A COUPLED ELASTIC-PLASTIC DAMAGE MODEL FOR RUBBER-MODIFIED EPOXY ADHESIVES

ULF EDLUND and ANDERS KLARBRING

Linköping Institute of Technology, Department of Mechanical Engineering, Division of Solid Mechanics and Strength of Materials, S-581 83 Linköping, Sweden

(Received 10 March 1992; in revised form 1 February 1993)

Abstract—A continuum damage model for rubber-modified epoxy adhesives is presented. The macro- and micromechanical observations are translated into a constitutive theory based on thermodynamics with internal state variables. An extension of the principle of strain equivalence, which makes a phenomenological derivation more systematic, is proposed. Important features of the material model are: (1) An isotropic damage measure is used. (2) The irreversible strain consists of a deviatoric part due to plastic flow and damage, and a dilatoric part due to damage. (3) The criteria for plastic flow and for damage growth are dependent on the hydrostatic stress. The model response in tension, compression and shear is studied. The model is likely to be valid for a wider class of polymeric materials.

1. INTRODUCTION

A body that is deformed generally undergoes irreversible and dissipative micromechanical processes. On the macromechanical level these processes lead to permanent deformations, loss of stiffness and heat generation. In many applications, however, only part of the deformation behaviour is of interest and therefore the effect of some processes can be excluded. In this paper, we are particularly interested in the form of a constitutive equation, which in addition to elastic-plastic behaviour also describes the fracture process in the material. The micromechanical process that ultimately leads to fracture through the formation of a macrocrack is related to initiation and growth of surface and volume discontinuities, i.e. growth of microcracks and microcavities. As a consequence the overall response of a damaged material volume is characterized by a softening behaviour. This local fracture process, which is referred to as material damage, was originally accounted for by Kachanov (1958) in a work concerning creep rupture in metals. Since then the damage process has been given a more general mathematical description in Continuum Damage Mechanics (CDM). A material model including damage reflects the physical processes leading to failure. This means that the corresponding load-bearing capacity of a body (i.e. the failure load) can be obtained without use of a postulated fracture criterion or the explicit assumptions needed in connection with a fracture mechanics approach. We regard this as an important improvement in the analysis process. Also from a computational point of view it seems natural to follow the material deterioration leading to the formation and propagation of a crack. Mathematically, the damage is accounted for through the introduction of a new field of scalar or tensorial character in the continuum description. Central questions in such a formulation are the definition of a damage measure, its impact on the mechanical behaviour and the form of an evolution law for the damage field. Within the framework of CDM, general formulations have been given to model creep damage [e.g. Hayhurst et al. (1984) and Krajcinovic (1983)], fatigue damage [e.g. Chrzanowski and Kolczuga (1980) and Marigo (1985)] and damage in brittle [see the comprehensive review article by Krajcinovic (1989)] and ductile materials [e.g. Ju (1989), Simo and Ju (1987a) and Benallal et al. (1988) in the case of small strains and Simo and Ju (1989) and Rousselier (1981) in the case of large strainsl.

Most theories published deal with the damage process in metallic materials. The aim of this work is to take into account the effects of damage on the constitutive behaviour of a polymeric adhesive material. The type of adhesive that we intend to model is an elastomer-

modified epoxy adhesive that is typically encountered in aerospace structural applications. This type of adhesive is also called a toughened epoxy adhesive due to the effect of the second phase (the rubber) added, and it is probably the adhesive material most frequently subjected to careful strength analyses. Although we have this particular material in mind, the description we derive is likely to be valid for a wider class of polymeric materials.

Material softening and its influence of the failure behaviour of adhesive joints have been investigated by Ottosen and Olsson (1988) and Gustafsson (1987) who established closed-form solutions for lap joints loaded in pure shear. An interesting observation can be made from the latter work: the load-carrying capacity of a joint mainly depends on the peak stress and on the fracture energy, i.e. the area under the stress-strain curve; the dependence of the shape of the curve seems to be less pronounced. A numerical treatment of adhesive joints using interface elements and a saw-tooth formed constitutive behaviour has been discussed by Stigh (1987).

The experimental determination of the complete response of a material is a difficult problem. To our knowledge a complete stress-strain curve for an epoxy adhesive is not available. In the case of wood adhesives, however, the softening behaviour has been determined by Wernersson and Gustafsson (1987) using a shear loaded specimen. See also Wernersson (1990) for a more detailed discussion. The possibility of determining the complete response of the adhesive indirectly through measurements of the deformation of a double cantilever beam has been discussed by Ungsuwarungsri and Knauss (1987).

When deriving a constitutive equation describing the macroscopic phenomena observed, two different paths can be followed: the macroscopic response is either derived from observations of the microscopic mechanisms by use of homogenization techniques, or a phenomenological standpoint is taken. In the latter approach the path goes in the opposite direction, i.e. the model is constructed mainly with the macromechanical observations in mind. This approach is the traditional one to deal with plastic and creep behaviour. In this paper we will derive a phenomenological model from a mathematical framework based on thermodynamical considerations where the micromechanical processes are accounted for by a set of internal variables (see Section 3). This means that we also use our qualitative knowledge of the micromechanical processes when we select the form of the theory. As a consequence the treatment of damage in our model becomes similar to the traditional treatment of plastic flow. Even though thermodynamical considerations are in a sense not necessary [see e.g. the model for fracture and slip of interfaces in cementitious composites derived by Stankowski et al. (1991)], it is believed that these considerations provide further insight into the model: the relation to other models is more easily established and generalizations follow more naturally.

