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The yielding of a two-dimensional void assembly in an organic glass

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This paper represents a first attempt to discuss the growth of groups of cavities in an isotropic polystyrene-like solid. Calculations are made on a two-dimensional array of cylindrical voids in a sheet of uniform thickness. Stresses and displacements throughout the solid are obtained by use of the finite element method for both uniaxial and biaxial loading conditions. Elastic perfectly-plastic and strain-softening-hardening material laws are considered. It is shown how groups of voids are much more easily formed than single separate voids. For a strain-softening-hardening constitutive law a craze formation mechanism is proposed and its stability examined.

1. Introduction

When an organic glass is subjected to a suitable level of stress, having a net hydrostatic tension [1], structures containing voids are formed and these are known as crazes. From the experimental point of view these processes have been extensively documented over the last 10 years. For example, Kambour [2] demonstrated the presence of very small rounded holes in a polycarbonate craze and then went on to demonstrate that cracking takes place by the splitting of a craze which is formed continuously at a crack tip [3]. More recently, evidence for expanded void structures has been found in fracture surfaces [4, 5] and electron micrographs have demonstrated craze structures in a wide range of glassy polymers [6-8].

Generally, these crazes start from the surface of a polymer where the existence of a stress-raising flaw or crack may be assumed. However, it has now been shown that this process can also take place internally within the body of a pure isotropic polymer glass [1, 9] and that this process can occur at stresses well below the yield stress of the polymer. Since the formation of voids must itself be a yield process the reason for this situation is not obvious.

Two approaches have been made to the solution of this problem. According to Gent [10] the glass transition temperature and the yield stress of the polymer are lowered by the occurrence of a hydrostatic tension in the polymer or at the tip of a crack. The difficulty with this proposal is that it appears to offer an alternative to the experimentally determined relation between yield stress and three-dimensional stress systems [1, 9, 11], which are not themselves adequate to predict the growth of a void in an isotropic solid [12]. The second approach suggests that voiding takes place only because the voids are produced in sheets or clusters. In this way, the formation of voids is made easier because the plastic yielding which takes place in the sphere surrounding a growing void is facilitated by similar yielding generated by the expansion of neighbouring voids [13]. This theory gains experimental support from morphological studies of fracture, which show that voids are indeed produced in groups but it suffers from a difficulty in making quantitative predictions. This paper represents a first attempt to discuss the growth of groups of cavities in an isotropic polystyrene-like solid.

2. Method of calculation

As it did not seem to be feasible at this stage to make calculations on a system of spherical voids in three dimensions, it was decided to simplify the problem by using a two-dimensional array of cylindrical voids in a sheet of unit thickness as shown in Fig. 1.

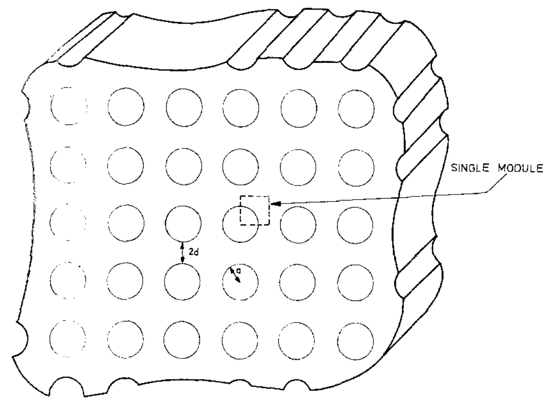


Figure 1 The two-dimensional cylindrical void model.

To this sheet two types of stress system were applied, namely 1. unidirectional tension, 2. two dimensional hydrostatic tension. In each case the plane strain constraint was imposed that the thickness should remain constant and deformation would take place solely by the enlargement of the initially cylindrical holes.

The method of solution employed was the finite element technique, which is a generalization of the stiffness method of structural analysis. Its basis lies in the subdivision of the structure into a discrete number of finite sized elements (as seen in Fig. 2) in each of which an assumed variation of the unknown quantities (usually displacements) is postulated. Continuity of the unknowns across interelement boundaries is invoked at only a finite number of points termed nodal points.

The method has the great advantage of being able to accommodate discontinuities and irregularities in material properties and geometry with ease and at present is probably the only feasible general method of solution of structures composed of materials with non-linear deformation characteristics. The finite element method is adequately explained in [14] and the details of its extension to the realm of non-linear material behaviour are to be found in [15-18].

