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Stress analysis near the tip of a curvilinear interfacial crack between a rigid spherical inclusion and a polymer matrix

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Craze and shear band formation at poorly adhering glass spheres in matrices of glassy polymers are known to be preceded by the formation of a curvilinear interfacial crack between sphere and matrix. In this study the axisymmetric finite element method has been used to analyse the stress situation near the tip of a curvilinear interfacial crack formed between a rigid spherical inclusion and a polymer matrix upon an applied uniaxial tension. Important factors that determine the stress state near the crack tip were found to be the crack length, the orientation of the crack tip with regard to the tension direction and the extent of interfacial slip between the inclusion and matrix. The results of the analyses were compared with the physical reality of craze and shear band formation at poorly adhering glass spheres. Reasonable agreement was found with respect to both the maximum interfacial crack length that can be reached until a craze or shear band forms at the crack tip and the planar orientation of craze growth perpendicular to the direction of the major principal stress.

1. Introduction

Rigid spherical inclusions in glassy polymers induce inhomogeneous stress fields and thus act as stress concentrators. Consequently, plastic deformation processes such as craze formation and shear band formation being at those inclusions. In recent studies the mechanisms for craze formation [1, 2] and shear band formation [3] were investigated at small glass spheres (diameter about 30 μm) embedded in matrices of, respectively, polystyrene (PS) and polycarbonate (PC) which were subjected to uniaxial tension. These microscopic studies have revealed the profound effect of the degree of interfacial adhesion on the mechanisms for craze and shear band formation. In the case of an excellently adhering glass sphere in a PS matrix the crazes were found to form near the poles of the sphere. From stress analysis around a completely bonded sphere it appeared that these are regions of maximum dilatation and of maximum principal stress. At an excellently adhering glass sphere in a PC matrix the shear bands were found to form near the surface of the sphere at an angle of 45° from the poles defined by the symmetry axis of the stressed sphere. These are regions of maximum principal shear stress and of maximum distortion strain energy density. In the case of poor interfacial adhesion between the glass sphere and polymer matrix, both craze and shear band formation were found to be preceded by dewetting along the interface between sphere and matrix. At dewetting a curvilinear interfacial crack is formed, starting at the pole and propagating along the interface in the direction of the equator until, at

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an angle of about 60° from the pole, a craze or shear band originates at the tip of this crack. As an example, Fig. 1 shows the craze pattern around a poorly adhering glass sphere in a PS matrix under uniaxial tension. The shadows at the poles of the sphere indicate that a pair of interfacial cracks has been formed.

The results of these studies prompted us to analyse the stress situation near the tip of a curvilinear interfacial crack between a glass sphere and a polymer matrix. The two-dimensional problem of a curvilinear interfacial crack between two further bonded dissimilar materials has been considered by several authors [4–6]. Their elastic analyses were based on the complex variable approach developed by Muskhelishvili. The obtained analytic solutions, however, are not appropriate for the present problem because the present problem is three-dimensional and, in physical reality, that part of the interface without interfacial crack is not perfectly bonded. Therefore, it is necessary to resort to numerical analyses and in this study the axisymmetric linear elastic finite element method has been used. This method is based upon assumptions for displacements which are defined in terms of polynomial functions over elements of finite size. Therefore, it is not possible to obtain an exact representation of the behaviour in the region of a singularity such as the tip of an interfacial crack. To overcome this difficulty, a finite element mesh has been used with a substantial refinement near the interface of sphere and matrix.

In the present paper two different kinds of problems are considered. Firstly, the problem of a partially unbonded–partially bonded sphere with the unbonded region of the interface increased stepwise from the pole to the equator. Secondly, the problem of a completely unbonded sphere with the extent of interfacial slip between sphere and matrix varied. With both problems the attention is focused on the stress state near the crack tip at the matrix side of the formed interfacial crack. The results of the second problem are compared with the physical reality of craze and shear band formation at poorly adhering glass spheres in matrices of glassy polymers.

