

## POLYMER CRAZING AND ACOUSTIC EMISSION

IGOR GRABEC

*Faculty of Mechanical Engineering, Edvard Kardelj University of Ljubljana, pp. 394, 61000  
Ljubljana, Yugoslavia*

Received 3 July 1986

UDC 541.64

Original scientific paper

The formation of fibrils at the growing craze-tip is a sequential process which should act as a sound source. In the article the mechanism of meniscus instability is applied for estimating the characteristic frequency and amplitude of the corresponding acoustic emission signal. The estimated value of frequency (typically about 1 Hz in mild environmental conditions) lies far below the frequency band of commercial acoustic emission monitors, while the estimated wave amplitude (typically of the order  $10^{-13}$  m) together with the broad spectral width indicate that corresponding signals are normally hidden in environmental and instrumental noise. This explains the absence of AE signals during the smooth development of crazes and at the same time leaves the mechanism of detectable acoustic emission in polymers to be explained on the grounds of cracking.

### *1. Introduction*

The strength of various glassy polymers in normal and aggressive environmental conditions is mainly limited by the development and growth of crazes. Extensive research work has therefore been done on this phenomenon including studies of morphology, fracture mechanics, kinetics and most recently also dynamics<sup>1,2,3</sup>. Due to the submicroscopic size of the craze tip region, information about the dynamic mechanism of craze development was obtained by morphological comparisons of electron microscope records of advancing craze-tip with similar images of discontinuities macroscopically developing in viscoplastic materials<sup>4,5,6</sup>.

The corresponding theoretical model of developing craze is based on the assumption that the structure of microvoids and fibrils develops due to meniscus instability caused by the stress gradient in viscoplastic polymer. Taking into account the material parameters of monodisperse polystyrene, Donald and Kramer<sup>4)</sup> indeed succeeded in calculating the characteristic distance between microfibrils in a craze-tip, which closely corresponds to experimental observation. This agreement indicates that not only morphological but also true dynamic parameters, such as the frequency of fibril formation etc., could be predicted from the same model, which is the aim of this article.

Changes in stress distribution caused by development of a craze are accompanied by emission of stress waves (acoustic emission AE). By the detection and analysis of AE signals information about source properties are obtained. Until now crazing or cracking of polymers was mainly characterized by the rate of AE events which can be correlated with the growth kinetics<sup>7,8,9)</sup>. Only recently the author attempted to determine forces released on a developing defect in a polymer by the deconvolution of the AE signal<sup>10)</sup>. Perhaps the most interesting result of all these studies is that acoustic emission is often not detectable in spite of evident crazing or even cracking. Until now only a plausible explanation of this observation has been forwarded by Peterlin<sup>11)</sup>. It is one of the tasks of this paper to give a quantitative basis for this explanation on the grounds of the mechanism mentioned of craze-tip advancement. For this purpose we try to estimate the characteristic frequency and amplitude of the AE signal generated by developing craze.

## 2. The mechanism of craze-tip advancement

Meniscus instability and its inclusion in models of propagating discontinuities in solids and liquids has been explained by various authors and is here only shortly reviewed<sup>4,5,6)</sup>. They assumed that a close similarity exists between a craze-tip and the meniscus of a liquid between two rigid plates as shown in Fig. 1. A charac-

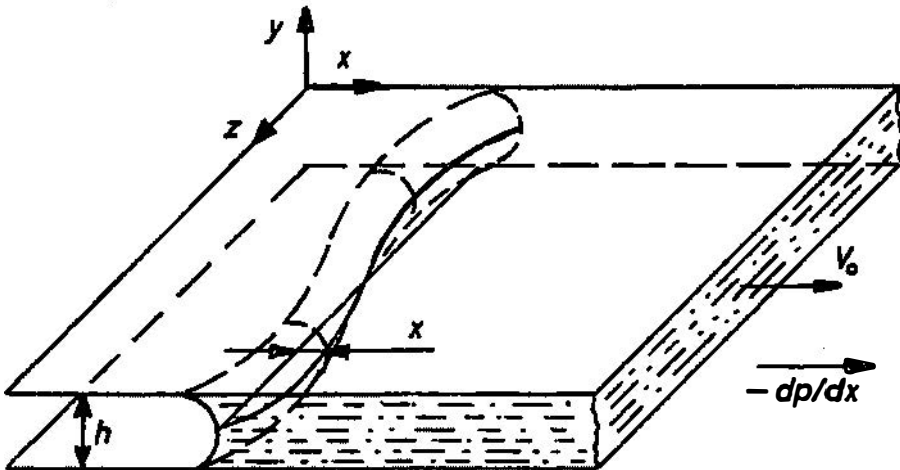


Fig. 1.

