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Crazing mechanism based on plastic instability

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The mechanism of the nucleation of craze from the region of pre-formed voids is discussed on the basis of the finite elements analysis for an elastic plastic material containing a two-dimensional array of cylindrical voids. Calculations are made for the two kinds of boundary conditions with respect to the constraint of the transverse strain under simple tension. The craze formation is considered to be an unstable concentration of plastic strain within the ligament between neighbouring voids. The present calculation shows that the constraint of the transverse strain is the essential factor in craze formation.

1. Introduction

It is well known that crazing is the direct precursor of fracture in polymeric materials [1]. The crazes contain a large number of fibrils and voids in a narrow disc-shaped zone propagating in the direction perpendicular to the maximum principal stress [2].

It is most fruitful to construct a theoretical model for crazing based on its microstructure paralleled to the ductile fracture which is a process of nucleation of holes, followed by their growth and coalescence by the plastic deformation. Haward and Owen [3] discussed a general growth condition of cavities in an isotropic solid by the use of the finite elements method (FEM) for a two-dimensional array of cylindrical voids. Argon and Hannoosh [4] have also proposed a criterion of craze nucleation based on the plastic expansion of voids at a certain level of porosity which depends on both the dilational stress and the shear stress.

Generally, it is recognized that the craze starts from the surface of polymer at a critical stress which is lower than the yield stress. On the other hand, we have already suggested [5–8] that the brittle fracture of ductile polymers with a round notch was initiated from the internal craze which was nucleated at the tip of the local plastic zone when the size of the local plastic zone initiated at the notch tip reached a certain critical size. In these studies, for notch brittleness of ductile polymers, it was indicated from analysis of plasticity that the stress for the nucleation of craze is larger than the yield stress of the polymer, except for crazes nucleated at extremely low temperature or high strain rate. In addition, the development of voids was observed between the internal craze and the plastic deformation zone for poly(vinyl chloride) in the amorphous glassy polymer and for polyethylene in the crystalline polymer. As the applied load increased, this region of voids could be deformed plastically and the crazes were nucleated at the tip of the region of voids when the size of the local plastic zone reached a certain critical size. It is suggested from this that the constraint of strain in the direction perpendicular to the direction of maximum principal stress is necessary for the nucleation of craze from the voids.

Because there is a large plastic strain concentration in the craze which is a narrow planar yield zone, elastic unloading of the surrounding material is required for the instability of plastic deformation.

The purpose of this study was to examine the crazing processes from the region of voids in polymethylpenthene(TPX), a transparent crystalline polymer, and to discuss the mechanism of the craze nucleation from the region of pre-formed voids on the basis of an FEM analysis for an elastic–plastic material.

2. Experimental details

The material used was a commercial grade of TPX(RT-18) (Mitsui Petrochemistry Co. Ltd) supplied in the form of pellets. The sheets of 1 and 6 mm thickness were compression moulded at 503 K. Specimens for tensile mechanical measurements were prepared by milling the 1 mm thick sheets. A round notch, of radius 0.5 mm, in the 6 mm thick sheets for three-point bending test was made by machining with a convex milling cutter. The specimens were loaded in three-point bending with a span length of 40 mm in an Instron-type testing machine (Auto Graph, Shimadzu DDS-5000). Tests were carried out at a bending rate of 1 mm min^{-1} at 296 K.

To examine the process of craze nucleation from the region of voids in the three-point bending under plane strain, thin sections of about 25 μm were cut normal to the plane of the initial notch from the unloaded samples, as shown in Fig. 1. The morphology of the craze and plastic deformation was studied in an optical microscope using polarized light and/or dark-field light. The changes in microstructure on the surfaces of cryogenically fractured samples were examined with a scanning electron microscope. Samples, which were subjected to the bending test, were first immersed in a liquid nitrogen bath for 5 min, and broken normal to the plane of the notch immediately after removal (Fig. 1). Because strain recovery on