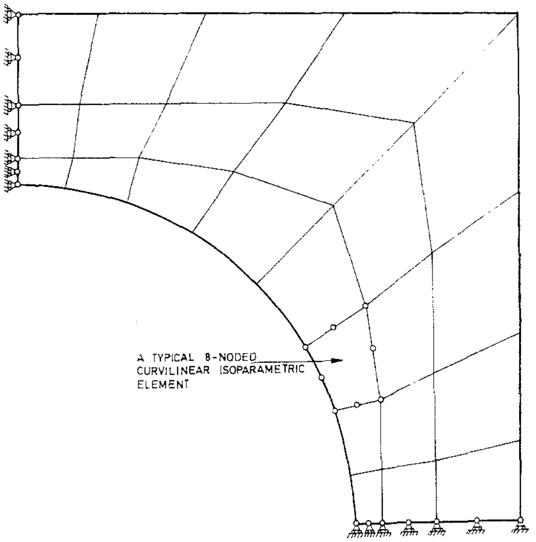


Figure 2 Typical finite element mesh of isoparametric elements.

Recent advances in both equation solution algorithms and individual element characteristics have resulted in the development of an economical computer program for the solution of elasto-plastic problems [16, 17]. Either small or large displacement problems can be accommodated subject to the restriction of small strains. The program, employing the isoparametric element concept, can readily incorporate any yield criterion for isotropic materials obeying the normality rule of plasticity with isotropic and/or kinematic hardening properties. A general purpose solution algorithm is included from which three options are available depending on the type of problem to be solved. Each method is based on the satisfaction of equilibrium by the redistribution of residual forces.

For the present study, the Von Mises yield criterion was adopted, the input information required being the elastic modulus, E, Poisson's ratio, ν, and the initial yield stress, σ_Y, of the material. The material behaviour after initial yield is determined by the effective stress versus effective plastic strain characteristic which, for the purpose of the numerical analysis, is prescribed in a piecewise linear manner.

The output quantities are displacements, stresses and strains at each nodal point as well as stresses and strains at several selected points (Gaussian integration points) within each element.

Owing to the fact that all models investigated contained a regular array of holes, it was possible to invoke symmetry to analyse only a single period or module of the model as indicated in Fig. 1. A typical finite element subdivision employed is shown in Fig. 2.

3. The selection of material constants

The first calculations were carried out using a model showing simple plasticity and having the material constants used in a previous paper [12], i.e., Young's modulus $E = 4.2 \times 10^3$ MN m^{-2};

Poisson's ratio $\nu = 0.33$; σ_Y yield strength in tension or compression without change in volume = 105 MN m^{-2}.

It was assumed that yielding is governed by the Von Mises yield criterion. This was of course a significant simplification of the actual behaviour of a polymer. For example, the effects due to different strain-rates were omitted and so were the frictional factors which are known to cause small differences between the yield stress in tension and compression for polystyrene [1, 9, 11]. However, since the work was directed to the elucidation of certain geometrical factors it was felt that these simplifications were acceptable. A much more serious limitation was thought to lie in the omission of both strain softening and orientation hardening effects. These are important since high deformations are an essential part of the processes under study. An attempt was made, therefore, in a second series of calculations to introduce a stress-strain relationship incorporating these factors.

For this purpose a basic stress-strain curve was derived from the compression curves published by Binder and Muller [19]. This gave very little difficulty as far as moderate strains were concerned but there is at present no adequate way in which the behaviour at the highest strains can be defined. Binder and Muller's results were affected by the fact that their test piece cracked at high compressive strains. Eventually, a vertical ordinate was assumed at the high strain end of the curve to provide a limiting plastic strain. Fortunately, however, none of the elementary units in our model reached a strain large enough for this limiting strain to affect the results.

Another very important difficulty which arises when an orientation hardening function is introduced into any plasticity model concerns that of the way in which it operates under different types of strain. Very little is known about this type of behaviour though some measurements on cellulose nitrate suggest that the most relevant feature may be the maximum tensile strain [20]. This was shown by the extension of a sheet material under biaxial and uniaxial tension whereupon it was found that the orientation hardening related much more closely to the maximum tensile strain than to the thickness strain. These results were analogous to those given by rubber [21].