2. PLASTIC FLOW AND FRACTURE OF RUBBER-MODIFIED EPOXY

The purpose of this section is to briefly summarize the characteristic macromechanical behaviour and the micromechanics of the fracture process in rubber-modified epoxy that motivates the form of the constitutive equation proposed. It was noted by Kinloch *et al.* (1983) that "there is considerable controversy surrounding the exact nature of the energy-dissipating deformations that occur in the vicinity of the tip of a stationary crack during loading or of a propagating crack...". This circumstance still prevails (Bascom and Hunston, 1989; Yee and Pearson, 1989). The mechanism for toughening and irreversible deformations advocated by Kinloch and co-workers seems, however, to be well-founded and is presented briefly in the following [see e.g. Kinloch and Young (1983) and Kinloch *et al.* (1983)].

Epoxy resins are widely employed as the basis for thermosetting adhesives and as the matrix material for fibre-composites. When cured it gives a material with high modulus and good performance at relatively high temperatures. Unmodified epoxies are relatively brittle materials with poor resistance to crack growth. A method to improve the fracture toughness is to incorporate a second phase of rubbery particles into the epoxy matrix. The cured material will then consist of small rubbery particles (typical size $1 \mu m$) dispersed in, and bonded to, the matrix of epoxy. Two interacting processes contribute to the increased

toughness. The first one is that the multiaxial state of stress at the crack tip causes failure and void formation in the rubber particles. The second, and most important, energy absorbing process is that of plastic shear flow in the matrix. Due to the stress concentrations associated with the small particles, extensive, but localized shear yielding takes place in the matrix. As a result, the fracture toughness of a rubber-modified epoxy can be 30 to 40 times that of the unmodified epoxy.

The micromechanical processes leading to initiation and propagation of a crack in a polymeric material appear not to be fully understood. It has been proposed that the crack is initiated in highly strained areas of intersecting shear bands. It also appears as if growth of a crack takes place through complicated processes due to extensive plastic flow ahead of the crack tip. The reader is referred to Kinloch and Young (1983) for a more detailed discussion in this matter. Furthermore, examinations of fracture surfaces have shown that [e.g. Kinloch et al. (1983), Yee and Pearson (1989) and Bascom and Hunston (1989)] the diameters of the cavities are larger than those of the undeformed cavities.

Plastic deformations in polymers can take place due to two different mechanisms: shear yielding (a deviatoric process) and craze yielding (i.e. formation of microcracks capable of transmitting loads across its faces; it is a dilatoric process) (Brown, 1986). In the type of polymer which we intend to model, a highly crosslinked polymer, craze yielding has not been observed in the matrix material [e.g. Kinloch et al. (1983) and Yee and Pearson (1989)] and consequently plastic flow takes place essentially at a constant volume. (Also, if craze yielding took place, it would be referred to as inelastic deformation due to damage in our model.) Furthermore, experiments [Bauwens (1970), Bowden and Jukes (1972) and Raghava et al. (1973)] have shown that the yield stress for polymers, unlike metallic materials, is dependent on the hydrostatic state of stress. Experimental results for a rubbermodified epoxy can be found in the paper by Sultan and McGarry (1973). The viscous behaviour of polymers is also well known. In the case of rubber-modified epoxy, experimental results are presented by Peretz and Weitsman (1982). In this paper we intend to deal primarily with the rate-independent, or short time behaviour. The model we derive allows, however, for a modification to include rate-dependency (see Remark 3.2).

From the observations made above we draw the following conclusions: (1) A pressure dependent yield criterion must be used for the undamaged material. Since we want to model plastic flow as a non-dilatoric process this implies that the flow law must be of nonassociated character. (2) It is obvious that a hydrostatic state of tensile stress promotes damage growth. This dependence must be considered by the evolution law for damage growth. (3) Since damage is associated with growth of microvoids and microcracks that probably do not close completely upon unloading, a model of damage should give a dilatoric contribution to the irreversible deformation. Also, the loss of bond between the small rubber particles and the matrix and the subsequent growth of the holes must be counted as damage. In addition we will make a number of assumptions. It will be assumed that the damage affects the material response isotropically. This implies that a scalar damage variable is sufficient. An anisotropic measure is probably more correct but tremendous experimental difficulties arise in connection with the calibration of such a material description to experimental results. In order to obtain as simple a model as possible it seems reasonable, as a first step, to adopt an isotropic damage measure. It will also be assumed that the undamaged material has a linear elastic range and that the material exhibits a hardening behaviour. For the sake of simplicity we will assume isotropic hardening.

3. A CONSTITUTIVE MODEL INCLUDING DAMAGE

3.1. Strain equivalence principle

The measure of damage and its influence on the mechanical behaviour is introduced through the concept of strain equivalence. If σ is the homogenized stress tensor (i.e. the stress tensor when the body is treated as a continuum), the effective stress tensor $\bar{\sigma}$ is obtained as

$$\bar{\boldsymbol{\sigma}} = \frac{1}{1 - d} \boldsymbol{\sigma},\tag{1}$$

where $d \in [0, 1)$ is a scalar that measures damage. The principle of strain equivalence now reads [see e.g. Lemaitre and Chaboche (1990)]:

"The constitutive law for the damaged material is given by the constitutive law for the undamaged material if the stress tensor is changed for the effective stress tensor."

The application of this principle does not fully specify a damage constitutive law since such a law also has to include an evolution law for the variable d. However, we propose here an extension of the principle which gives guidance in this respect. It is based on the following observations: (1) In the thermodynamic framework which we use, certain thermodynamic forces occur. (2) It is seen that not only the stress but all of these forces, except the one associated with d, naturally appear in both effective and homogenized form; the one associated with d has only the local, or effective character. It seems natural to treat all forces on an equal basis and, thus, we propose the following extended principle of strain equivalence:

"The constitutive law should be formulated in terms of effective thermodynamic forces, and the constitutive law for the undamaged material should be recovered from that of the damaged by setting $\dot{d} = 0$ and changing the effective thermodynamic forces back to the homogenized ones."