2. Finite element analysis
2.1. Analysed system

As the principles of the axisymmetric linear elastic finite element method and the application of this method for spherically filled materials have been described in detail elsewhere [7, 8], only some main points will be briefly discussed here. The unit cell of the analysed system is shown in Fig. 2. The z-axis is the axis of revolution. The glass sphere occupies about 2 vol % and it has been checked that at this low percentage the analysed system represents the situation of an isolated sphere in an infinite matrix. The applied uniaxial tension was simulated by
stretching the system in the z-direction without restraints in the r-direction. The model of Fig. 2 was subdivided into triangular elements (TRIA3X3) as shown in Fig. 3. This was done automatically with a mesh generator named TRIQUAMESH [9]. Fig. 3 shows the substantial refinement of the mesh near the interface. A roughness function, determining the size of the elements, was kept constant along the entire interface to minimize the influence of the element size on the results under various conditions of crack length. The axisymmetric finite element analyses were executed using the FEMSYS finite element computer program written by Peters [10]. The elastic constants used are:

- Polymer matrix: Young's modulus 3 GPa, Poisson's ratio 0.35
- Glass sphere: Young's modulus 70 GPa, Poisson's ratio 0.22

The computer program calculates for each element the stresses \( \sigma_z \), \( \sigma_r \), \( \sigma_\theta \) and \( \tau_\theta \) acting at the centroid of the element. \( \sigma_\theta \) acts in the circumferential direction and is one of the three principal stresses since \( \tau_\theta \) and \( \tau_r \) are equal to zero.

### 2.2. Boundary conditions at the interface

The boundary conditions for relative displacements between the adjacent faces of the sphere and matrix are different for unbonded and bonded regions. Along unbonded regions of the interface relative displacements are permitted in both the normal and the tangential direction (n and t in Fig. 2), whereas along bonded regions relative displacements are not permitted at all. It is important to realize that a segment of an unbonded region may only be regarded as a part of the interfacial crack if the adjacent faces of the sphere and matrix open up upon application of the uniaxial tension. In that case the relative displacement of the matrix with respect to the sphere in the normal direction is positive, and the stress on that segment in the normal direction is zero. If, however, a segment of an unbonded region remains closed upon application of the uniaxial tension, it may not be regarded as a part of the interfacial crack. In that case there is no relative displacement of the matrix with respect to the sphere in the normal direction, and a normal stress which is negative (or zero) acts on that segment. Of course a relative displacement in the tangential direction (interfacial slip) still remains possible, in contrast to the case of a bonded region.

The extent of interfacial slip between unbonded adjacent faces of the sphere and matrix is dependent on the friction forces acting in the tangential direction. In the present analyses the friction force acting on a certain segment in the tangential direction, \( F_t \), is modelled as being proportional to the force acting on that segment in the normal direction, \( F_n \), by the proportionality constant \( \mu \). In formula:

\[
F_t = \mu F_n
\]

Accordingly, if a segment of an unbonded region opens up because of tension, no friction force acts on that segment since in that case \( F_t \) is equal to zero. If, however, a segment remains closed and \( F_n \) has a negative value, then a friction force proportional to \( F_n \) acts in the negative tangential direction opposing interfacial slip in the positive tangential direction.

### 3. Results and discussion

#### 3.1. Partially unbonded–partially bonded sphere

This section deals with the partially unbonded–partially bonded sphere. To study the effect of the length of an interfacial crack (at a further bonded sphere) on the stress state near the crack tip, the unbonded region of the interface was increased stepwise from the pole (\( \theta = 0^\circ \), completely bonded) to the equator (\( \theta = 90^\circ \),

![Figure 3 Finite element mesh (683 nodes, 1210 elements).](image-url)
completely unbonded). The unbonded region was assumed to be frictionless, thus $\mu = 0$.

From the analyses it appeared that up to a length of the unbonded region represented by $\theta = 64^\circ$, the entire unbonded region may be regarded as an interfacial crack. If, however, the unbonded region exceeds an angle $\theta$ of $64^\circ$, the adjacent faces of the sphere and matrix do not open up along the entire unbonded region but a part closest to the equator remains closed. This result can be understood from the contraction of the matrix perpendicular to the direction of the applied tension. This implies that the effect of the interfacial crack length on the stress state near the tip can only be studied up to a crack length represented by $\theta = 64^\circ$. In Fig. 4 the stresses in the matrix element having its centre closest to the interfacial crack tip have been plotted against the crack length. The stresses are presented as a ratio, $\sigma/T$ or $\tau/T$, where $T$ is the applied uniaxial tension. The stresses plotted at $\theta = 0$ are the stresses in the matrix element having its centre closest to the pole of a completely bonded sphere. Apart from $\sigma_\alpha$, $\sigma_\beta$, $\sigma_\gamma$, and $\tau_{rz}$, Fig. 4 shows the value of the ratio of the major principal stress, $\sigma_1$, which represents the maximum stress concentration. In this connection it must be noted that for all analysed systems with an interfacial crack, the maximum stress concentration always occurred in the matrix element nearest to the crack tip, thus in the element for which the stresses are given in Fig. 4.