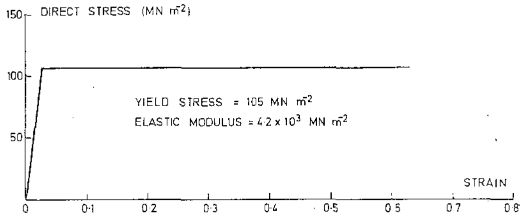
This means that the equation for plastic strain becomes

$$\text{stress} = KF(\epsilon)$$

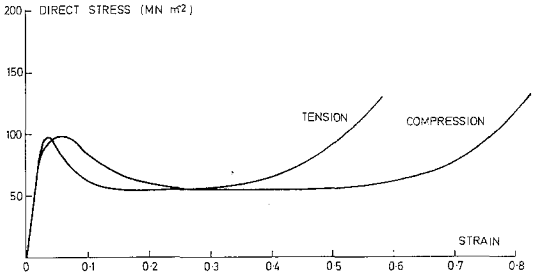
where ϵ is the maximum tensile strain. Since Binder and Muller's results were measured in compression, the relevant values of ϵ for tension are given by

$$\left(\frac{h_0}{h}\right)^{\frac{1}{2}} = \frac{l}{l_0} = 1 + \epsilon$$

where h, h_0 refers to the change in height during compression, l, l_0 to change in length in tension and ϵ is the plastic extension. Thus the assumed relation between the work hardening curves for tension and compression are as given in Fig. 3.



(a) ELASTIC - PERFECTLY PLASTIC (MODEL I)



(b) STRAIN - SOFTENING - HARDENING (MODEL II)

Figure 3 Material properties of the two-dimensional cylindrical void model. (a) Elastic-perfectly plastic (model I). (b) Strain-softening-hardening (model II).

4. Simple plastic strain model

This model did not contain an orientation hardening factor. It was studied in uniaxial and biaxial tension using the four standard values of the ratio d/a (Fig. 1) which were employed throughout these studies and which include the volume fractions known to be of interest for polymer craze structures (0.40 to 0.50 voids) [22].

Condition	d/a	Volume fraction of holes
I	0.1	0.64
II	0.5	0.35
III	1.0	0.19
IV	2.0	0.087