teristic parameter of discontinuity is the spacing  $h$  between the plates at the distance from the tip where meniscus curvature is already negligible. The liquid is sucked by a negative pressure gradient  $-dp/dx$  with constant velocity  $v_0$  in the direction of growing discontinuity. Let us assume that in a steady state the meniscus is straight and has a curvature  $R$  at the head. If its straight form is perturbed from the coordinate  $x_0$  to

$$x = x_0 + s \cos 2\pi z/\lambda \quad (1)$$

the head curvature is changed for

$$1/R = d^2x/dz^2 = -s(2\pi/\lambda)^2 \cos 2\pi z/\lambda. \quad (2)$$

The surface tension  $T$  tends to diminish this disturbance by the equivalent pressure difference  $\Delta p_s = T/R$  while the pressure gradient amplifies it by  $\Delta p_g = -s dp/dx$ . The disturbance is therefore unstable if the condition

$$\Delta p_g + \Delta p_s > 0 \quad (3)$$

is fulfilled. By assuming just unstable disturbance we get the expression for the lower limit of wavelength:

$$\lambda = 2\pi \sqrt{T/|dp/dx|}. \quad (4)$$

More detailed analysis of meniscus instability showed<sup>5)</sup> that the wavelength of disturbance with maximal increment is given by

$$\lambda_0 = \sqrt{3} \lambda = 2\pi \sqrt{3T/|dp/dx|}. \quad (5)$$

Although possible in principle, it is difficult to measure the pressure gradient, therefore it is better to express it by velocity  $v_0$  of meniscus propagation. In the case of viscous flow between two plates we can apply the standard formula for meniscus head velocity:

$$v_0 = -\frac{h^2}{8\eta} \left( \frac{dp}{dx} \right) \quad (6)$$

in which  $\eta$  denotes liquid viscosity. This velocity is greater by a factor 8/12 than the average flow velocity usually taken into account by other authors. Inserting Eq. 6 into (5) we get

$$\lambda_0 = 2\pi \sqrt{3Th^2/8v_0\eta}. \quad (7)$$

Taking into account a more general flow law of type:

$$\dot{\epsilon} = \dot{\epsilon}_0 (\sigma/\sigma_0)^n \quad (8)$$

Fields and Ashby<sup>6)</sup> derived an equivalent expression applicable also for plastic flow:

$$\lambda_0 = 2\pi \sqrt{3 \sqrt{3} Th/2\sigma_0}. \quad (9)$$

Here  $n$ ,  $\sigma_0$  and  $\dot{\epsilon}_0$  are material parameters, which for the Newtonian liquid are given by:  $n = 1$  and  $\eta = \sigma_0/3\dot{\epsilon}_0$ .

Donald and Kramer<sup>4)</sup> used the following data for the monodisperse polystyrene:  $T = 0.04 \text{ J/m}^2$ ;  $\dot{\epsilon}_0 = 1.7 \cdot 10^{-5} \text{ s}^{-1}$ ;  $\sigma_0 = 35 \text{ MPa}$ ,  $n = 17$ ,  $v_0 = 10^{-8} \text{ ms}^{-1}$  that give for the craze thickness of the order  $h = 10 \text{ nm}$  the characteristic wavelength  $\lambda_0 = 20 \text{ nm}$ , which is in good agreement with the experimental results.

