JOURNAL OF MATERIALS SCIENCE 19 (1984) 3271–3275

Shear band formation in polycarbonate–glass bead composites

M. E. J. DEKKERS, D. HEIKENS
Eindhoven University of Technology, Laboratory of Polymer Technology, PO Box 513, 5600 MB Eindhoven, The Netherlands

The shear band formation at glass beads embedded in a polycarbonate matrix subjected to a uniaxial tension has been investigated by microscopic *in situ* observation. The degree of interfacial adhesion was varied by different glass surface treatments. To gain insight into the three-dimensional stress field requirement for shear band formation, the distributions of several elastic failure criteria around an isolated adhering glass sphere in a polycarbonate matrix have been computed with the aid of finite element analysis. It was found that the mechanism for shear band formation is fundamentally different for adhering and non-adhering glass beads. In the case of excellent interfacial adhesion, the shear bands form near the surface of the bead in regions of maximum principal shear stress and of maximum distortion strain energy. In the case of poor interfacial adhesion, shear band formation is preceded by dewetting along the interface between bead and matrix.

1. Introduction

Shear deformation in glassy polymers takes place by co-operative movement of molecular segments without loss of intermolecular cohesion. Many details concerning shear deformation and shear yielding are given in a number of reviews [1, 2, 3]. Shear processes may be diffuse or localized into shear microbands. As pointed out by Bowden *et al.* [4, 5], the tendency towards shear band formation at the expense of diffuse shear deformation increases with the size of the strain inhomogeneities. Thus in a glassy polymer such as polycarbonate (PC), which at room temperature under tensile conditions deforms by diffuse shearing [6], shear bands can be generated by incorporation of artificial stress concentrating heterogeneities. In the present study small glass beads are used for this purpose. The mechanism for shear band formation at the glass beads is investigated by microscopic *in situ* observation in the course of a tensile test. From previous investigations [7, 8, 9] it is known that the degree of interfacial adhesion has a pronounced effect on the mechanism for craze formation in polystyrene–glass bead composites. Therefore special attention is paid to the effect of interfacial adhesion on the mechanism for shear band formation.

Another interesting feature is the criterion for shear band formation or, in other words, the kind of critical elastic limit that must be reached in order to start shear band formation. Several authors have attempted to formulate a criterion for shear yielding [1, 10, 11]. However, all those criteria are based on the macroscopic yield behaviour of polymers in mechanical tests and therefore do not refer directly to the microscopic shear processes that occur locally within the material. In this study the distributions of a number of simple elastic failure criteria around an isolated adhering glass sphere in a PC matrix are computed with the aid of finite element analysis. By examining the location at the adhering sphere at which the shear band originates during the tensile test, information is obtained about the three-dimensional stress field requirement for shear band formation on a microscopic level.

2. Experimental details

The PC used was Makrolon 2405 (Bayer). The glass beads (Tamson 31/20) have an average diam-