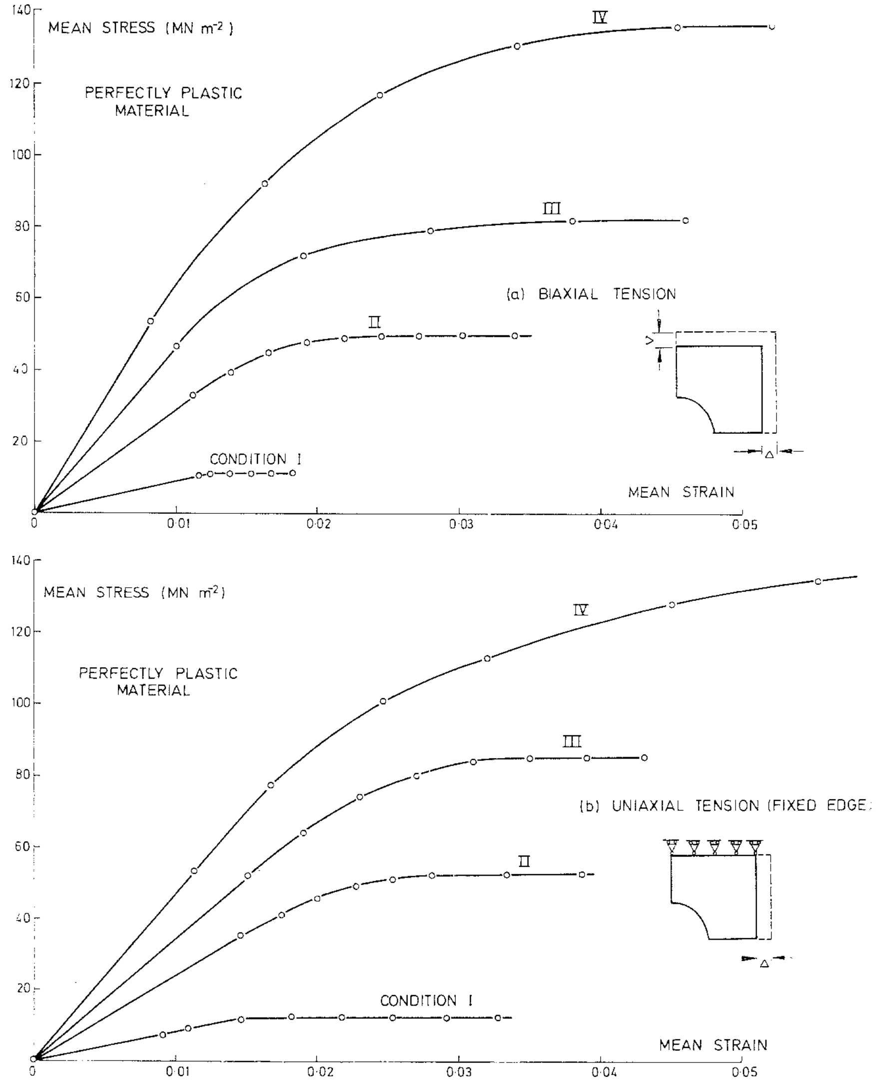


Figure 4 Mean stress-mean strain characteristics for an elastic-perfectly plastic material subjected to two loading conditions. (Mean stress = [total force/unit thickness along edge]/edge length.) (a) Biaxial tension. (b) Uniaxial tension (fixed edge).

Under these conditions both the systems have a relatively simple response. The stress-strain curve (Fig. 4) is relatively linear until plastic yield sets in, at which point yielding occurs and the whole system collapses. This collapse behaviour which occurs at a stress of 41 MN m^{-2} for a 40% volume fraction of voids, compared with an assumed yield stress of 105 MN m^{-2}, shows that structures of the type assumed will indeed yield or fail at stresses well below that required for bulk polymer. Naturally, the collapse stress will depend on the volume fraction of voids for each stress system. These results are shown in Fig. 5. In drawing these curves we have assumed that at zero volume fraction the appropriate "collapse stress" is that required to enlarge a cylinder in an infinite volume of polymer as provided by Hill's formula [23]. The significance of the results will be discussed later.

5. The use of a strain-softening, orientation hardening model

Similar calculations were then made using a stress-strain curve of the type shown in Fig. 3,

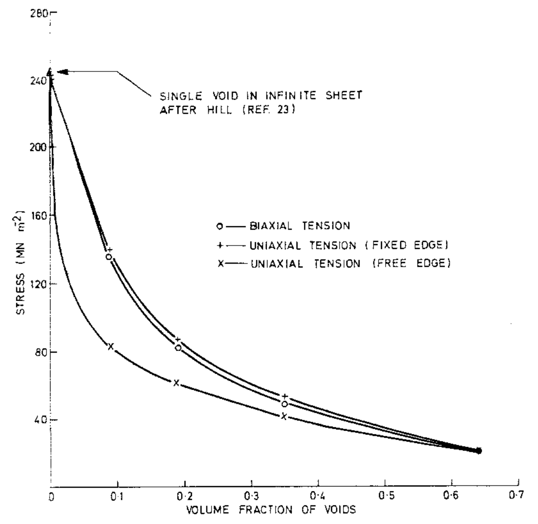


Figure 5 Collapse stresses for various void ratios and loading conditions (elastic-perfectly plastic material).

showing strain softening at intermediate strains and orientation hardening at high strains.

Examples of curves calculated in this way for the two highest cavity volumes are given in Fig. 6 for uniaxial and biaxial tension (condition II) and for biaxial tension (condition I). Fig. 7 shows the corresponding spread of plastic zones with increasing load. The load deformation behaviour for the condition I model has been extended to very high strains in an approximate manner by utilizing the fact that plastic straining is confined to the region of minimum cross-sectional area of the model as is evident from Fig. 7. This yielded area is marked ABCD in Fig. 7a. Therefore, the plastic deformation of the model can be approximated to by considering the yielding of a bar under uniaxial loading and subjected to the same plane strain condition as the model. This calculation is simple and was used to extend the load/deformation characteristic for the condition I model beyond the limit set by the computer program. The results justify two conclusions. 1. Where strain softening occurs in the tensile and compression curves, a strain softening feature is also seen in the cavity model. 2. Where orientation hardening occurs in tension or compression this will ultimately be observed in the cavity model too. This conclusion has a high degree of probability for all cavity volumes, although it could be calculated, and then only approximately in the case where the volume fraction of voids was highest (condition I).