3.2. A damage model derived as a Generalized Standard Material

If the concept of strain equivalence is the first cornerstone of our theory, the second one is the structure brought by the concept of generalized standard material, to be described in a restricted setting below. Firstly, the total strain tensor ε is split into the reversible (elastic) part ε^c and the irreversible one ε^{ir} :

$$\varepsilon = \varepsilon^{c} + \varepsilon^{rr}. \tag{2}$$

Next, the state variables of the theory have to be decided on: as discussed earlier, the undamaged material is taken as linear elastic-plastic with isotropic hardening and the hardening is represented by a scalar internal variable p. As a further internal variable we take the damage measure d. A choice of the homogenized free energy which leads to an identification of effective and homogenized forces is:

$$\Psi(\mathbf{\varepsilon}^{c}, p, d) = (1 - d)\Psi^{0}(\mathbf{\varepsilon}^{c}, p). \tag{3}$$

The free energy for the undamaged material is taken as

$$\Psi^{0}(\boldsymbol{\varepsilon}^{c}, p) = \frac{1}{2}\boldsymbol{\varepsilon}^{c} : \mathbf{E} : \boldsymbol{\varepsilon}^{c} + h(p), \tag{4}$$

where the function h represents the hardening behaviour and \mathbf{E} is the fourth-order elasticity tensor. The form of h is left unspecified at this stage.

Once the internal variables are specified, consequences of Clausius-Duhem's inequality are obtained by Coleman's method [Coleman and Noll (1963) and Coleman and Gurtin (1967)]. They are, the state law

$$\sigma = \frac{\partial \Psi}{\partial s^{c}} = (1 - d)\mathbf{E} : \mathbf{\varepsilon}^{c}$$
 (5)

and the reduced dissipation inequality

$$D = \boldsymbol{\sigma} : \dot{\boldsymbol{\varepsilon}}^{\text{ir}} - R\dot{p} - Y\dot{d} \geqslant 0, \tag{6}$$

where, by definition,

Continuum damage model

$$R = \frac{\partial \Psi}{\partial p} = (1 - d) \frac{\mathrm{d}h}{\mathrm{d}p},\tag{7}$$

2697

$$Y = \frac{\partial \Psi}{\partial d} = -\Psi^0(\mathbf{\epsilon}^c, p), \tag{8}$$

and a superposed dot denotes time derivative. Note that (5) implies that $\bar{\sigma} = \mathbf{E} : \varepsilon^{\epsilon}$ and (7) shows that R has a homogenized character: the corresponding effective thermodynamic force is R/(1-d). As pointed out earlier, Y has a naturally effective character. This thermodynamic force is called the damage energy release rate and can be thought of as a "damage driving" force.

The inequality (6) should be satisfied for all processes. An elegant way of ensuring this, which stems from the work of Moreau (1974), Ziegler (1983) and Onsager (1931a, b), is to assume the existence of a potential

$$\phi = \phi(\sigma, R, Y),\tag{9}$$

which is convex but generally non-differentiable. If

$$\phi(\mathbf{0}, 0, 0) = 0, \qquad \mathbf{0} \in \partial \phi(\mathbf{0}, 0, 0) \tag{10}$$

and

$$(\dot{\mathbf{z}}^{ir}, -\dot{p}, -\dot{d}) \in \partial \phi(\boldsymbol{\sigma}, R, Y)$$
 (11)

then it is seen (Moreau, 1974) that (6) is always satisfied. Here, $\partial \phi$ denotes the subdifferential of ϕ (see the Appendix). Equation (11) represents a generalized normality principle and materials which are specified in this way by a free energy Ψ and a potential ϕ (actually the dual of a dissipation potential) are called Generalized Standard Materials (GSM) (Halphen and Nguyen, 1975).

The class of materials covered by the above analysis can be slightly extended, without conflicting with (6), by including the state variables as parameters in ϕ , i.e.

$$\phi = \phi(\sigma, R, Y; \varepsilon^{e}, p, d). \tag{12}$$

The subdifferentiation is still taken with respect to the first three arguments. The possibility of this extension is utilized in this work and it is needed if we are to comply with our modified strain equivalence principle.

In this work rate-independent materials will be primarily considered. This implies that ϕ should be an indicator function (see the Appendix) of a closed convex set C, i.e.

$$\phi = I_C. \tag{13}$$

The convex set will be specified by functions

$$f_1 = f_1(\sigma^{D}/(1-d), R/(1-d); \varepsilon^{c}),$$
 (14)

$$f_2 = f_2(\sigma_{\rm m}/(1-d), Y; p, d),$$
 (15)

so that

$$C = \{ (\sigma, R, Y) \mid f_1 \le 0, f_2 \le 0 \}.$$
 (16)

Here, the stress tensor is split into its deviatoric and spherical parts σ^D and $\sigma_m I$, respectively, i.e.

$$\boldsymbol{\sigma} = \boldsymbol{\sigma}^{\mathrm{D}} + \sigma_{\mathrm{m}} \mathbf{I},\tag{17}$$

where I is the identity tensor. We see that C can be interpreted as a permitted domain for the thermodynamic forces. The coupled functions f_1 and f_2 can be interpreted as a flow surface and a damage surface, respectively. To be more specific, we will use here a modification of the von Mises yield criterion. By including $\sigma_m/(1-d) = \frac{1}{3} \text{tr}(\mathbf{E} : \mathbf{e}^c)$ as a parameter in f_1 , the yield stress becomes dependent on the spherical part of the stress tensor as has been experimentally observed for polymers. However, by expressing this parameter in terms of \mathbf{e}^c , the flow law becomes non-associated in character, and plastic flow becomes deviatoric, and consequently, volume preserving, as is also experimentally observed for polymers. The modification of the yield stress due to hardening is represented by the effective thermodynamic force R/(1-d). Thus

$$f_1 = J_2 \left(\frac{\boldsymbol{\sigma}^{\mathrm{D}}}{1 - d} \right) - g_1 \left(\frac{1}{3} \text{tr}(\mathbf{E} : \boldsymbol{\varepsilon}^{\mathrm{c}}) \right) - \frac{R}{1 - d}, \tag{18}$$

where $J_2(\boldsymbol{\sigma}^D) = (\frac{3}{2}\boldsymbol{\sigma}^D : \boldsymbol{\sigma}^D)^{4/2}$ is the second invariant of the stress deviator and the function g_1 represents the initial yield stress.

