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Microscopic *in situ* observation of craze formation in polystyrene-glass bead composites

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When a glass bead-filled polystyrene (PS) sample is subjected to a uniaxial tension, crazes form at the stress concentrating glass beads. In a recent paper [1] the effect of interfacial adhesion on the mechanism for this craze formation was reported. The degree of interfacial adhesion was varied by using different silane coupling agents. Tensile specimens with a very low percentage of glass beads (average diameter = 3×10^{-5} m) were strained uniaxially on a tensile tester. Afterwards the craze patterns around the glass beads were examined with a light microscope. It was found that the degree of interfacial adhesion has consequences for the location near the surface of the glass bead at which the craze originates during the tensile test. In the case of excellent interfacial adhesion the craze forms near the pole of the bead. However, in the case of poor interfacial adhesion the craze forms at the interface between the pole and equator at a polar angle, θ , of about 60° . This indicates a mechanism for craze formation different from that of the excellent adhesion case. It was proposed that in the case of poor interfacial adhesion the formation of crazes is preceded by dewetting along the interface between the bead and matrix. In the present study this proposed mechanism is confirmed by means of microscopic *in situ* observation of the craze formation process in the course of a tensile test.

The experimental procedure to obtain unoriented PS-glass bead composites with poor interfacial adhesion has been described previously [1]. In the present study dumbbell-shaped specimens (narrow section 4 mm $\times$ 1.5 mm) were strained uniaxially on a small tensile apparatus which was fitted to the stage of a Zeiss light microscope. In this way the craze formation process at the poor adhering glass bead can be followed *in situ* by continuous microscopic observation. At any stage of the tensile test, it is

possible to interrupt the test briefly in order to take a photograph with a camera fitted to the microscope. Photographs of important successive stages are shown in Fig. 1.

Fig. 1a shows the poor adhering glass bead embedded in a PS-matrix before straining. Shortly after the tensile test is started, a sickle-shaped shadow appears at the poles of the bead indicating that, indeed, dewetting takes place and a small cap-shaped cavity is formed (Fig. 1b). By continuing the tensile test the edge of the cavity shifts into the direction of the equator until, at a polar angle, θ , of about 60° , a craze originates at the edge of the cavity (Fig. 1c).

As a result of dewetting the stress situation around the glass bead changes. As a consequence of this the craze does not only originate at a location different to the case of excellent interfacial adhesion, but also expands into the matrix in a direction deviating initially from the direction perpendicular to the applied tension. Only at some certain distance from the bead, where the propagating craze tip leaves the "sphere of influence" of the bead, the craze bends toward this direction, as is clearly visible in Fig. 2b of [1]. Such curvilinear crazes were also found by Sternstein *et al.* [2] around a hole in a thin polymethyl methacrylate sheet. Based on stress field calculations around the hole, they concluded that in structurally isotropic glassy polymers real craze growth occurs along a path such that the major principal stress always acts perpendicular to the craze plane. This implies that near the point where the craze originates at the poorly adhering bead the directions of the major principal stress and the applied tension do not coincide.

Finally Fig. 1d shows the situation after removal of the applied strain. The shadows of the cavities have disappeared and the bead and matrix touch each other again. Only the two crazes remain visible.

Figure 1 Successive stages of the craze formation process at a poorly adhering glass bead. (a) Specimen before straining; (b) dewetting; (c) craze formation; (d) craze pattern after removal of the applied strain. The arrow indicates the applied strain direction. Note that, besides at the glass bead, crazes are also formed at surface flaws. Two small surface crazes are clearly visible in Figs. 1c and d at the left side of the bead near the equator. These crazes were observed to grow from the surface of the specimen into the material where they reached the bead.

In conclusion, it can be stated that microscopic *in situ* observation of the craze formation process at a poor adhering glass bead provides conclusive evidence for the mechanism proposed previously: craze formation is preceded by dewetting along the interface between the bead and matrix.

References

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