6. Changes in the shape of the voids

In all cases, the voids were initially assumed to have a circular cross-section. In all cases the circular form becomes distorted as the system is strained. With the simple plastic strain model a smooth distortion of the cylinders takes place in uniaxial tension and also in biaxial tension as shown in Fig. 8a and b. On the other hand, the incorporation of strain softening into the constitutive law leads to neck formation and to a localized distribution of plastic strain as shown in Fig. 8c and d. In each case, however, the cylinders become elongated under the action of a unidirectional stress. The nature of this distortion is roughly equivalent to that which appears to

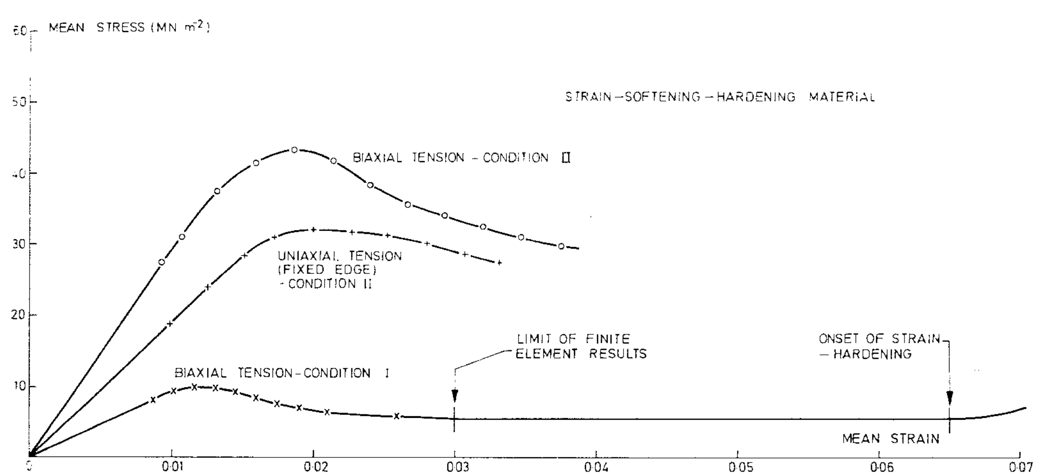


Figure 6 Mean stress—mean strain characteristics for a strain-softening-hardening material.

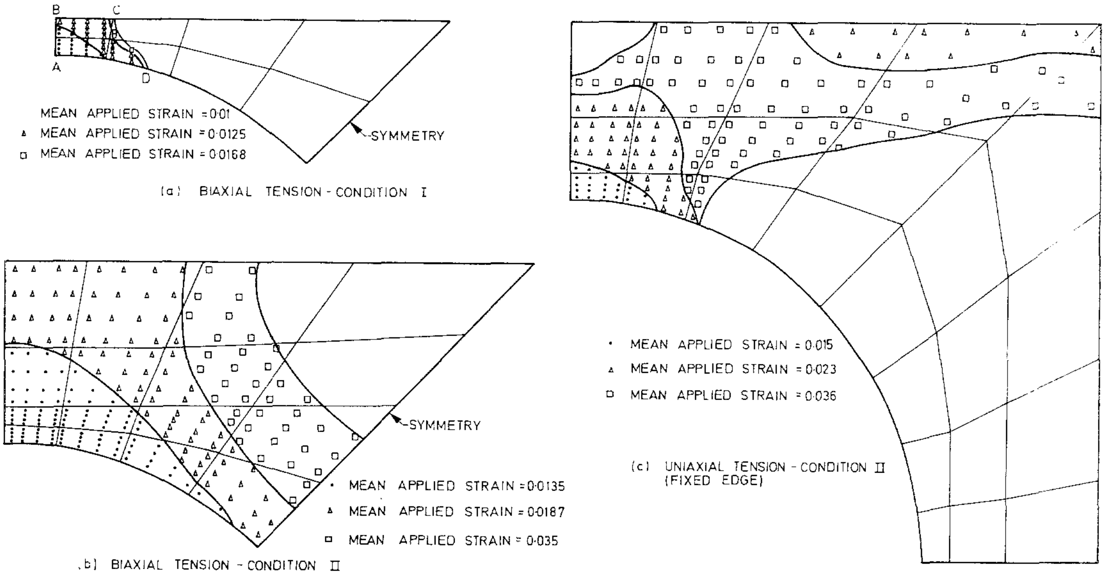


Figure 7 Development of plastic zones for an elastic-perfectly plastic material.

take place in some polystyrene crazes. See Fig. 9.