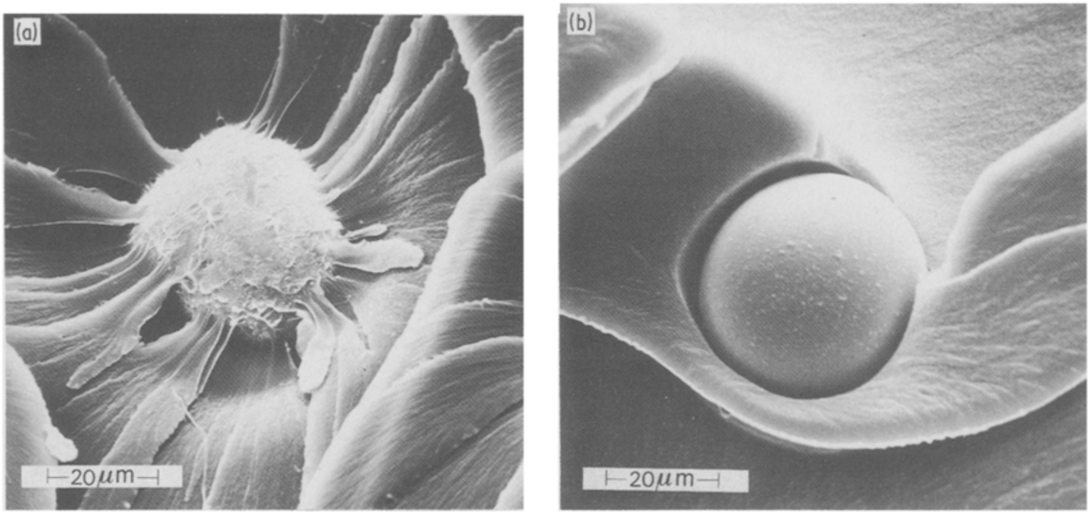


Figure 1 Scanning electron micrographs of fracture surfaces of PC-glass bead composites. (a) Excellent interfacial adhesion obtained with γ-aminopropylsilane; (b) Poor interfacial adhesion obtained with silicone oil.

eter of about 3×10^{-5} m. Composites were made containing 0.5 vol% of glass beads. Before being dispersed in PC, the glass beads were surface treated differently to obtain various degrees of interfacial adhesion. For excellent interfacial adhesion the beads were treated with γ-aminopropyltriethoxysilane (Union Carbide A-1100), for poor interfacial adhesion with a silicone oil (Dow Corning DC-200). Intermediate adhesion was obtained with untreated beads.

The surface treatments were executed as follows: the glass beads were first cleaned by refluxing isopropyl alcohol for 2 h and vacuum dried for 1 h at 130° C.

1. *γ-aminopropylsilane*: 75 g of refluxed glass was stirred for 1 h at room temperature in a 2% solution of silane in methanol containing 1% 2 M hydrochloric acid (200 ml in total). After this the glass was allowed to dry in air for 1 h and was then cured for 1 h at 100° C under vacuum.

2. *Silicone oil*: 100 g of refluxed glass was stirred for 3 h at room temperature in a 1% solution of silicone oil in toluene (200 ml in total). After this, the glass was dried for 1 h at 100° C under vacuum.

To avoid orientation effects, the composites were not prepared by injection moulding but by melt-mixing on a laboratory mill at 235° C. The hot crude mill sheets were compression moulded at 260° C. Dumb-bell shaped tensile specimens (narrow section 4 × 1.5 mm) were machined from the compression moulded sheets. To reduce thermal stresses the specimens were annealed at 80° C for 24 h and then conditioned at 20° C and 55% relative humidity for at least 48 h before testing.

The tensile tests were performed by straining the specimens uniaxially on a small tensile apparatus which was fitted to the stage of a Zeiss light microscope. In this way the shear band formation process at the glass beads could be followed *in situ* by continuous microscopic observation. At important stages of the tensile test, it was interrupted briefly to take a photograph with a camera fitted to the microscope.

In order to investigate the degree of interfacial adhesion between glass and PC, fracture surfaces of the specimens were examined with a Cambridge scanning electron microscope.

3. Results and discussion

3.1. Mechanism for shear band formation

The difference in PC–glass bead adhesion due to different glass surface treatments is demonstrated by the fracture surfaces in Fig. 1. The beads treated with γ-aminopropylsilane show excell interfacial adhesion, the beads treated with sili oil show poor interfacial adhesion. Intermed adhesion was obtained with untreated beads a will not be considered further.

The shear band pattern around an excellently adhering glass bead is shown in Fig. 2. In this case, during the tensile test the shear bands originate near the surface of the bead at about 45° from the