From Fig. 4 it appears that the maximum stress concentration near the tip first increases with increasing curvilinear crack length, up to a value of $\theta$ of about $45^\circ$, and then decreases. Obviously it does not simply increase with increasing crack length as is the case with a rectilinear crack at the interface of two bonded dissimilar materials [11, 12]. This must be attributed to the fact that the angle between the direction of the applied tension and the normal of the crack plane near the tip increases with
increasing crack length which, of course, is unfavourable for high stress concentrations near the tip. Thus two factors opposing each other are introduced when the curvilinear crack length is varied, resulting in a maximum value of the maximum stress concentration at a crack length represented by \( \theta \) of about 45°.

3.2. Completely unbonded sphere with varied resistance to interfacial slip

This section deals with the completely unbonded sphere. The resistance to interfacial slip along the unbonded interface was varied by taking the value of \( \mu \) stepwise from 0 (frictionless) to 5.

From the analyses it appeared that the length of that part of the unbonded interface that may be regarded as an interfacial crack depends slightly on the value of \( \mu \). For values of \( \mu \) from 0 to 2 the adjacent faces of the sphere and matrix open up, up to \( \theta = 70° \) whereas the remaining part of the interface remains closed. For values of \( \mu \) from 3 to 5 the unbonded interface only opens up, up to \( \theta = 68° \), so the interfacial crack for-

3.3. Comparison of the results with the physical reality

3.3.1. Maximum interfacial crack length

When comparing the results of the completely unbonded sphere with the physical reality of craze and shear band formation at poorly adhering glass spheres, it must be realized that the analyses performed in this study do not take into account the thermal residual stresses set up around the glass sphere in consequence of the mismatch between the coefficients of thermal expansion (\( \alpha_{\text{polymer}} \) is about ten times \( \alpha_{\text{glass}} \)). Accordingly, in the present stress analyses the formation of the entire interfacial crack is represented to occur momentarily upon tension application. In practice, however, the formation of the interfacial crack is supposed to be a process, starting at the pole of the sphere because the radial thermal compressive stresses are first balanced at this location [1]. As the tensile test proceeds, the crack will propagate in the direction of the equator until a craze or shear band originates at the tip. The interfacial crack propagation is hindered by the radial thermal compressive stresses around the sphere and this will become more serious as the crack tip approaches the equator. The difference between the stress analysis situation and the physical reality must be the cause of the small difference between the computed and experimentally
Figure 6 The principal stresses near the tip of the curvilinear interfacial crack at a completely unbonded sphere as a function of the resistance to interfacial slip.

measured critical angle, $\theta$, that the interfacial crack can maximally reach until a craze or shear band forms at the tip. This angle was found experimentally to be about 60°, computed to be 68 to 70° depending on the value of $\mu$. In any case, it has clearly been demonstrated that in the case of a completely unbonded glass sphere the maximum stress concentration does not occur at the equator, as suggested by other authors [13, 14], simply because the interfacial crack tip cannot reach the equator.

3.3.2. Planar orientation of craze growth
An interesting feature is the planar orientation of craze growth. Areal craze growth in isotropic glassy polymers has been reported to occur along a path such that the major principal stress always acts perpendicular to the craze plane [15]. A craze that originates near the pole of an excellently adhering glass sphere in a PS matrix expands into the matrix in the direction perpendicular to the applied tension [1]. The directions of the major principal stress and of the applied tension indeed coincide near the pole of a completely bonded sphere. A craze that originates at the tip of the interfacial crack of a poorly adhering glass sphere, however, expands into the matrix in a direction deviating initially from the direction perpendicular to the applied tension as is clearly visible in Fig. 1. Only at some certain distance of the glass sphere, where the propagating craze tip leaves the “sphere of influence” of the glass sphere, the craze bends towards this direction. The initial deviation amounts to about 20°. Calculations of the direction of the major principal stress $\sigma_1$ in the matrix element nearest to the interfacial crack tip of a completely unbonded sphere revealed that the deviation between the direction of the major principal stress and the applied tension varies from 19° to 13° if $\mu$ is varied from 0 to 5. Bearing in mind the differences between the stress analysis situation and the physical reality, these values agree reasonably well with the experimental value of about 20°.

3.3.3. Criteria for craze formation and shear band formation
As known from previous studies [1, 3], at an excellently adhering glass sphere a craze forms near the pole of the sphere in the region of maximum dilatation, $\Delta$, and of maximum principal stress, $\sigma_1$. A shear band forms near the surface of the sphere at an angle of 45° from the pole in the region of maximum principal shear stress, $\tau_1$, and of maximum distortion strain.
energy density, $W_d$. The expressions of these elastic failure criteria in terms of the three principal stresses can be found elsewhere [3]. It is of practical interest whether craze and shear band formation occur easier (i.e. at a lower applied tensile stress level) at the tip of the interfacial crack of a poorly adhering sphere than at an excellently adhering sphere. Comparison of the maximum values of $\Delta$, $\sigma_1$, $\tau_1$ and $W_d$ at a completely bonded sphere with the values of these criteria near the interfacial crack tip of a completely unbonded sphere might provide an insight into this matter: if the values of the relevant criteria are clearly higher near the crack tip, a lower applied stress level required to start craze or shear band formation is to be expected.