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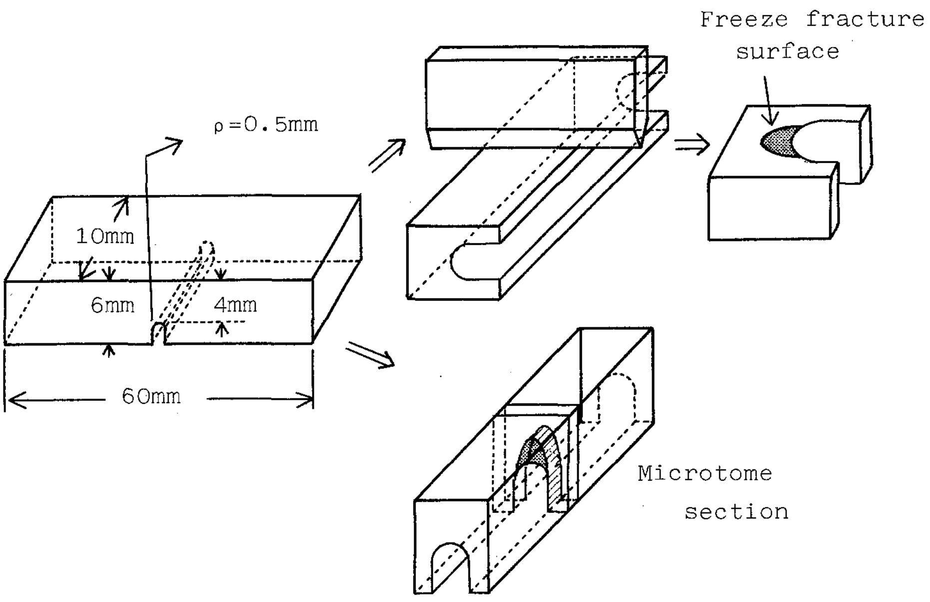


Figure 1 Dimension of the test sample and the cutting direction for observation of deformation zones.

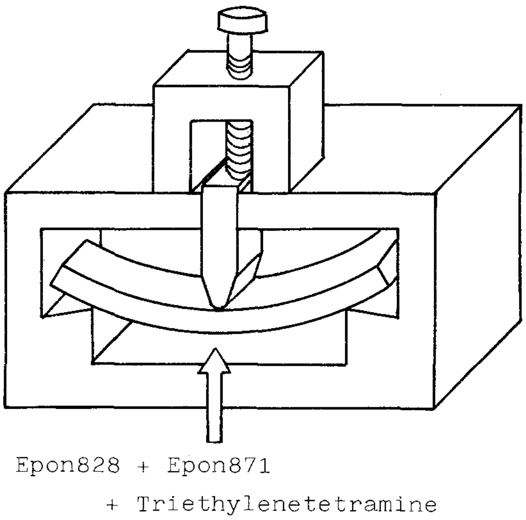


Figure 2 Fixation of strain by casting in epoxy resin.

unloading influences significantly the morphology of deformation zone, the deformation by three-point bending was fixed by casting in epoxy resin, which consisted of 60 p.h.r. Epon 828, 40 p.h.r. Epon 871 and 9 p.h.r. triethylene–tetramine, as shown in Fig. 2.

3. Results

Fig. 3 shows the bending moment displacement curves of TPX and the deformation processes associated with the curve, which were observed by dark-field microscopy. The strain of samples was fixed by casting in epoxy resin in order to observe the microtomed sections in the microscope. It was found that the development of stress whitening initiated from the notch tip at about 60% applied bending moment of the fracture moment (A). The region of stress whitening spread as the applied moment was increased. At about 80% applied bending moment of the fracture moment (B), the craze, which was propagated in a direction normal to the direction of maximum principal stress as the local concentrated plastic deformation band, was nucleated at the tip of the region of stress whitening. There is a higher birefringence in the craze. Fig. 4 shows scanning electron micrographs of the deformation zone obtained on the sample in which the strain was fixed by epoxy resin at 80% applied bending moment of fracture moment. Numerous stable voids, similar to micro-crazes, had developed in the region of stress whitening. It was clear that the density of the nucleation of voids increased with increasing distance from the notch tip. The craze was