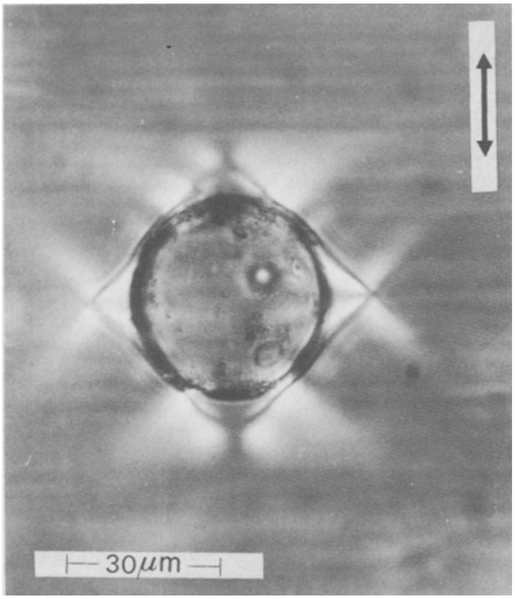


Figure 2 Light micrograph of the shear band pattern around an excellently adhering glass bead viewed between crossed polars. The arrow indicates the direction of applied strain.

poles defined by the symmetry axis of the stressed sphere. After this, the bands expand into the matrix at an angle of 45° to the tension direction. This inclination is consistent with the predictions of plasticity theory for isotropic materials deforming at constant volume [1].

It should be noted that, of course, the shear bands form axisymmetrically with respect to the polar axis. So, though visible as bands, there are actually two shear regions, both in the shape of the outline of a right circular cone.

Fig. 3 shows successive stages of the shear band formation process at a poorly adhering glass bead. In this case, the formation of shear bands is preceded by dewetting along the interface between bead and matrix. This is demonstrated by Fig. 3b which shows the situation shortly after the tensile test has started. The sickle-shaped shadow at the poles of the bead is the indication for a small cap-shaped cavity formed as a result of dewetting. A similar behaviour was reported previously for craze formation at a poorly adhering glass bead embedded in a polystyrene matrix [7, 8]. As the strain is further increased, the edge of the cavity shifts into the direction of the equator until, at a polar angle of about 60°, a shear band originates at the edge of the cavity (Fig. 3c). As with excellent interfacial adhesion, the shear bands expand into the matrix at an angle of 45° to the tension direction.

Summarizing, it appears that the degree of interfacial adhesion has a pronounced effect on the mechanism for shear band formation. In the case of excellent interfacial adhesion the shear bands form near the surface of the bead at a polar angle of about 45°. In the case of poor interfacial adhesion shear band formation is preceded by dewetting along the interface between bead and matrix.

3.2. Criterion for shear band formation

In order to gain insight into the three-dimensional stress field requirement for shear band formation on a microscopic level, the distributions of a number of simple elastic failure criteria around an isolated adhering glass sphere in a PC matrix were calculated. As the procedure followed in this study is the same as described previously [7], only some main points will be briefly discussed.

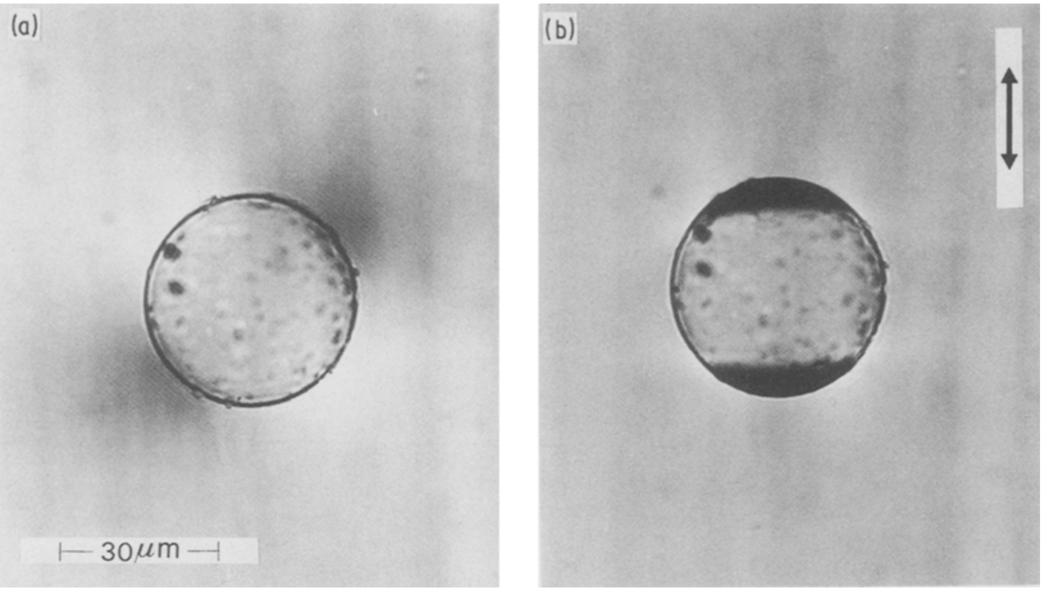
The three-dimensional stress situation around the sphere caused by uniaxial tension was computed with the aid of axisymmetric finite element analysis for spherically filled materials. The thermal shrinkage stresses around the sphere induced by cooling from the annealing temperature to room temperature (temperature difference 60° C) were computed using the equations derived by Beck *et al.* [12]. Superposition of the mechanical and thermal stresses yielded the three-dimensional stress situation with which the distributions of the failure criteria were calculated.