The values of the four elastic failure criteria in the matrix element nearest to the interfacial crack tip of a completely unbonded sphere were calculated with the principal stresses given in Fig. 6. The results are plotted in Fig. 7 where the horizontal lines represent the maximum values of the criteria at a completely bonded sphere. From Fig. 7 it appears that the values of the criteria near the crack tip are not necessarily higher than at a completely bonded sphere but that this depends on the value of $\mu$. At $\mu = 0$, thus when the interface is frictionless, the criteria $\sigma_1$, $\tau_1$, and $W_d$ are of the same order of magnitude as at a completely bonded sphere. If $\mu$ is increased, thus if the extent of interfacial slip is reduced, the values of these criteria increase and become clearly higher compared with a bonded sphere. At $\mu = 0$, the value of the dilatation criterion $\Delta$, which represents the sum of the three principal stresses, is clearly lower compared with a bonded sphere and only becomes higher if $\mu$ exceeds a value of 1.5. This difference with the other three criteria can be understood by comparing the stress state near the interfacial crack tip with that near the pole of a completely bonded sphere. There the stress state is triaxial whereas near the crack tip the stress state is biaxial. So near the crack tip dilatation is to be produced by only two stresses and therefore, at low values of $\mu$, its value remains behind compared with the value of dilatation near the pole of a completely bonded sphere.

On the basis of the results presented in Fig. 7 it is rather difficult to determine if craze and shear band formation occur easier at the tip of the interfacial crack of a poorly adhering glass sphere than at an excellently adhering sphere. Apart from the fact that the exact formulations of the criteria for craze and shear band formation are unknown, it now appears that near the crack tip the values of the elastic failure criteria that rule craze and shear band formation are strongly determined by the extent of interfacial slip. This makes the extent of interfacial slip a very critical factor at comparing the values of the criteria near the crack tip with the values at a completely bonded sphere. In the present analyses the friction forces that oppose slip were
modelled as being proportional to the normal forces by the proportionality constant $\mu$. In practice, $\mu$ is known as the coefficient of static friction and practical values of $\mu$ between glass and glassy polymers have been reported to amount to about 0.5. However, in the physical reality of a poorly adhering glass sphere in a polymer matrix, the extent and character of the interfacial slip and the forces that oppose slip are not precisely known. It is uncertain if a value of 0.5 for a coefficient of static friction between glass and polymer has any physical relevance to this specific case. It is also uncertain if the modelling of the friction forces as being proportional to the normal forces is realistic. Thus, as the extent of interfacial slip appears to be so critical but is not precisely known, based on the present results definite conclusions on whether craze and shear band formation occur easier at the interfacial crack tip of a poorly adhering glass sphere than at an excellently adhering sphere cannot be made.

4. Conclusions

Application of the finite element method provides only approximate solutions in the region of a singularity. Nevertheless, the present study provides a good insight into the factors that determine the stress state near the tip of a curvilinear interfacial crack between a rigid spherical inclusion and a polymer matrix. It has been demonstrated that the maximum stress concentration near the tip of a curvilinear interfacial crack at a further bonded sphere does not simply increase with increasing crack length, but is also determined by the orientation of the crack tip with regard to the applied tension direction. The analyses for a completely unbonded sphere have shown that an interfacial crack at a completely unbonded sphere cannot become larger than a critical length represented by $\theta = 68$ to $70^\circ$. The stress state near the tip is biaxial and is strongly determined by the extent of interfacial slip along that part of the unbonded interface that remains closed. The values of the elastic failure criteria that rule craze formation and shear band formation increase substantially as the extent of interfacial slip reduces.

Comparison of the results of the completely unbonded sphere with the physical reality of craze and shear band formation at poorly adhering glass spheres has shown reasonable agreement with respect to the critical interfacial crack length that can maximally be reached until a craze or shear band forms at the tip. Also the experimental and calculated planar orientation of areal craze growth perpendicular to the direction of the major principal stress agree reasonably well. Definite conclusions on whether craze and shear band formation occur easier at the interfacial crack tip of a poorly adhering glass sphere than at an excellently adhering glass sphere could not be made because the extent and character of the interfacial slip between a poorly adhering glass sphere and a polymer matrix are not precisely known.

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