7. Discussion

For several reasons, the calculations just described can only be generally indicative of the stresses which occur during the growth of a craze-like structure in a glassy polymer. In the first place the model is only two-dimensional and based on cylindrical cavities, whereas the actual craze has cavities which may start as spheres and extend in three dimensions. There is also no fully satisfactory way in which the model can be used to calculate the stresses in a system of voids which grow from initially very small volumes. Surface tension effects are also omitted, though these would help to develop and retain a circular cross section in the smaller sized holes; their inclusion would, of course, also somewhat increase the work required to form a craze.

If we consider first the results from the first model showing simple plasticity, we find that the calculations lead in each case to a calculated collapse stress at which the structure would yield. If we now go on to assume that a craze would grow to a void content of 0.40 or 0.50 [22] if the stress applied were equal to the average collapse stress over the range of void volumes (Fig. 10), then we find from Fig. 5, that with uniaxial tension and a fixed edge these averages come out as shown.

Void volume fraction of finished "craze"	0.40	0.50	
Average stress to form "craze"	98	85	MN m^{-2}

and this compares with a yield stress of 105 MN m^{-2} i.e., in each case the stress to form the craze is below the yield stress (as required). Further, at a void content of 40%, the collapse stress is found to be 41 MN m^{-2} which is (probably somewhat coincidentally) close to the fracture stress of polystyrene [24]. This shows that the concept that the growth of a group of voids happens much more easily than with single voids is a viable one in the context.

On the other hand, it is clear that with a material having only simple plasticity, no craze could exhibit any degree of stability. As the volume fraction of holes increases the stress required to enlarge them decreases, and hence, once started a craze-like structure would proceed catastrophically to fracture.

At this point the second calculation may be considered. Here there are similar effects due to the volume fraction of holes, but it can also be shown that strain-softening will cause a reduction in stress in the early stages of the yield process. The same model, however, also strongly predicts, that at later stages, orientation hardening, if shown by the polymer, will cause an increase in the stress necessary for further