The method applied is based on perfect interfacial adhesion between sphere and matrix. Therefore the results of the analysis may only be compared with the situation of excellent interfacial adhesion between glass and PC.

The physical constants used for the calculations are listed in Table 1. The applied tension was taken at 25 MPa because at about this stress level shear deformation starts in PC–glass bead composites with excellent interfacial adhesion [6]. The maxi-

TABLE I Physical constants of the materials

Material	Young's modulus (MPa)	Poisson's ratio	Coefficient of thermal expansion (K^{-1})
Polycarbonate	2300	0.4	6.5×10^{-5}
Glass	70000	0.22	7×10^{-6}



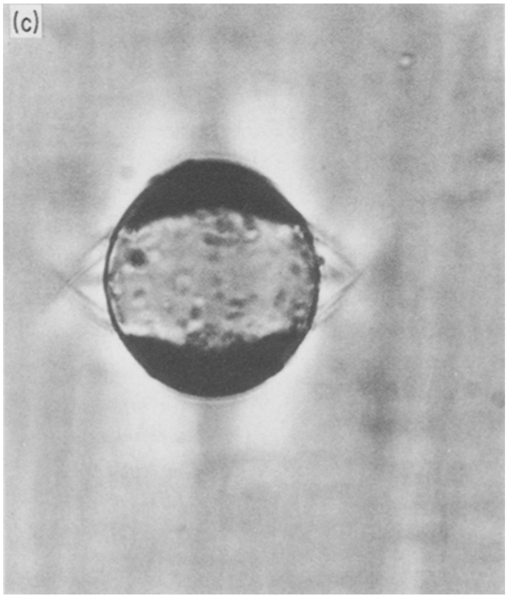


Figure 3 Light micrographs of successive stages of the shear band formation process at a poorly adhering glass bead. (a) Specimen before straining; (b) Dewetting; (c) Shear band formation. The arrow indicates the direction of applied strain. (crossed polars).

mum value of the radial thermal compressive stress was found to be 3.7 MPa.

Table 2 gives the expressions for the criteria under investigation in terms of the three principal stresses in the three-dimensional stress system. Fig. 4 shows the positions of the absolute maxima of these criteria marked in the unit cell of the system under analysis. It should be noted that the omission of thermal stresses from the calculations did not significantly change the positions of the maxima.

Microscopic observation *in situ* revealed that at an adhering glass bead shear bands form near the surface of the bead at a polar angle of about 45°. From Fig. 4 it appears that only the maxima of the principal shear stress and the distortion strain energy are located near this point. The maxima of the other criteria under investigation are clearly located at some distance from this point: maximum total strain energy occurs at a polar angle of 40° whereas maximum dilatation, maximum principal stress and maximum principal strain occur near the pole of the sphere. Thus at an adhering glass bead in a PC matrix the shear bands form in regions of maximum principal shear stress and of maximum distortion strain energy.