For f_2 we simply take it as a linear function of Y and introduce a dependence on spherical stress through a function g_2 . The presence of this function introduces an irreversible dilatational strain. Furthermore, a function κ is introduced which describes how the threshold for damage growth decreases as the material properties deteriorate with plastic strain and also how it increases with further damage. Thus

$$f_2 = -Y + g_2 \left(\frac{\sigma_{\rm m}}{1 - d}\right) - \kappa(p, d). \tag{19}$$

In this paper we will consider a case when the desired form of g_2 makes f_2 non-differentiable. Therefore, we generalize the description of the domain $f_2 \le 0$ by using two differentiable functions, f_{21} and f_{22} , so that

$$C = \{ (\boldsymbol{\sigma}, R, Y) \mid f_1 \leq 0, f_{21} \leq 0, f_{22} \leq 0 \}, \tag{20}$$

where

$$f_{21} = -Y - \kappa(p, d) \tag{21}$$

and

$$f_{22} = -Y + g_2 \left(\frac{\sigma_{\rm m}}{1 - d}\right) - \kappa(p, d). \tag{22}$$

This means that we express the original damage surface by two damage surfaces that intersect in a non-smooth fashion.

From (A4) of the Appendix it is now clear that the complementary constitutive law (11) corresponding to this choice of C becomes

$$\dot{\boldsymbol{\varepsilon}}^{ir} = \dot{\lambda} \frac{\partial f_1}{\partial \boldsymbol{\sigma}^{D}} : \frac{\partial \boldsymbol{\sigma}^{D}}{\partial \boldsymbol{\sigma}} + \dot{\mu}_2 \frac{\partial f_{22}}{\partial \sigma_{m}} \frac{\partial \sigma_{m}}{\partial \boldsymbol{\sigma}} = \frac{\dot{\lambda}}{1 - d} \frac{3}{2} \frac{\boldsymbol{\sigma}^{D}}{J_2(\boldsymbol{\sigma}^{D})} + \frac{\dot{\mu}_2}{1 - d} g_2' \left(\frac{\sigma_{m}}{1 - d}\right) \frac{1}{3} \mathbf{I}, \tag{23}$$

$$-\dot{p} = \lambda \frac{\partial f_1}{\partial R} = -\frac{\dot{\lambda}}{1 - d},\tag{24}$$

$$-\dot{d} = \dot{\mu}_1 \frac{\partial f_{21}}{\partial Y} + \dot{\mu}_2 \frac{\partial f_{22}}{\partial Y} = -(\dot{\mu}_1 + \dot{\mu}_2), \tag{25}$$

$$f_1 \leqslant 0, \qquad \hat{\lambda} \geqslant 0, \qquad f_1 \hat{\lambda} = 0,$$
 (26)

$$f_{21} \le 0, \qquad \dot{\mu}_1 \ge 0, \qquad f_{21} \dot{\mu}_1 = 0,$$
 (27)

$$f_{22} \le 0, \qquad \dot{\mu}_2 \ge 0, \qquad f_{22}\dot{\mu}_2 = 0,$$
 (28)

where g'(x) = dg/dx. Note that our multisurface description of C leads to a form of the complementary law that resembles that proposed by Koiter [see e.g. Koiter (1960)] for multisurface plasticity. Also note that $\lambda/(1-d)$ can be substituted for a new multiplier, say $\bar{\lambda}$, so that when damage is not present, i.e. $\dot{\mu}_1 = \dot{\mu}_2 = 0$, the constitutive law becomes independent of d. Since $\sigma^D/J_2(\sigma^D)$ is of order zero in σ^D , this implies that the strain equivalence principle is satisfied. From (23) it is seen that the irreversible part of the strain consists of a deviatoric part due to plastic flow and damage and a spherical part due to damage. A standard calculation shows that the deviatoric effective plastic strain can be identified with the internal variable p, i.e.

$$\dot{p} = (\frac{2}{3}\dot{\mathbf{e}}^{\text{irD}} : \dot{\mathbf{e}}^{\text{irD}})^{1/2} = \frac{\dot{\lambda}}{1 - d}.$$
 (29)

Next, we will investigate the dissipation in our model. From (6) and (23)–(25) we obtain

$$D = \lambda \left(J_2 \left(\frac{\boldsymbol{\sigma}^{\mathrm{D}}}{1 - d} \right) - \frac{R}{1 - d} \right) + \dot{\mu}_1 (-Y) + \dot{\mu}_2 \left(\frac{\sigma_{\mathrm{m}}}{1 - d} g_2' \left(\frac{\sigma_{\mathrm{m}}}{1 - d} \right) - Y \right)$$
(30)

and since $f_1 = f_{22} = 0$ when $\lambda > 0$ and $\mu_2 > 0$, we find in this case that

$$D = \lambda g_1(\frac{1}{3} \operatorname{tr}(\mathbf{E} : \boldsymbol{\varepsilon}^{\mathrm{e}})) + \dot{\mu}_1(-Y) + \dot{\mu}_2\left(\frac{\sigma_{\mathrm{m}}}{1-d}g_2'\left(\frac{\sigma_{\mathrm{m}}}{1-d}\right) - g_2\left(\frac{\sigma_{\mathrm{m}}}{1-d}\right) + \kappa(p,d)\right)$$
(31)

which shows that we must have $g_1 \ge 0$ and a sufficient condition for (6) to hold is

$$g_2'\left(\frac{\sigma_{\rm m}}{1-d}\right)\frac{\sigma_{\rm m}}{1-d} - g_2\left(\frac{\sigma_{\rm m}}{1-d}\right) \geqslant 0$$
 and $\kappa \geqslant 0$.