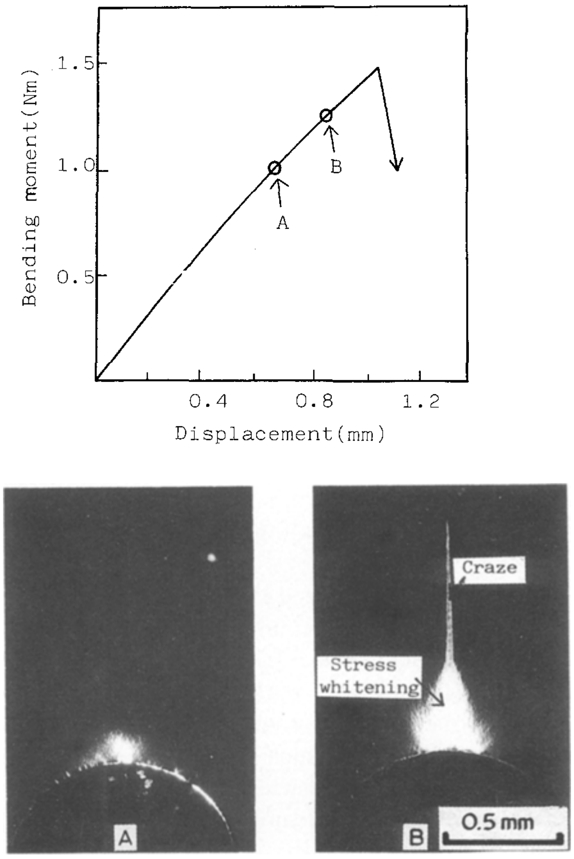


Figure 3 Bending moment displacement curve of TPX with a round notch and deformation processes associated with the curve.

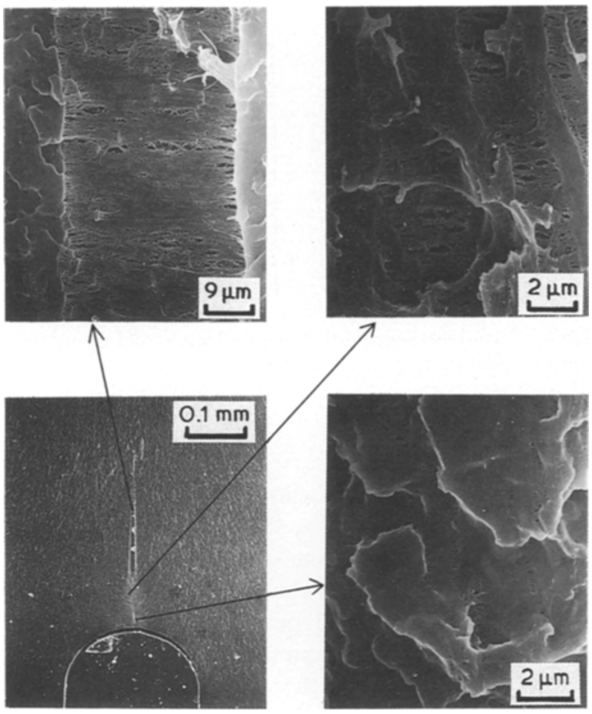


Figure 4 Scanning electron micrographs of the deformation zone.

nucleated at the tip of the region of stress whitening constructed by the numerous voids. They contain the fibrils oriented in the direction of maximum principal stress, which arose from the local concentration of plastic strain, and the elongated voids between these fibrils. Fig. 5 shows the fracture surface of TPX. The dimple-type fracture surface, which is the most typical surface for ductile fracture, was clearly observed in the region of crazing. Thus it was concluded for TPX that crazing is a process of nucleation of holes, followed by their growth and coalescence by plastic deformation.

