operating (°C). This gave  $\Delta T = -40^{\circ}$ C and  $\Delta T = -115^{\circ}$ C for the room and low temperature cases, respectively. For strength prediction, a bilinear approximation to the adhesive elasto-plastic behaviour was used. The failure criteria were applied at every Gauss point in the adhesive except for the composite failure, in which case the peel (normal tensile) stress at the interface was calculated.

All dimensions in mm



FIGURE 6 Geometry of the single lap joints tested.

#### 4.1 MY 750/HY 956

The predicted and measured strengths for joints with the MY 750/HY 956 adhesive are compared in Figures 7a,b.

One noticeable feature is that the maximum strain criterion predicts slightly higher strengths than the maximum stress. The suggestion that points of maximum stress are not necessarily at maximum strain can be partly explained by the fact that the joint contains thermal strains. While the strains alone are not harmful, they give rise to stresses which are. Therefore, the stress criterion will generally predict lower strengths.

The predictions suggest that the composite is more likely to fail before the adhesive. In the case of the MY 750/HY 956 joint at room temperature, the aluminium is covered by composite, suggesting that failure might indeed have occurred initially in the composite.

The accuracy of the composite failure predictions is good except at the largest overlap. This is not surprising since, at large overlaps, the adherends will be slightly bent due to the thermal mismatch. Compounding this with the fact that the overlap can no longer be treated as rigid will undermine the basis of the edge bending moment calculations. In ignoring these detrimental effects, the theory overestimates the joint strength.

At the low temperature, the adhesive behaves in an elastic, brittle manner. Theoretical predictions were, therefore, based on an elastic analysis and are compared with experiment in Figure 7b. The theory predicts that failure will occur in the adhesive whereas the observed mode appeared to be a failure in the composite. However, it is difficult to be certain of the true failure mode from a study of the adherend surfaces since failure may have begun in the adhesive and progressed along the composite in an interlaminar fashion.



FIGURE 7 Predicted and experimental mean strength for joints with MY 750/HY 956 at 20°C (a) and  $-55^{\circ}$ C (b).

If it is assumed that failure occurs in the composite, then predicted strengths are too high. Therefore, the present analysis, in conjunction with the data available, is unable to provide a conclusive explanation of MY 750/HY 956 joint failures at low temperatures.

#### 4.2 MY 750/2005B

At room temperature, MY 750/2005B behaves in an elastic-perfectly plastic manner and can withstand a stress of 52 MPa. This is similar to the transverse strength of

the composite (45 MPa). Therefore, failure in either the adhesive or composite can be expected to occur.

The theoretical and experimental predictions are compared in Figure 8a. A maximum stress criterion is not included since the adhesive is perfectly plastic. As with MY 750/HY 956, the maximum strain criterion overestimates the strength slightly. The plastic field appears to be a better guide to adhesive failure since the predictions are very close to the experimental values. At most overlaps, however, there is little difference in the predictions between composite and adhesive failure.



FIGURE 8 Predicted and experimental mean strength for joints with MY 750/2005B at  $+20^{\circ}$ C (a) and  $-55^{\circ}$ C (b).

It was difficult to establish the mode of failure since there are traces of both composite and adhesive on the aluminium surface.

Since the failure may have been in the adhesive, strength predictions based on the EMM are included in Figure 8a. The predictions are within 15%, which suggests that the method can be used to estimate strength for this type of joint using this adhesive.

The strengths at  $-55^{\circ}$ C are compared in Figure 8b. From the failure surfaces, it was not possible to establish where failure began. The theory predicts failure in the adhesive, but at lower loads than were observed. This discrepancy probably arises from using the failure quantity measured in the bulk specimen. If composite failure is assumed, then the theoretical results are in better agreement with experiment. Even so, the predictions are underestimates, suggesting the theoretical stresses are too large. This can easily occur at singularity points in a linear analysis such as this, since the stress-relieving mechanism of plastic deformation is not allowed to occur.

#### 4.3 VOX 501

The room temperature predicted and measured strengths are given in Figure 9a. The theoretical stresses at the interface were too low to predict composite failure, even at high loads. This is not surprising considering the fact that the maximum stress that VOX 501 can sustain (23 MPa) is much lower than the composite transverse strength (45 MPa). Therefore, a composite failure criterion is not included in Figure 9a.

Adhesive failure based on maximum strain is once again found to be an overestimate. The remaining criteria, maximum stress and plastic strain/work give very similar strengths. These compare very well with experiment except at the large overlap. This is very encouraging since it shows that the theory is able to predict strength even when the adhesive exhibits an excessive amount of plasticity. Less accurate is the EMM which underestimates strength by up to 25%. Nevertheless, it does provide a reasonable prediction, even for this extremely ductile adhesive.

At  $-55^{\circ}$ C, the theoretical strengths, based on maximum stress, are lower than those observed, as shown in Figure 9b, whereas the predictions based on the maximum strain criterion agree well with experiment.

VOX 501 is an excellent example of the beneficial effect of ductility. At  $+20^{\circ}$ C it is able to withstand large amounts of deformation which results in high joint strengths. At  $-55^{\circ}$ C, however, the material supports a similar stress but hardly deforms before failure. The resulting joint strength is, therefore, significantly lower.

#### 5. DISCUSSION AND CONCLUSIONS

The present method can be used to predict strengths for elasto-plastic adhesives at room temperature, despite neglecting plasticity in the aluminium adherend. For most cases, predictions are within 5% of the experimental values. The main exception is at the 63.5 mm overlap which appears to be at the edge of the range of validity for the theory.