Remark 3.1. It is of interest to make some remarks on the relation of the present model to the similar one presented by Ju (1989). [See also Simo and Ju (1987a).] First of all it should be noted that Ju's model is not derived within the framework of GSM. Furthermore, an important difference, as compared to our model, is that Ju aims at modelling only microcracks which do not result in a volume change. Thus, the function g_2 is constant in that work. Another important point when comparing with Ju's work is that we take κ to depend on plasticity (through p). Ju seems to attempt to introduce the influence of plasticity on damage through the generalized force Y only. That is, the example of h(p) that he gives is $h(p) = R_0 p + \frac{1}{2} \theta p^2$, where R_0 is said to denote the initial yield radius and θ the hardening slope. However, such a free energy implies that no plastic work is dissipated. The present framework allows for the construction of a mechanically equivalent but thermodynamically more sound theory.

Remark 3.2. In our constitutive model we did not take into account the viscous behaviour of the material. The present model allows, however, for a modification in this direction. Mathematically, this can be accomplished by a regularization of the non-differentiable dissipation potential ϕ , i.e. a regularization of the indicator function I_C . For example, if

$$C = \{ \mathbf{x} \mid f_i(\mathbf{x}) \leq 0, i = 1, 2, ..., n \}$$

then a possible regularized form $\Omega(\mathbf{x})$ or $I_C(\mathbf{x})$ is defined by:

$$\Omega(\mathbf{x}) = \sum_{i=-1}^{n} \overline{\Omega}_{i}(\mathbf{x}),$$

$$\overline{\Omega}_{i}(\mathbf{x}) = \omega(f_{i}(\mathbf{x})),$$

where

$$\omega(f) = \begin{cases} \frac{K}{n+1} \binom{f}{K}^{n+1} & f > 0, \\ 0 & f \leq 0. \end{cases}$$

and K and n are material constants. The possibility of performing only a "partial" regularization leading to e.g. viscous plastic and a non-viscous damage behaviour, is also interesting. In that case ϕ becomes the sum of a differentiable and a non-differentiable part.

4. MODEL RESPONSE

The purpose of this section is to show the typical material response of our rate-independent constitutive model and how variations of the material parameters affect the response for the following choice of the yet unspecified functions h, g_1 , g_2 and κ :

$$h(p) = \frac{1}{2}\theta p^{2},$$

$$g_{+}(x) = k - \xi x,$$

$$g_{2}(x) = \eta x,$$

$$\kappa(p, d) = l - \alpha p + \beta d.$$

It is easily verified that g_2 fulfills the sufficient requirement for non-negative dissipation. The functions f_1 , f_{21} and f_{22} are now fully specified. In Figs 1 and 2 the flow surface and the damage surface are shown. The material constants introduced have the following interpretations: θ is a plastic hardening constant; k is the initial yield stress; ξ represents the yield stress dependence on the hydrostatic state of stress; l is the initial damage threshold; α and β describe how the damage threshold is affected by plastic flow and damage; the constant η is the damage growth sensitivity due to a hydrostatic state of stress. Furthermore, we assume that the initial reversible (elastic) response is isotropic. We let E and ν denote Young's modulus and Poisson's ratio, respectively.

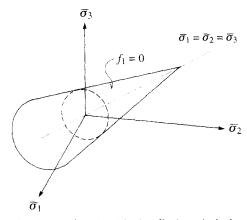


Fig. 1. The flow surface $f_1 = 0$ in the effective principal stress space.

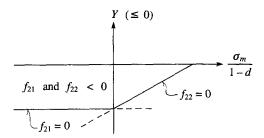


Fig. 2. The damage surface $f_{21} = 0$, $f_{22} = 0$.

We are primarily interested in the material response when it is used as a thin adhesive layer between two stiff bodies. Therefore we will investigate the stress-strain relation of the canonical deformation modes (see Fig. 3) of such a layer, i.e. uniaxial straining with no contraction (only $\varepsilon_{11} \neq 0$) and pure shear strain (only $\varepsilon_{12} \neq 0$).

Our material description involves nine parameters $(E, v, \xi, k, \theta, \eta, l, \alpha)$ and β) which have to be determined from experiments. The most convenient situation is of course if the constitutive law can be integrated to give closed-form expressions of the material response for some simple loading situations. When the corresponding experimental results are available the material parameters can be calculated so that the constitutive response fits the experimental result approximately. Unfortunately, it was found that it is practically impossible to obtain closed-form expressions even for the simple loading cases mentioned above. Instead, the constitutive equation has to be integrated numerically. We have chosen to make use of the "operator splitting" methodology originally proposed for elastoplasticity by Simo and Ortiz (1985). We have used the three-step operator split algorithm for rate independent problems involving damage evolution proposed by Ju (1989). See also Simo and Ju (1987b) for a discussion of this type of algorithms in connection with damage evolution problems.