4. Discussion

It is already known that, under the plane strain state, the stress distribution of the local plastic zone (Fig. 6) which developed at the tip of a round notch, may be expressed by the use of the mechanics of plasticity [9]

$$\sigma_y = \sigma_{\mathrm{mean}} + \tau \qquad (1)$$

$$\sigma_x = \sigma_{\mathrm{mean}} - \tau \qquad (2)$$

$$\sigma_{\mathrm{mean}} = \tau[1 + 2\ln(1 + x/\rho)] \qquad (3)$$

where σ_{mean} is the mean stress ($\sigma_{\mathrm{mean}} = (\sigma_x + \sigma_y + \sigma_z)/3$), τ is the shear yield stress, ρ the notch radius and x the distance from the notch tip. The stress at the tip of notch on this stress distribution satisfies the yield criterion on tension under the plane strain state, that is $\sigma_y = 2\tau$, $\sigma_z = \tau$ and $\sigma_x = 0$. The stress of σ_y is increased with increasing distance from the notch tip by the development of the stress σ_x in the x-direction caused by the constraint of strain. There is a maximum stress at the tip of the local plastic zone. The formation of numerous voids occurred by this dilational stress, as observed on TPX. The development of voids caused by the dilational stress has been previously observed on PE for a crystalline polymer and PVC for a glassy amorphous polymer. Generally, an isolated void introduces a stress concentration around it. However, when the number of voids is increased, the stress of σ_x introduced by the constraint of strain is relaxed, because the stress in this direction cannot be borne by these voids. Berg [10] suggested a yield criterion of ductile materials containing voids by considering the above condition

$$(\sigma_1 - \sigma_2)^2 + (\sigma_2 - \sigma_3)^2 + (\sigma_3 - \sigma_1)^2 = 6[\tau_0 - g(p)/2]^2 \qquad (4)$$

where σ_1, σ_2, σ_3 are the principal stresses, τ_0 is the shear yield stress of continuous material and $g(p)$ is the parameter which increases with an increase in both the content of voids and the dilational stress. This equation is formally equivalent to the pressure-dependence yield criterion. Thus the stress of the local plastic zone within the numerous voids is decreased in comparison with that of the continuous material, as shown in Fig. 7. The craze nucleation, in which the plastic strain was locally concentrated between neighbouring voids, occurred at the tip of the plastic zone, when the constraint of strain was further enhanced by the expansion of the plastic zone, as can be seen in Fig. 3. In order to concentrate the plastic strain between the voids, it is necessary to unload the surrounding elastic region because of the similarity of the craze to the crack. Argon and Hannoosh [4] proposed the criterion for the nucleation of a craze which can be stated as a sufficient condition for the rapid plastic cavitation of the region of voids by elastic–plastic unloading. However, we have suggested [1] experimentally that the calculated value from Argon's equation over estimates the stress required to nucleate the internal craze of PC.

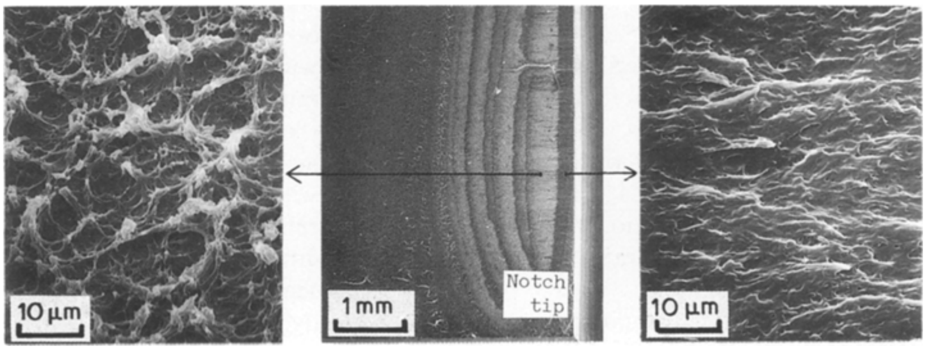


Figure 5 Scanning electron micrographs of the fracture surface of TPX.