FIGURE 9 Predicted and experimental mean strength for joints with VOX 501 at 20°C (a) and  $-55^{\circ}$ C (b).

It is not possible to identify one criterion for all the modes of failure. At room temperature, the plastic work and the maximum stress criteria seem appropriate. However, at  $-55^{\circ}$ C, the material is brittle, so the plastic criterion is invalid, while the maximum stress criterion is less accurate than maximum strain.

Through the use of a few simple continuum failure criteria, it has been found that the present theory is capable of accurate joint strength predictions for elasto-plastic adhesives. When the adhesive behaviour is brittle, the predictions are less accurate. It has not been possible to establish whether this is due to a deficiency in the method, but it is suspected that this may be due to scatter in the mechanical property data. The percentage deviation of the fracture quantities at  $+20^{\circ}$ C was far less than at  $-55^{\circ}$ C. Finally, it was shown that adoption of a new approach, the Effective Modulus Method, enables joint strength to be estimated through a relatively simple linear analysis, even when the adhesive behaviour is non-linear.

#### **APPENDIX I**

#### Theory of Plasticity and the Elasto-Plastic Modulus

The mathematical theory of plasticity provides a relationship between stress and strain for a material which exhibits an elasto-plastic response. A material is said to deform plastically when it suffers irreversible strains which are not time dependent. Here, it is assumed that the material behaves in a *linear* elastic manner until it reaches a certain stress. This stress state,  $\{\sigma\}$ , is given by a yield criterion of the form

$$\mathbf{f}(\{\sigma\}) = 0 \tag{A.1}$$

where f is some function, usually defined in terms of the hydrostatic stress,  $J_1$ , and the deviatoric stress,  $J_2$ , where

$$J_1 = \sigma_x + \sigma_y + \sigma_z$$
  
$$J_2 = (1/2)\{(\sigma_x - J_1/3)^2 + (\sigma_y - J_1/3)^2 + (\sigma_z - J_1/3)^2 + (\tau_{xy})^2 + (\tau_{xz})^2 + (\tau_{yz})^2\}$$

For ductile materials, the von Mises yield function

1

$$f(\{\sigma\}) = (3J_2)^{1/2} - Y_T = 0$$
(A.2)

is often used.  $Y_T$  is the yield stress derived from a uniaxial tensile test on the material. For polymers, the yield in tension and compression are often different. This is accounted for in Raghava's criterion<sup>12</sup> by including the hydrostatic stress, J<sub>1</sub>:

$$\{J_1(S-1) + (J_1)^2(S-1)^2 + 12J_2S\}/2S = Y_T^2$$
(A.3)

where S is the ratio of the yield stress in compression to the yield stress in tension. Until initial yield, the strain  $\{\epsilon\}$  is related to the stress  $\{\sigma\}$  according to the elastic law

$$\{\boldsymbol{\epsilon}\} = [\mathbf{D}]^{-1}\{\boldsymbol{\sigma}\} \tag{A.4}$$

where [D] is the modulus matrix. After yielding, the total strain,  $\{\epsilon\}$ , will be partly elastic,  $\{\epsilon_e\}$ , and partly plastic  $\{\epsilon_p\}$ ;

$$\{\boldsymbol{\epsilon}\} = \{\boldsymbol{\epsilon}_{e}\} + \{\boldsymbol{\epsilon}_{p}\} \tag{A.5}$$

While the elastic part is given by eqn (A.4), the plastic strain is determined from the theory of plasticity. However, the theory provides a rule for the determination of plastic strain *increments*. This "flow" rule is of the form

$$d\{\boldsymbol{\epsilon}_{p}\} = \lambda(\partial f/\partial\{\boldsymbol{\sigma}\}) \tag{A.6}$$

where  $d{\epsilon_p}$  is the vector of plastic strain increments and  $\lambda$  is an instantaneous constant that can vary throughout loading. The flow rule ensures that the plastic

strain increment is normal to the yield surface defined by f and also that the stress remains within the surface  $(f({\sigma})<0)$ .

To accommodate the incremental nature of plasticity theory, increments of strain have to be considered. Rewriting eqn (A.5) in incremental form gives

$$d\{\epsilon\} = d\{\epsilon_e\} + d\{\epsilon_p\}$$
(A.7)

Substituting eqns (A.5) and (A.6) into eqn (A.7) and re-arranging, we get

$$d\{\sigma\} = [D]d\{\epsilon\} + [D]\{\alpha\}\lambda$$
 (A.8)

where  $\{\alpha\} = (\partial f / \partial \{\sigma\})$ . It can be shown that

$$[\mathbf{D}]\{\alpha\}\lambda = [\mathbf{D}_{\mathbf{p}}]\{\epsilon\}$$
(A.9)

where

$$[\mathbf{D}_{\mathsf{p}}] = \frac{[\mathbf{D}]\{\alpha\}\{\alpha\}^{\mathsf{T}}[\mathbf{D}]}{\mathbf{A} + \{\alpha\}^{\mathsf{T}}[\mathbf{D}]\{\alpha\}}$$
(A.10)

and A is equal to the instanteous gradient of the uniaxial stress-plastic strain curve. Therefore, we have the following relationship between stress and strain:

$$\mathbf{d}\{\boldsymbol{\sigma}\} = [\mathbf{D}_{ep}]\mathbf{d}\{\boldsymbol{\epsilon}\} \tag{A.11}$$

where  $[D_{ep}] = [D] - [D_p]$  and is known as the elasto-plastic modulus.

If the material strain *softens*, then A will be negative. This can lead to a situation where the stiffness matrix is not positive definite. Therefore, at this stage in the present work, only strain hardening has been considered.

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