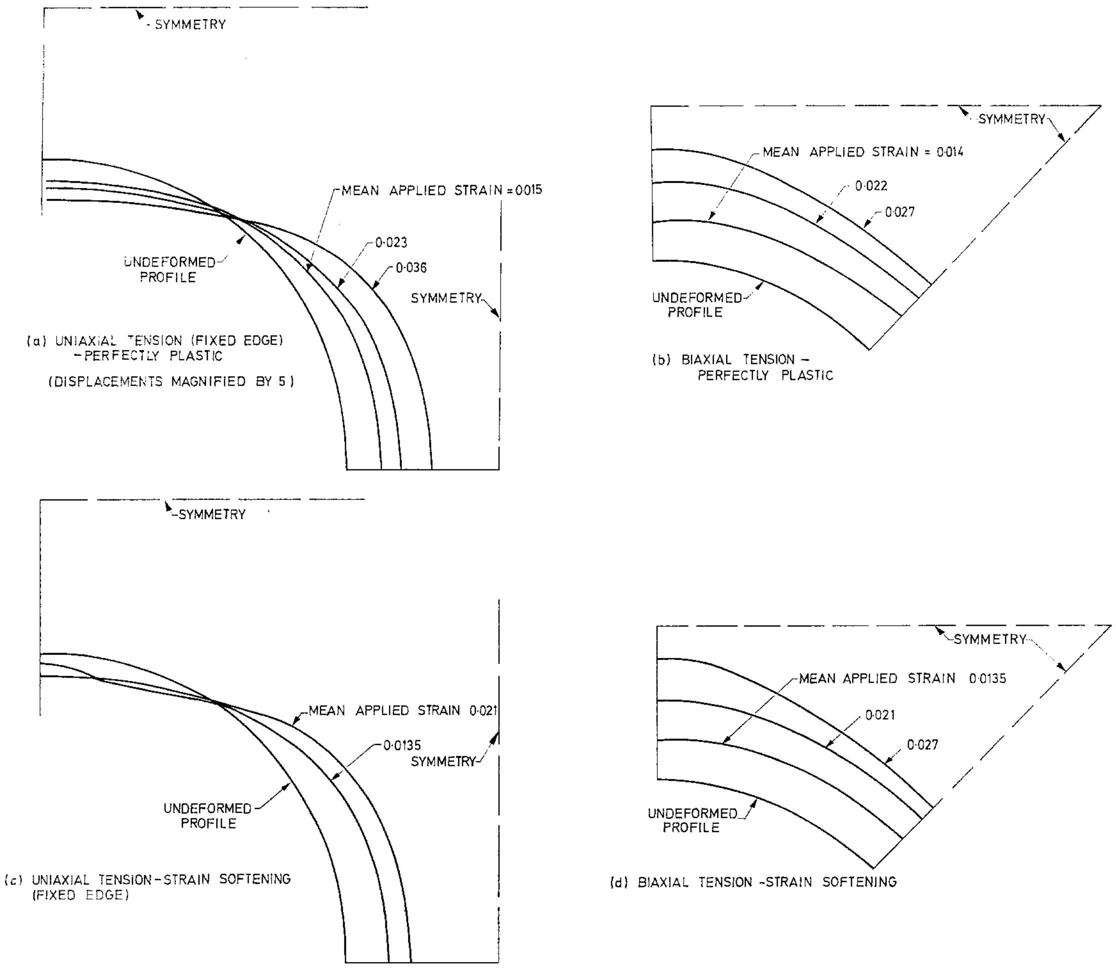


Figure 8 Distortion of holes for condition II model

growth, and so could stabilize a craze as indicated in Fig. 11. Here the existence of a region of the craze where voiding and strain softening had taken place would lead to local yielding and void expansion up to a point where orientation hardening stabilized the craze. Yielding in the partly expanded part of the craze would then lead to higher stresses at the craze tip which would allow it to propagate. This whole argument is closely analogous to that describing the formation of a stable neck during a tensile test [25].

The treatment proposed in this paper is of limited application and can provide only a part of the explanation of craze initiation. Apart from the limitations implied by the use of a cylindrical model instead of an assembly of spherical voids, which will introduce some quantitative differences but probably does not affect the form of the results, the problem of the formation of the first void remains. Some process to achieve this step still appears to be necessary to initiate a process of "overlapping proliferation" as described here.

This difficulty arises mainly in the case of the initiation of internal crazes in isotropic polymers. At the surface it is likely that there will be sufficient irregularities present to initiate local yielding at average bulk stresses below the yield stress. Such local yielding will generate a surface depression and when this has reached the point where orientation hardening sets in the material just behind it will be in a strain-softened condition. Here groups of voids can be formed whose plastic zones may in fact overlap with the surface as well as each other. These can propagate in the manner already described.

However, it is clear from the electron micrographs [7, 8] that the initial voids can be very small, perhaps less than 5 nm diameter, corresponding to a volume of some 50 nm^3. This is less than the volume occupied by a polymer molecule of mol wt 100000 and not much more than the volumes of moving segments which may be derived from the application of the Eyring equation (3 to 20 nm^3) to yield phenomena in polymers [26].

Very little is known in general about processes on this scale. In the first place, it is not at all clear that the values of yield stress obtained from macroscopic experiments actually apply. Secondly, it is probable that irregularities of structure (or flaws or impurities) will exist on this scale in a normally clear and orientation free organic glass. Finally, changes in this range may be affected by

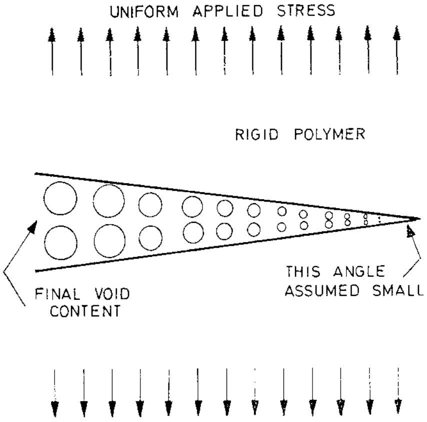
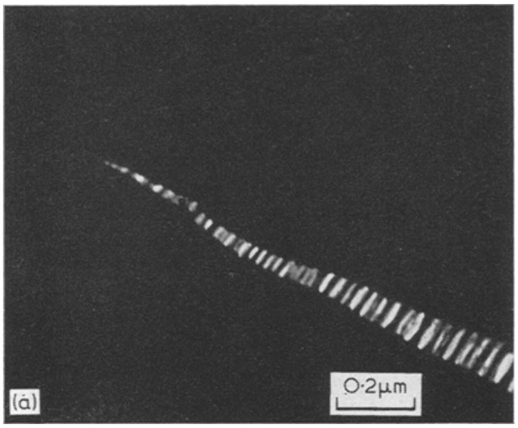