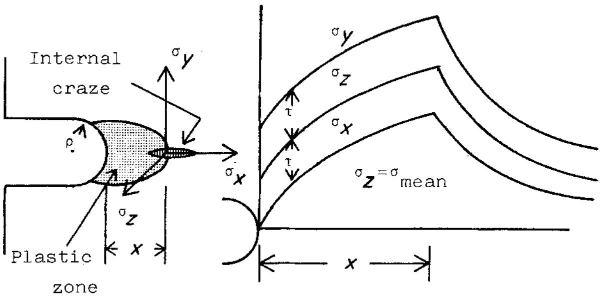


Figure 6 Definition of the stress components.

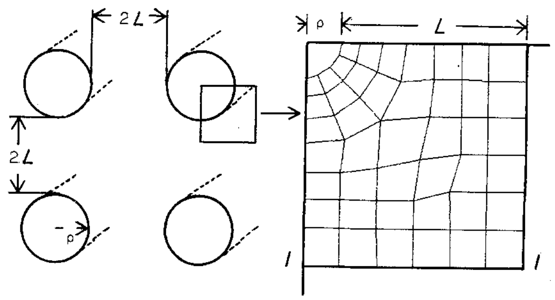


Figure 8 Two-dimensional cylindrical void model and typical finite element mesh of isoparametric elements.

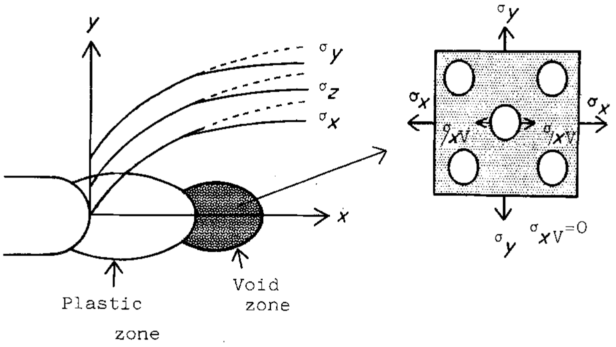


Figure 7 Variation of stress distribution caused by nucleation of voids.

The crazing processes contain the nucleation of voids, followed by their growth and coalescence by plastic deformation. In addition, it may be surmised that the constraint of strain in the direction perpendicular to the maximum principal stress is required to propagate the craze, because the origin of stress concentration is the constraint of strain. We attempted to simulate the growth of voids on the two distinct states of strain constraint by the use of FEM for an elastic–plastic material. It is reasonable to use the model of a two-dimensional array of cylindrical voids because the plane strain state is assumed in this problem. Fig. 8 shows a two-dimensional cylindrical voids model and a typical finite element mesh of isoparametric elements. In this model two types of boundary condition were adapted for strain constraint as follows:

1. the strain perpendicular to the maximum principal stress equals zero ($\sigma_y = 0$) (constraint of strain);
2. the mean stress perpendicular to the maximum principal stress equals zero($\sigma_y = 0$) without the distortion of line l–l (uniaxial tension(fixed edge)).

It was assumed that yielding is governed by a Von Mises yield criterion. The effects of both pressure and orientation hardening were omitted. The calculations were carried out using a model with material constants as follows: Young's modulus $E = 4.2 \times 10^3\,\mathrm{MN\,m^{-2}}$; Poison's ratio $\nu = 0.33$; yield stress in tension under plane strain without change in volume $2\tau = 105\,\mathrm{MN\,m^{-2}}$.

Fig. 9 shows the calculated stress–strain curve for the model constraining the strain in a direction perpendicular to the tension direction, and the development of a plastic zone associated with the curve. It was found that the mean tensile stress was increased over the yield stress under plane strain tension due to the plastic constraint. It was characteristic that the void was plastically expanded primarily in the direction perpendicular to the maximum principal direction, because the volume of the model was maintained constant during plastic deformation. As a result, the ligament thickness between neighbouring voids was decreased with increasing strain. When the ligament thickness was decreased below a certain critical thickness, the mean stress reached a maximum volume and then decreased in analogy with necking in the tensile test of a polymeric material, that is, elastic–plastic unloading took place. Therefore, the plastic strain was unstably concentrated into the ligament between neighbouring voids and the nucleation of a craze occurred. On the other hand, in the calculated results for the model without the strain constraint (uniaxial tension under plane strain), unloading was not observed in the calculated stress–strain curve, as shown in Fig. 10. The stable plastic zone initiating from the void was spread in the direction 45° to the maximum principal stress in contrast to the model constraining the strain. The circular form void was elongated into an ellipsoid shape under the action of unidirectional stress as the system was strained. The degree of the growth of voids is smaller than that for the model constraining the strain. These calculated results clearly conclude that the formation of crazes, which contains the processes of the nucleation of voids, their growth and then elastic–plastic unloading, followed by the unstable concentration of plastic strain into the ligament between voids, requires the constraint of transverse strain under the plane strain state.

