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Cavitation in model elastomeric composites

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Layers of transparent silicone rubber were bonded between two steel spheres or between two parallel steel cylinders, to make simple mechanical models of particle-filled and fibre reinforced elastomers. When the steel end-pieces were pulled apart, visible cavities appeared suddenly in the rubber layer between them, at well-defined tensile loads and displacements. The critical conditions for cavity formation are shown to be in good agreement with a theoretical criterion for the unbounded elastic expansion of a microscopic precursor void within the rubber: that the local triaxial tensile stress attains a value of $5E/6$, where E is Young's modulus for the rubber. When the rubber layer was extremely thin, however, less than about 5% of the steel end-piece diameter, then the stress required to form a cavity was greater than this, and it increased rapidly as the rubber thickness was reduced further.

1. Introduction

Composite materials show internal cracking when a sufficiently large stress is applied. In order to elucidate the mechanism of cracking, a number of studies have been carried out using simple models, in which a stiffer inclusion is incorporated into an elastic solid and the onset of failure near or at the surface of the inclusion is examined. Such studies have included thoria spheres embedded in a glass matrix [1], glass spheres embedded in a glassy polymer matrix [2, 3], and glass and steel spheres embedded in a rubber matrix [4, 5].

Variations in the strength of interfacial adhesion have been shown to affect not only the magnitude of the critical applied stress at which failure initiates, but also the nature of the failure itself [2, 5]. For a weakly-bonded rigid sphere in a rubber matrix, sudden detachment at the poles is observed when the applied stress reaches a sufficiently large value. On the other hand, when the rubber is well-bonded to the inclusion, a characteristic internal failure, termed cavitation, takes place near the poles of the inclusion, in the direction of the applied tension [4, 5]. This process consists of the sudden appearance of a void within the rubber itself, close to the surface of the inclusion but separated from it by a thin layer of still-attached rubber. It has been attributed to elastic expansion of a microscopic precursor void under the action of the local dilatant stress (negative hydrostatic pressure) $-P$ until the maximum extensibility of the material is exceeded and the void then grows to a visible size by tearing [6]. This hypothetical mechanism of cavity formation in elastomers has been shown to account quantitatively for the appearance of cavities under triaxial tensions [6], under the action of dissolved supersaturated gases [7], and at points near spherical and rodlike inclusions where a sufficiently large triaxial tension is set up by an applied far-field tensile stress [4, 5, 8].

Unbounded elastic expansion of a spherical cavity in a rubber block is predicted to occur when the local dilatant stress exceeds a critical value, given by

$$-P_c = 5E/6 \qquad (1)$$

where E is Young's modulus of the rubber [6]. Good agreement is generally obtained with experimentally-observed conditions for the formation of visible cavities in soft rubbery solids.

One exception must be noted. When a rigid spherical inclusion is present, and is small in size, the critical far-field tensile stress for cavity formation is found to be considerably larger than that calculated from Equation 1, and it increases steadily as the diameter of the inclusion is reduced [5]. This anomaly is tentatively attributed to a second feature of the cavitation process: when the volume of material subjected to the dilatant stress is extremely small, say less than about 10^{-15} m^3, then the probability of finding a relatively large precursor void within it is also small. And when the precursor void is less than about 10^{-7} m in diameter, an additional restraint on its expansion becomes significant, arising from its own surface energy, so that cavitation becomes more difficult on this account [9].

We now turn to another aspect of internal void formation, and that is the effect of the close proximity of two inclusions upon the occurrence of voids between them. Cavities appear midway between two closely-spaced spherical inclusions lying in the direction of the applied tension [5], presumably when the dilatant stress set up there is sufficiently large. Simple models have therefore been constructed to represent highly-filled composites. They consist of two steel spheres or two parallel steel cylinders, bonded together with a layer of strongly-adhering transparent silicone rubber between them. They have been subjected to tensile forces in the direction of the two steel end-pieces, large enough to induce cavitation in the rubber phase. Results for the critical loads and deflections are reported here and compared with the predictions of Equation 1, using a simple approximate solution for the dilatant stress set up in a layer of an incompressible elastic material bonded between two rigid spheres or

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two rigid parallel cylinders, when it is subjected to a tensile load in the direction of their centres.