As a consequence of the need for a numerical integration of the constitutive equation, numerical techniques must also be used to fit the response to experimental data. A discussion of the various optimization algorithms available can be found in Häggblad (1985). The

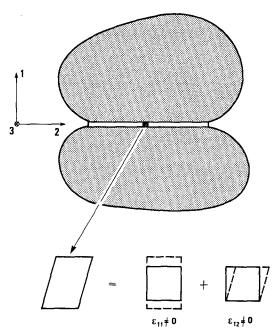


Fig. 3. The canonical deformation modes (only $\varepsilon_{11} \neq 0$, only $\varepsilon_{12} \neq 0$) of a thin adhesive layer joining to stiff bodies.

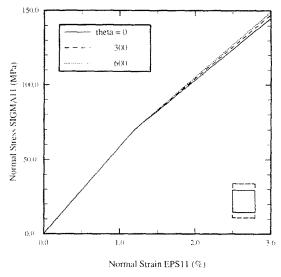


Fig. 4. Response in tension when varying θ . See Table 1 for input data.

optimization algorithms have in common that a rough estimate of the parameters are necessary as starting values for the optimization procedure. In Figs 4–13, the influence of the different material parameters on the response in tension, compression and shear is systematically examined. Finally, a typical response in a tension–compression test and the corresponding damage evolution is shown in Fig. 14. The input data corresponding to each figure is shown in Table 1. This clarifies the role of the parameters so that reasonable input data for an optimization procedure can be established.

5, CONCLUSIONS AND DISCUSSION

A constitutive model, which in addition to elastic-plastic behaviour also describes the effects of material deterioration, has been derived using a phenomenological approach. It rests on the mathematical structure brought by GSM and the principle of strain equivalence. We have proposed an extension of this principle that makes the reasoning more coherent. The most important features of the model are: (1) An isotropic damage measure is used. (2) The irreversible strain consists of a deviatoric part due to plastic flow and damage, and

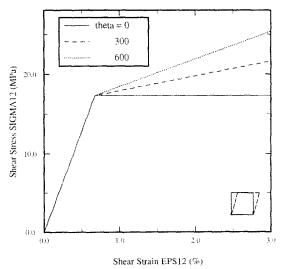


Fig. 5. Response in shear when varying θ . See Table 1 for input data.

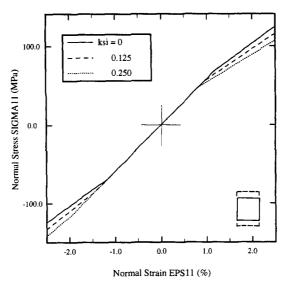


Fig. 6. Response in tension and compression when varying ξ . See Table 1 for input data.

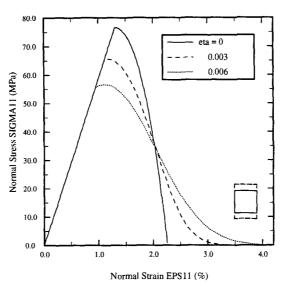


Fig. 7. Response in tension when varying η . See Table 1 for input data.

1 aut	1	mpt	ii ua	ia i	O PI	gs 4-	-12

Fig. No.	<i>θ</i> [MPa]	ξ [—]	η [—]	β [MPa]	α [MPa]	/ [MPa]	k [MPa]	Remark
4, 5	see Fig.	0	()	()	()	[]	30	$\dot{\mu}=0,t,s$
6	0	see Fig.	$\dot{}$	$\dot{}$	Ó	ÌÌ	30	$\dot{\mu}=0,t,c$
7	0	0	see Fig.	1.0	Ò	0.5	[]	$\lambda = 0, t$
8, 9	()	()	0	see Fig.	$\dot{}$	0.5	ίi	$\lambda = 0, t, s$
10, 11	0	0	0	0.5	see Fig.	0.5	30	t, s
12, 13	300	0.125	0.003	0.5	100	see Fig.	30	t, c, s
14	300	0.125	0.003	0.5	100	0.750	30	, . , .

^() means that output is independent of this parameter.

^[] means that the parameter has been given a large value ($\Rightarrow \lambda$ or $\dot{\mu} \equiv 0$).

t =tension, s =shear, c =compression.

E = 3500 MPa, v = 0.36.

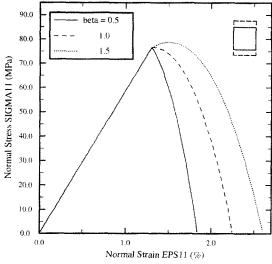


Fig. 8. Response in tension when varying β . See Table 1 for input data.

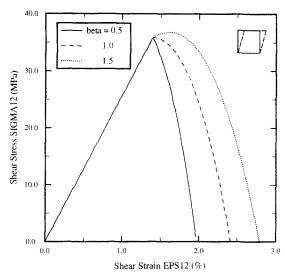


Fig. 9. Response in shear when varying β . See Table 1 for input data.

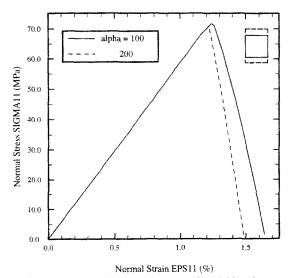


Fig. 10. Response in tension when varying α . See Table 1 for input data.

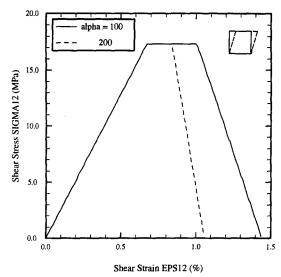


Fig. 11. Response in shear when varying α . See Table 1 for input data.

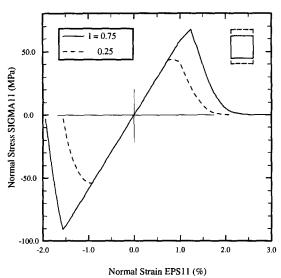


Fig. 12. Response in tension and compression when varying l. See Table 1 for input data.