It is already known that the ratio of the critical stress for craze nucleation to the shear yield stress is decreased with decreasing temperature and/or increasing strain rate [11]. It can be suggested from the crazing mechanism mentioned above that the ligament thickness between neighbouring voids is decreased with decreasing temperature and/or increasing strain rate. Thus the void content in the critical condition for craze nucleation is increased with decreasing temperature and/or increasing strain rate. If a perfectly rigid–plastic body for the material is assumed, the general yield stress of the ligament between neighbouring voids is given [9] by:

for $\rho/L < 1/(e^{\pi/2} - 1)$

$$\sigma_{y\mathrm{crit}} = 2\tau\{(1 + \pi/2) - [e^{\pi/2} - (1 + \pi/2)]\} \times L/(L + \rho) \qquad (5)$$

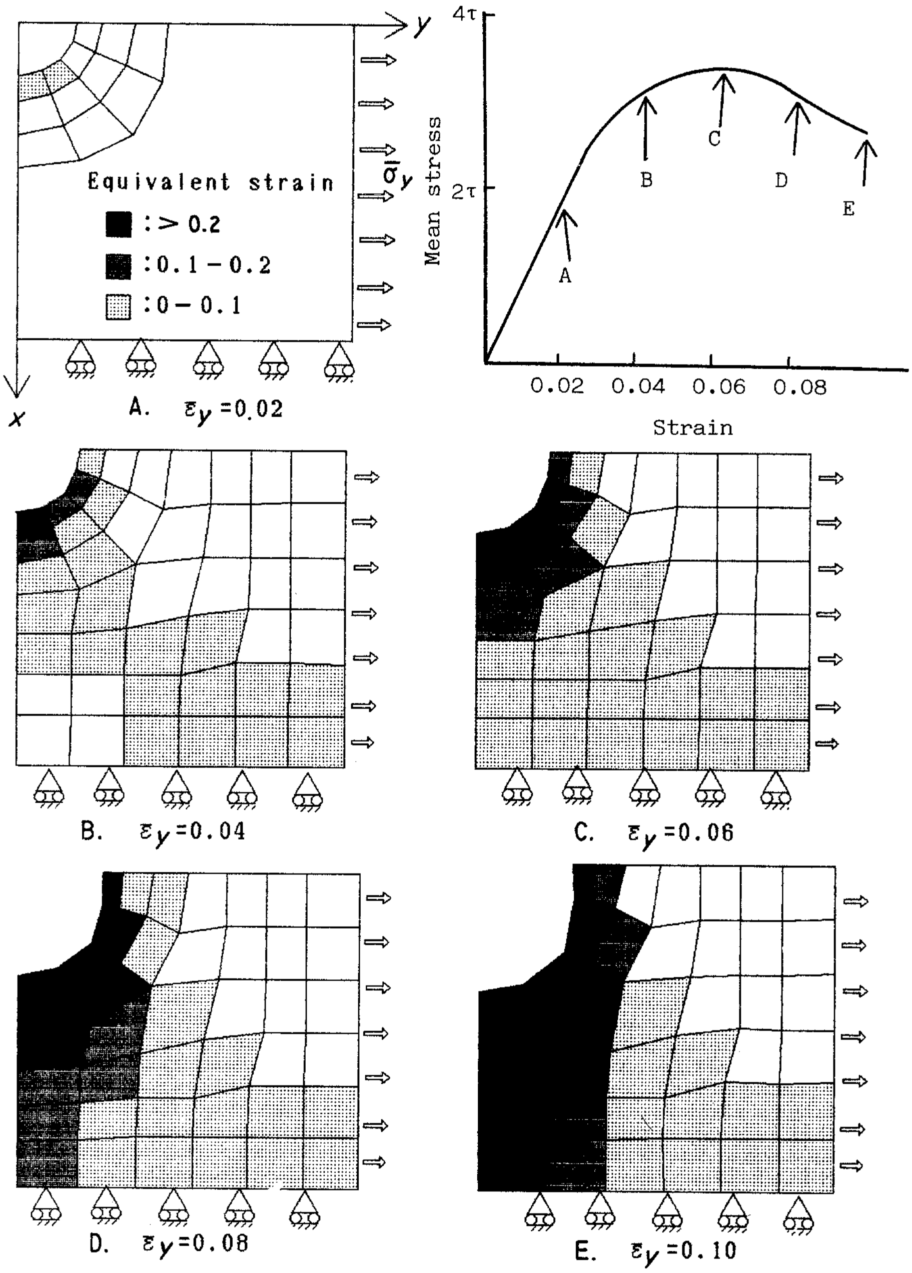
for $\rho/L > 1/(e^{\pi/2} - 1)$

$$\sigma_{y\mathrm{crit}} = 2\tau[(1 + \rho/L)\ln(1 + L/\rho)] \times L/(L + \rho) \qquad (6)$$

In this study, the general yield stress is about $2\tau \times 1.84$ for $L/\rho = 6$. This volume is nearly equal to that estimated from the calculated results by the use of FEM. The critical content of voids for craze nucleation can be calculated from the ratio of ligament thickness to the radius of the void using the following equation

$$V_{\mathrm{cr}} = 074[1/(L/\rho + 1)]^3 \qquad (7)$$

which is obtained on the assumption of regular close packing [12]. For example, the critical content for PC at room temperature is about 1 vol % and for PMMA at room temperature and a strain rate of 100% min^{-1} is 3 vol %. When the density of the development of voids on the nucleation of a craze is increased, the ratio of the stress for craze nucleation to the shear yield stress is decreased. Above 3.7 vol % voids content, the stress for craze nucleation is smaller than the yield stress. A typical example of this type of craze is shown by polypropyrene in which the stress for craze nucleation is nearly equal to the yield stress and in polystyrene in which the critical content of 6.7 vol % is estimated for a ratio of critical stress to yield stress of 0.8. These results point out that void nucleation is a most fundamental process for craze nucleation.



A. $\varepsilon_y = 0.02$

B. $\varepsilon_y = 0.04$

C. $\varepsilon_y = 0.06$

D. $\varepsilon_y = 0.08$

E. $\varepsilon_y = 0.10$

Figure 9 Calculated stress–strain curve for the model constraining the strain in the direction perpendicular to the direction of tension and development of a plastic zone associated with the curve.