2. Theoretical considerations

Values of the maximum hydrostatic tension $-P$ set up in the rubber layer can be obtained from an approximate stress analysis, assuming that the rubber is an incompressible, linearly-elastic solid. The results take the form [10],

$$-P_m/E = e_m/4A(A-1)^2 \quad (2)$$

for a layer bonded between two rigid spheres, Fig. 1a, and

$$-P_m/E = e_m/2A(A-1)^2 \quad (3)$$

for a layer bonded between two parallel rigid cylinders, Fig. 1b. The term A denotes

$$A = 1 + (h/D) \quad (4)$$

where h is the separation distance of the spheres or cylinders and D is their diameter, and e_m is the maximum tensile strain set up in the rubber, given by the ratio of the displacement δ of one sphere or cylinder away from the other to the initial separation h: $e_m = \delta/h$.

The relations between e_m and the applied force F are somewhat complex; they are given in the original paper [10]. However, they are not very different from the simple results:

$$e_m = \sigma/E \quad (5)$$

for layers between spherical end-pieces, and,

$$e_m = 3\sigma/4E \quad (6)$$

for layers between cylindrical end-pieces, where σ denotes the mean applied stress [10]. With spherical end-pieces close together the observed and calculated strains e_m are somewhat larger than predicted by Equation 5 and with closely-spaced cylindrical end-pieces they are smaller than predicted by Equation 6, by a factor of up to 2× [10].

3. Experimental details

3.1. Preparation of test-pieces

Four sizes of steel balls were used to construct test-pieces having spherical end-pieces, Fig. 1a. The diameters were 6.35, 9.50, 18.8 and 49.3 mm. Thin steel rods were welded to the outer poles of the balls to hold them in a stretching device subsequently. Stainless steel tubes having an outer diameter of 9.55 mm, and of varied lengths in the range 12.5 to 50 mm, were used to construct test-pieces having cylindrical end-pieces, Fig. 1b.

Surfaces of the steel balls and tubes were polished with fine emery paper and then coated with a special primer (Primer 92-023, Dow Corning Corporation) to give good adhesion to the silicone elastomer used. After the primer coating had dried, rubber layers were cast in the gap between two identical steel spheres or two identical cylinders using a mixture of 100 parts of Sylgard S-184 silicone polymer and six parts of Sylgard C-184 curing agent, both of which were obtained from Dow Corning Corporation. The mixture was degassed under vacuum for 30 min and then poured into a mould surrounding the metal end-pieces and cured for 12 h at 120° C. Tensile measurements on a cured slab of the same formulation gave a value for Young's modulus E of 0.96 ± 0.05 MPa.

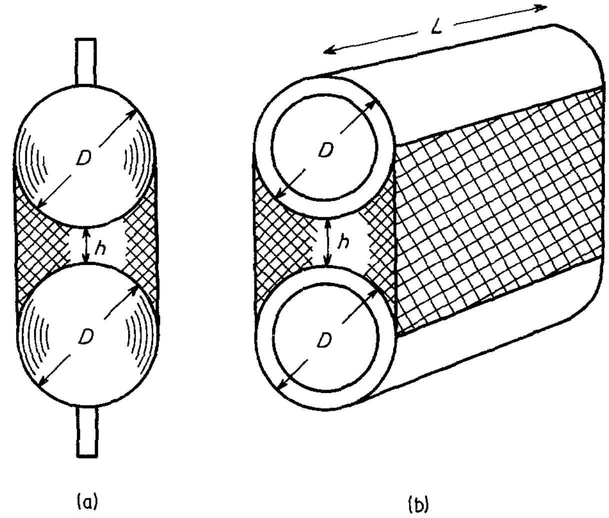

Figure 1 (a) Rubber layer (cross-hatched) bonded between two steel spheres. (b) Rubber layer (cross-hatched) bonded between two parallel steel tubes.

Specimens were prepared with various spacings h so that the ratio h/D ranged from about 0.02 up to about 0.35.

3.2. Measurement of applied forces or displacements at which cavitation occurred

Specimens with spherical end-pieces were stretched under an optical projector having a magnification of about 50×. The displacement δ at which a cavity was seen to appear suddenly in the rubber layer was measured directly in this way and employed using Equation 2 to calculate the corresponding value of the hydrostatic tension $-P_m$.