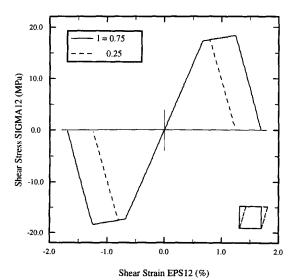


Fig. 13. Response in shear when varying I. See Table 1 for input data.

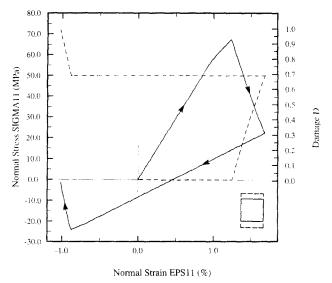


Fig. 14. Typical response in a tension-compression cycle (prescribed total strain) until material is completely damaged. See Table 1 for input data.

a dilatoric damage part. (3) The criterion for plastic flow and for damage growth is dependent on the hydrostatic stress. Note that, since the plastic-damage part of flow law is volume preserving, it is non-associated in character. The model is probably valid for a larger class of polymeric materials than the specific one we have in mind.

A constitutive model including damage is of interest in practical calculations because failure loads can be obtained without use of a postulated fracture criterion or fracture mechanics. Such methods are usually based on the stress or strain state obtained neglecting the effects of material damage. Also, typical assumptions in fracture mechanics as crack length, direction and location are avoided. Also, couplings to other fields in the continuum description can be taken into account. In a material description including damage, the fracture behaviour is inherent in the constitutive model. For instance, a domain with completely damaged material constitutes a crack.

We have aimed at deriving as simple a model as possible. Therefore an isotropic damage measure was adopted and the strains were assumed to be small. Some of the recently developed adhesives can probably not be treated under these assumptions. The generalization of a model of this kind to include large deformations is therefore an important point for future work. It should, however, be remembered that a formulation of a large strain theory for plastic flow (even without damage) still is a matter of controversy and no unified treatment seems to exist. Also, the need for experimental techniques and results to support a phenomenological derivation is obvious.

REFERENCES

Bascom, W. D. and Hunston, D. L. (1989). The fracture of epoxy and elastomer-modified epoxy polymers. In Treatise on Adhesion and Adhesives (Edited by R. L. Patrick), Vol. 6, pp. 123–185. Marcel Dekker, New York.
 Bauwens, J. C. (1970). Yield condition and propagation of Lüders' lines in tension torsion experiments on poly(vinyl chloride). J. Polymer Sci. 8, 893–901.

Benallal, A., Billardon, R. and Doghri, I. (1988). An integration algorithm and the corresponding consistent tangent operator for fully coupled elastoplastic and damage equations. *Comm. Appl. Num. Meth.* 4, 731–740. Bowden, P. B. and Jukes, J. A. (1972). The plastic flow of isotropic polymers. *J. Mater. Sci.* 7, 52–63.

Brown, N. (1986). Yield behaviour of polymers. In Failure of Plastics (Edited by W. Brostow and R. D. Corneliussen), pp. 98-118. Hanser.

Chrzanowski, M. and Kolczuga, M. (1980). Continuous damage mechanics applied to fatigue failure. Mech. Res. Commun. 7(1), 41-46.

Coleman, B. D. and Gurtin, M. E. (1967). Thermodynamics with internal state variables. *J. Chem. Phys.* **47**(2), 597–613.

Coleman, B. D. and Noll, W. (1963). The thermodynamics of elastic materials with heat conduction and viscosity. *Arch. Rat. Mech. Anal.* 13, 167-178.