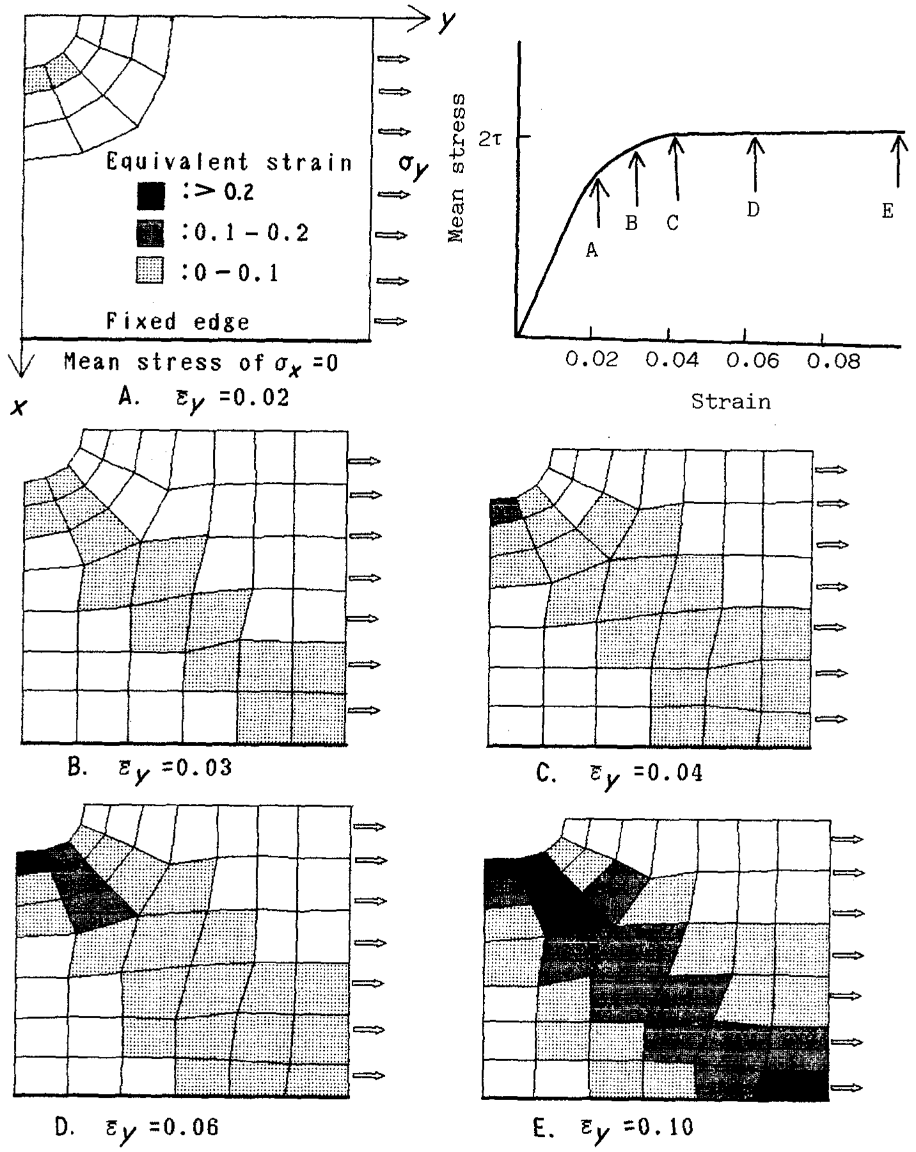


Figure 10 Calculated stress–strain curve for the model without constraint of strain and development of the plastic zone associated with the curve.

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References

1. J. P. BERRY, *J. Polym. Sci.* **50** (1961) 107.
2. R. P. KAMBOUR and A. S. HOLIK, *J. Polym. Sci. A-2* **7** (1969) 463.
3. R. N. HAWARD and D. R. J. OWEN, *J. Mater. Sci.* **8** (1973) 1136.
4. A. S. ARGON and J. G. HANNOOSH, *Phil. Mag.* **36** (1977) 1195.
5. M. ISHIKAWA, I. NARISAWA and H. OGAWA, *J. Polym. Sci. Polym. Phys. Edn* **15** (1977) 197.
6. M. ISHIKAWA, H. OGAWA and I. NARISAWA, *J. Macromol Sci. Phys. Edn* **B19** (1981) 421.
7. M. ISHIKAWA and I. NARISAWA, *J. Mater. Sci.* **18** (1983) 1974.
8. *Idem, ibid.* **18** (1983) 2826.
9. R. HILL, "The Mathematical Theory of Plasticity" (Clarendon Press, Oxford, 1950).
10. C. A. BERG, in "Inelastic Behavior of Solids", edited by M. F. Kanninen, W. F. Addler, A. R. Rosenfield and R. I. Jaffer (McGraw-Hill, New York, 1971) p. 171.
11. M. ISHIKAWA, H. OGAWA and I. NARISAWA, *J. Macromol Sci. Phys. Edn* **B19** (421) 1989.
12. M. ISHIKAWA and I. CHIBA, *Polymer* **31** (1990) 1231.

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