It should be realized that in the present study only a few simple criteria are considered. For macroscopic shear yielding more complicated criteria were proposed, e.g. the modified Tresca criterion [1] and the modified von Mises criterion [10]. These criteria contain, in addition to a shear stress term and a distortion strain energy term respectively, a dilatation term to account for the effect of hydrostatic pressure on the yield behaviour. (An increase in dilatation was found to result in a decrease in yield stress). In the present study such combinations could not be investigated since the relative contributions of the terms are unknown

TABLE II The expressions of the elastic failure criteria under investigation in terms of the three principal stresses $\sigma_1 > \sigma_2 > \sigma_3$. E and ν are, respectively, the Young's modulus and the Poisson's ratio of the matrix material

Criterion	Expression
Maximum principal stress, σ	σ_1
Maximum principal strain, ϵ	$\epsilon_1 = (1/E)[\sigma_1 - \nu(\sigma_2 + \sigma_3)]$
Maximum principal shear stress, τ (Tresca)	$\tau_1 = (1/2)(\sigma_1 - \sigma_3)$
Maximum dilatation, Δ	$\Delta = [(1 - 2\nu)/E](\sigma_1 + \sigma_2 + \sigma_3)$
Maximum total strain energy density, W_S	$W_S = (1/2E)[\sigma_1^2 + \sigma_2^2 + \sigma_3^2 - 2\nu(\sigma_1\sigma_2 + \sigma_1\sigma_3 + \sigma_2\sigma_3)]$
Maximum distortion strain energy density, W_D (von Mises)	$W_D = [(1 + \nu)/6E][(\sigma_1 - \sigma_2)^2 + (\sigma_2 - \sigma_3)^2 + (\sigma_3 - \sigma_1)^2]$

and can neither be determined by a simple uniaxial tensile test. Therefore a combination of dilatation with principal shear stress or distortion strain energy cannot be ruled out completely. In any case, on the basis of the fact that maximum dilatation clearly occurs at a point remote from that point at which the shear bands form, it can be concluded that principal shear stress and distortion strain energy play a dominant role in microscopic shear band formation.

4. Conclusions

From microscopic observation *in situ* it appears that the mechanism for shear band formation is fundamentally different for adhering and non-adhering glass beads. In the case of excellent interfacial adhesion the shear bands form near the surface of the bead at a polar angle of about 45°. In case of poor interfacial adhesion, shear band formation is preceded by dewetting along the interface between bead and matrix. The consequences of those different mechanisms on the mechanical behaviour of PC–glass bead composites will be reported in a subsequent paper [6].

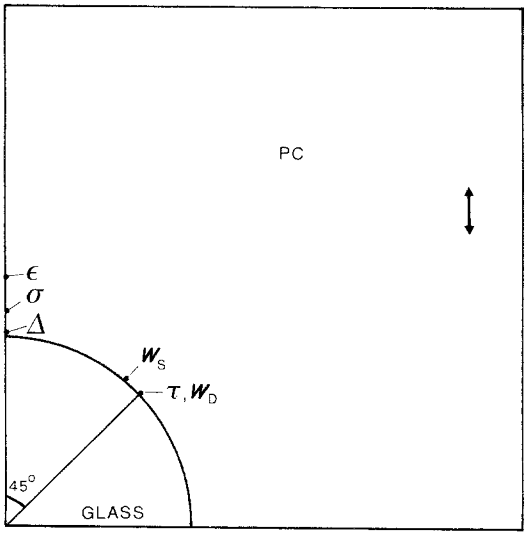


Figure 4 Unit cell of the analysed system with the positions of the absolute maxima of the following elastic failure criteria: principal stress (σ), principal strain (ϵ), principal shear stress (τ), dilatation (Δ), total strain energy density (W_S) and distortion strain energy density (W_D). The applied tension and the maximum thermal stress were assumed to be 25 and 3.7 MPa, respectively. The arrow indicates the applied strain direction.

From stress analysis it appears that microscopic shear band formation occurs in regions of maximum principal shear stress and of maximum distortion strain energy. Based on the results of the applied method, a choice between these two criteria cannot be made. The present study provides no direct indication that dilatation plays a role in shear band formation in PC–glass bead composites.

Acknowledgement

The assistance of H. C. B. Ladan in obtaining the scanning electron micrographs is gratefully acknowledged.

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*Received 29 November
and accepted 20 December 1983*