Figure 10 The growth of a cylindrical void assembly in a rigid but perfectly plastic material. Voids assumed to remain spherical.



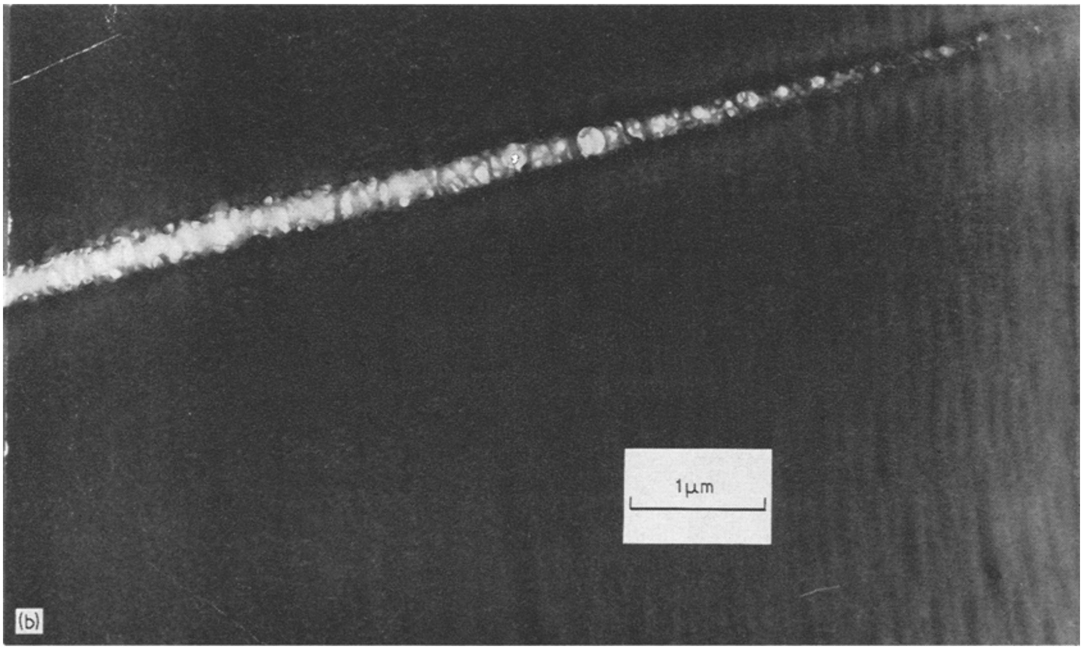


Figure 9 (a) Transmission electron micrograph of the end of a craze formed in a thin section of polystyrene. Photograph by Beahan *et al* [8]. (b) One half of a craze in homopolystyrene impregnated with iodine-sulphur eutectic. Crack initiation thought to have occurred to the left of left edge of photograph. Craze propagated left to right and top to bottom. Note progressive coarsening of structure. Photograph by Kambour and Russell [7].

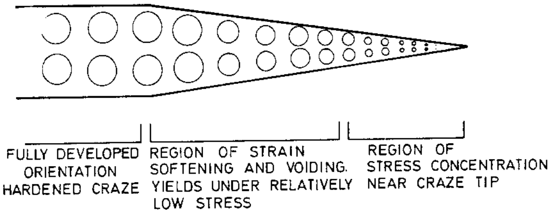


Figure 11 Craze with orientation hardening and strain softening.

energy fluctuations not different in principle from those which are assumed to cause chemical reactions which must involve changes on a scale only a factor of 10 below those considered here. A statistical mechanical treatment of a process of this type has already been applied by Fisher [27] to the problem of void nucleation in the liquid state, a process which is also concerned with voids having diameter of 1 to 5 nm, although the problem of viscous forces (yield stresses) was excluded in this case.

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