- Gustafsson, P. J. (1987). Analysis of generalized Volkersen-joints in terms of non-linear fracture mechanics. In *Mechanical Behaviour of Adhesive Joints* (Edited by G. Verchery and A. H. Cardon), pp. 323–338. Edition Pluralis. Paris.
- Häggblad, H.-Å. (1985). Constitutive modelling of hard metal powder. Licentiate Thesis 1985: 003L, Division of Computer Aided Analyses and Design, Luleå University of Technology, Sweden.
- Halphen, B. and Nguyen, Q. S. (1975). Sur les matériaux standards genéralisés. J. Mécanique, 14(1), 39-63.
- Hayhurst, D. R., Dimmer, P. R. and Morrison, C. J. (1984). Development of continuum damage in the creep rupture of notched bars. *Phil. Trans. Roy. Soc. Lond.* 311, 103-129.
- Ju, J. W. (1989). On energy-based coupled elastoplastic damage theories: Constitutive modeling and computational aspects. Int. J. Solids Structures 25(7), 803-833.
- Kachanov, L. M. (1958). Time of the rupture process under creep conditions. Izv. Akad. Nauk. S.S.S.R., Otd. Tekh. Nauk. 8, 26-31.
- Kinloch, A. J., Shaw, S. J., Tod, D. A. and Hunston, D. L. (1983). Deformation and fracture behaviour of a rubber-toughened epoxy: I. Microstructure and fracture studies. *Polymer* 24, 1341-1354.
- Kinloch, A. J. and Young, R. J. (1983). Fracture Behaviour of Polymers. Elsevier Applied Science, Amsterdam.
- Koiter, W. T. (1960). General theorems for elastic-plastic solids. In *Progress in Solid Mechanics* (Edited by I. N. Sneddon and R. Hill), Vol. 1. North-Holland, Amsterdam.
- Krajcinovic, D. (1983). Creep of structures—a continuous damage mechanics approach. J. Struct. Mech. 11(1), 1--11.
- Krajcinovic, D. (1989). Damage mechanics. Mech. Mater. 8, 117-197.
- Lemaitre, J. and Chaboche, J.-L. (1990). *Mechanics of Solid Materials*. Cambridge University Press, Cambridge (translation of the 1985 French original edition).
- Marigo, J. J. (1985). Modelling of brittle and fatigue damage for elastic material by growth of microvoids. *Engng Fract. Mech.* **21**(4), 861–874.
- Moreau, J. J. (1974). On unilateral constraints, friction and plasticity. In *New Variational Techniques in Mathematical Physics* Coordinated by G. Capriz and G. Stampacchia), pp. 173–322. Edizioni Cremonese, Rome.
- Onsager, L. (1931a). Reciprocal relations in irreversible processes—I. Phys. Rev. 37, 405-426.
- Onsager, L. (1931b). Reciprocal relations in irreversible processes—II. *Phys. Rev.* **38**, 2265–2279. Ottosen, N. S. and Olsson, K.-G. (1988). Hardening/softening plastic analysis of an adhesive joint. *J. Engng*
- Ottosen, N. S. and Oisson, K.-G. (1988). Hardening/softening plastic analysis of an adhesive joint. J. Enging Mech. 114(1), 97–116.
- Peretz, D. and Weitsman, Y. (1982). Nonlinear viscoelastic characterization of FM-73 adhesive. *J. Rheology* **26**(3), 245–261.
- Raghava, R., Caddell, R. M. and Yeh, G. S. Y. (1973). The macroscopic yield behaviour of polymers. J. Mater. Sci. 8, 225-232.
- Rousselier, G. (1981). Finite deformation constitutive relations including ductile fracture damage. In *Three-Dimensional Constitutive Relations and Ductile Fracture* (Edited by S. Nemat-Nasser), pp. 331–335. North-Holland, Amsterdam.
- Simo, J. C. and Ju, J. W. (1987a). Strain- and stress-based continuum damage models—I. Formulation. *Int. J. Solids Structures* 23(7), 821–840.
- Simo, J. C. and Ju, J. W. (1987b). Strain- and stress-based continuum damage models—II. Computational aspects. *Int. J. Solids Structures* 23(7), 841–869.
- Simo, J. C. and Ju, J. W. (1989). On continuum damage-elastoplasticity at finite strains. Comput. Mech. 5, 375–400.
- Simo, J. C. and Ortiz, M. (1985). A unified approach to finite deformation elastoplastic analysis based on the use of hyperelastic constitutive equations. *Comput. Meth. Appl. Mech. Engng* 49, 221–245.
- Stankowski, T., Runesson, K. and Sture, S. (1991). Fracture and slip of interfaces in cementitious composites. *J. Engng Mech.* (submitted).
- Stigh, U. (1987). Initiation and growth of an interface crack. In *Mechanical Behaviour of Adhesive Joints* (Edited by G. Verchery and A. H. Cardon), pp. 237–248. Edition Pluralis, Paris.
- Sultan, J. N. and McGarry, F. J. (1973). Effect of rubber size on deformation mechanism in glassy epoxy. *Polymer Engng Sci.* 13(1), 29–34.
- van Tiel, J. (1984). Convex Analysis—An Introductory Text. Wiley, New York.
- Ungsuwarungsri, T. and Knauss, W. G. (1987). The role of damaged-softened material behaviour in the fracture of composites and adhesives. *Int. J. Frac.* 35, 221-241.
- Wernersson, H. (1990). Wood adhesive bonds—fracture softening properties in shear and in tension. Report TVSM-3012, Lund Institute of Technology, Division of Structural Mechanics, Sweden.
- Wernersson, H. and Gustafsson, P. J. (1987). The complete stress-slip curve of wood-adhesives in pure shear. In *Mechanical Behaviour of Adhesive Joints* (Edited by G. Verchery and A. H. Cardon), pp. 139–150. Edition Pluralis, Paris.
- Yee, A. F. and Pearson, R. A. (1989). Fractography and failure mechanisms of rubber modified epoxide resins. In *Fractography and Failure Mechanisms of Polymers and Composites* (Edited by A. C. Roulin-Moloney). Elsevier Science Publishers, Amsterdam.
- Ziegler, H. (1983). An Introduction to Thermomechanics. North-Holland, Amsterdam.

APPENDIX

This Appendix gives a short presentation of the concepts and results of convex analysis, essential for the developments in Section 3 of the paper. An introductory treatment of convex analysis is given by van Tiel (1984). Let $\Phi \colon \mathbf{R}^n \to \mathbf{R}$ be convex but generally non-differentiable. At a point $\mathbf{x} \in \mathbf{R}^n$, where Φ attains a finite value, the subdifferential $\partial \Phi(\mathbf{x})$ of Φ is the set of all $\mathbf{y} \in \mathbf{R}^n$ which satisfy

$$\Phi(z) - \Phi(x) \geqslant y \cdot (z - x) \,\forall \, z \in \mathbb{R}^n, \tag{A1}$$

where the dot denotes inner product.

The indicator function of a closed convex set $C \subset \mathbb{R}^n$ is the function

$$I_C(\mathbf{x}) = \begin{cases} 0 & \text{if } \mathbf{x} \in C, \\ \infty & \text{if } \mathbf{x} \notin C. \end{cases}$$
 (A2)

If the set C is given by

$$C = \{\mathbf{x} \in \mathbf{R}^n | f_i(\mathbf{x}) \le 0, \quad i = 1, 2, \dots, n\},\tag{A3}$$

where f_i are continuously differentiable real-valued convex functions, then

$$\partial I_{\mathcal{C}}(\mathbf{x}) = \{ \mathbf{y} \in \mathbf{R}^n \mid \mathbf{y} = \sum_{i=1}^n \lambda_i \nabla f_i(\mathbf{x}), \quad \lambda_i \geqslant 0, \quad f_i(\mathbf{x}) \geqslant 0, \quad \lambda_i f_i(\mathbf{x}) = 0, \quad i = 1, 2, \dots, n \},$$
(A4)

where ∇ is the gradient operator.