### 3. The properties of AE generated by developing craze-tip

Due to meniscus instability a sinusoidal disturbance first develops which, under favourable conditions, is then further transformed into a fingerlike structure<sup>6)</sup>. Images of craze-tips show that fingers are also unstable and decay into series of microfibrils between both sides of a craze. They are spaced approximately the same distance apart as the fingers themselves<sup>4)</sup>. A detailed physical description of filament evolution is at present still outside our scope, but we can suppose that the same mechanism that governs the development of fingers is also responsible for their further decay. Although both phenomena are physically similar there exists a marked difference caused by geometrical properties. Formation of a fingerlike structure in the longitudinal direction of flow velocity is a smooth continuous process, while the transversal decay of fingers into fibrils must take place sequentially with a certain characteristic frequency  $f$ . This frequency is determined by the ratio of meniscus head velocity  $v_0$  and characteristic distance between fibrils, which approximately equals  $\lambda_0$ :

$$f = v_0/\lambda_0 = \frac{1}{2\pi} \sqrt{\frac{2v_0^2 \sigma_0}{3\sqrt{3} h T}} \quad (10)$$

From the quoted data for monodisperse polystyrene we get an estimation  $f = 0.5 \text{ Hz}$ .

It is interesting that characteristic frequency  $f$  can be assessed directly from experimentally determined  $\lambda_0$  and  $v_0$  without any assumption about the mechanism of craze development. Experiments on different materials (PMMA, PS, polycarbonate etc.) show that the craze velocity in mild environmental conditions ranges from zero to the values a few orders of magnitude above or below  $10^{-8} \text{ ms}^{-1}$ , while the fibril spacing at the craze tip is of the order  $10 \text{ nm}$ <sup>1)</sup>. A typical frequency is therefore of the order  $1 \text{ Hz}$  or below.

If the stepwise fibril formation is accompanied by stress fluctuation, then the frequency of the corresponding acoustic signal lies far below the frequency band  $0.1\text{--}1 \text{ MHz}$  in which acoustic emission is usually monitored. This great discrepancy indicates that by the currently available AE instrumentation the development of crazes under normal environmental conditions practically cannot be detected.

The same conclusion stems also from energy or amplitude considerations. Let us assume that the craze proceeds for  $\lambda_0$  in a typical time  $t_0 = 1/f = \lambda_0/v_0$ . The volume of a polymer transformed into microvoid and fibril in this step is of the order  $V_0 = \lambda_0^3$ . Experimental investigations of Lauterwasser and Kramer<sup>3)</sup> showed that the stress on the craze is only slightly different from the applied stress

in increased material. We can therefore suppose that in the assumed step of microtransformation only elastic energy contained in the volume  $V_0$  is released. It is equal to

$$W = \frac{E \varepsilon^2}{2} \lambda_0^3 \quad (11)$$

Here  $E$  is the modulus of elasticity and  $\varepsilon$  is the strain at the craze-tip which is of the same order as the average value in the sample. The order of magnitude of  $W$  is most simply assessed if for  $\varepsilon$  the yield or the critical value for the craze development is taken into account<sup>1)</sup>.

The released elastic energy is partly spent for plastic deformation related with fibril formation and partly emitted by elastic wave. In the most favourable situation the latter part is approximately of the same order as the released energy  $W$ . Due to wave propagation the energy liberated in the time interval of span  $t_0$  is spread into the volume of the order  $V = (ct_0)^3$  where  $c$  is the speed of sound. If the distance  $ct_0$  is greater than specimen dimensions then wave reflections on the boundary complicate the situation but we can still expect that the energy density in the emitted wave is of the order

$$\frac{W}{V} = \frac{E \varepsilon^2 V_0}{2 (ct_0)^3} = \frac{E \varepsilon^2}{2} \left( \frac{v_0}{c} \right)^3 \quad (12)$$

Expressing this density by the wave amplitude and frequency we get the equation

$$\frac{W}{V} = \frac{q A^2 (2\pi f)^2}{2} = \frac{E \varepsilon^2}{2} \left( \frac{v_0}{c} \right)^3 \quad (13)$$

in which  $q$  denotes the polymer density. By introducing the relation  $c^2 = E/q$  we derive for the wave amplitude the expression

$$A = \frac{c}{2\pi f} \left( \frac{v_0}{c} \right)^{3/2} \quad (14)$$

Applying the following values for the monodisperse polystyrene:  $\varepsilon = 0.5$ ;  $v_0 = 10^{-8} \text{ ms}^{-1}$ ,  $f = 0.5 \text{ s}^{-1}$ ,  $c = 2.5 \cdot 10^3 \text{ ms}^{-1}$  we obtain  $A \cong 10^{-14} \text{ m}$ . This value represents an overestimated amplitude of acoustic wave corresponding to a single fibril-void formation.

The craze development is composed of a series of such single events which are more or less synchronous. Electron microscope images indicate that typical correlation length of fibril spacing is of the order of a few  $\lambda_0$ . This means that the number of synchronously cooperating microevents is of the order 10. The wave amplitude is therefore raised by an order of magnitude. As a consequence the emission process is expected to be represented by a signal of effective amplitude below  $10^{-13} \text{ m}$  and typical frequency about 1 Hz. The characteristic correlation time should be approximately an order of magnitude greater than  $t_0$  which shows that the signal has a broad frequency spectrum. Such a signal corresponds to a random noise which is practically impossible to differentiate from the background noise of currently available instrumentation.

#### 4. Conclusion

The aim of the article was to assess some characteristic properties of acoustic signal emitted during the smooth development of crazes. The estimated parameters indicate that the corresponding signals fall into a frequency band which is several orders of magnitude below that of normal acoustic emission monitoring systems. Beside this the amplitude estimated by the consideration of craze development mechanism falls outside the region of detectability. These estimations explain why acoustic emission is so often absent during the smooth advancement of crazes but at the same time indicate that there must be a concurrent mechanism which is responsible for detectable acoustic emission in several cases, especially under aggressive environmental conditions<sup>7,8,9</sup>). Presumably only cracking of crazed polymer could be considered for the explanation of this phenomenon.

#### References

- 1) R. P. Kambour, *J. Polym. Sci., Macromol. Rev.* **7** (1973) 1;
- 2) E. J. Kramar, *Development of Polymer Fracture 1*, Ch. 3, Edited by E. H. Andrews, Appl. Sci. Pub. Ltd, 1978;
- 3) B. D. Lauterwasser and E. J. Kramar, *Phil. Mag.* **39** (1979) 469;
- 4) A. M. Donald and E. J. Kramar, *Phil. Mag.* **43** (1981) 857;
- 5) A. S. Argon and M. M. Salama, *Mat. Sci. Eng.* **23** (1976) 219; *Phil. Mag.* **36** (1977) 1217;
- 6) R. J. Fields and M. F. Ashby, *Phil. Mag.* **33** (1976) 33;
- 7) I. Grabec and A. Peterlin, *J. Polym. Sci., Polym. Phys. Ed.* **14** (1976) 651;
- 8) K. Jakus, J. E. J. R. Ritter and C. A. Larsen, *Polym. Eng. Sci.* **21** (1981) 854;
- 9) T. Nishiura, T. Joh, S. Okuda and M. Miki, *Polym. J.* **13** (1981) 89; **13** (1981) 611;
- 10) I. Grabec, *Fortschritte der Akustik, FASE/DAGA, Göttingen 1982*, p. 696, 1982;
- 11) A. Peterlin, *AE of Polymers under Tensile Load*, in *Adv. in Chem. Series No 174*, 1979, — *Probing Polym. Structures*, Ed. J. L. Koenig.

### RAZVOJ VLAKNASTIH RAZPOK V POLIMERU IN AKUSTIČNA EMISIJA

IGOR GRABEC

*Fakulteta za strojništvo, pp. 394, 61000 Ljubljana.*

UDK 541.64

Tvorba vlaken v konici vlaknaste razpoke je postopni proces, ki naj bi deloval kot izvor zvoka. V članku je uporabljen mehanizem meniskusne nestabilnosti za ocenjevanje karakteristične frekvence in amplitude ustreznega signala akustične emisije. Ocenjena vrednost frekvence (tipično okoli 1 Hz v blagih pogojih okolja) leži daleč pod frekvenčnim pasom komercialnih detektorjev akustične emisije. Ocenjena valovna amplituda (tipično reda velikosti  $10^{-14}$  m) nakazuje skupaj s široko spektralno širino, da so ustrezni signali akustične emisije normalno skriti v šumu okolja in inštrumentarija. To pojasnjuje nezaznavnost akustične emisije med počasnim razvojem vlaknastih razpok in hkrati nakazuje, da je potrebno zaznavno akustično emisijo jemati na osnovi skokovitega pokanja polimera, ki sledi razvoju vlaknastih