Some experimentally-determined relations between tensile load, represented by the mean applied stress σ, and the corresponding displacement δ, are shown in Fig. 2. They are seen to be approximately linear, up to maximum tensile strains of about 80%, and values of the tensile stiffness K obtained from their slopes were

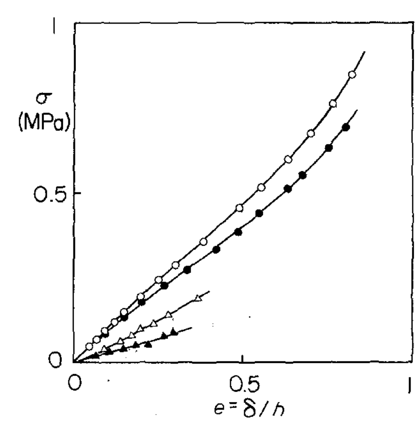

Figure 2 Experimental relations between mean tensile stress σ and ratio e of displacement δ to initial separation h for rubber layers bonded between two steel spheres. (○) $h/D = \infty$, (●) 0.34, (△) 0.058, (▲) 0.015.

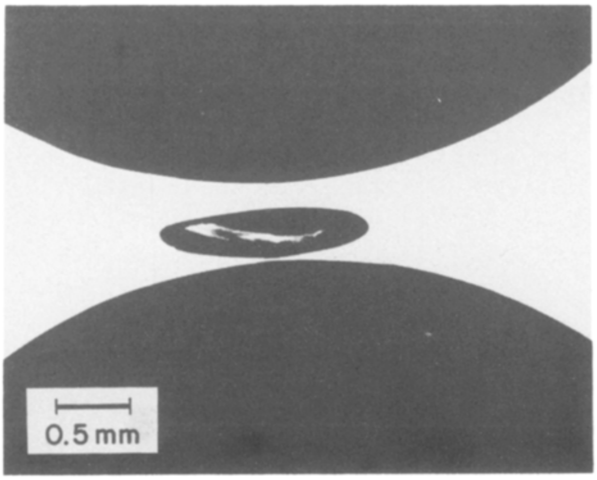

Figure 3 Cavity formed in a silicone rubber layer bonded between two steel spheres at a tensile strain $e_m = 0.23$. Initial separation $h = 0.43$ mm, diameter $D = 9.50$ mm.

TABLE I Effect of separation distance h on the cavitation strain e_m and dilatant stress $-P_m$ for specimens with spherical end-pieces of diameter D

D (mm)	h (mm)	h/D	δ (mm)	e_m	$-P_m/E$
6.34	0.95	0.150	0.87	0.92	1.33
	1.33	0.210	1.52	1.14	1.12
	1.93	0.304	1.95	1.01	0.64
9.51	0.19	0.020	0.05	0.26	3.22
	0.22	0.023	0.06	0.27	2.91
	0.37	0.039	0.07	0.19	1.16
	0.38	0.040	0.11	0.29	1.74
	0.43	0.045	0.10	0.23	1.24
	0.74	0.078	0.25	0.34	1.00
18.80	0.27	0.014	0.04	0.15	2.68
	0.73	0.039	0.22	0.30	1.85
49.30	0.69	0.014	0.14	0.20	3.58
	0.84	0.017	0.21	0.25	3.50
	2.51	0.051	0.76	0.30	1.40
	2.93	0.058	1.02	0.35	1.46
	16.96	0.335	18.32	1.08	0.62

in good agreement with those obtained previously for similar test-pieces subjected to small compressions [10]. Thus, the assumption of linear elastic behaviour appears to hold reasonably well for these specimens, even up to moderately high tensile strains.

For specimens with cylindrical end-pieces, the critical loads at which a visible cavity suddenly appeared were measured directly using a tensile test apparatus. The specimens were stretched at a rate of about $10\,\mu m\,sec^{-1}$, until a cavity appeared, and the corresponding displacements were then calculated from the measured critical loads using theoretical stiffness values for such test-pieces [10]. The corresponding values of hydrostatic tension $-P_m$ were obtained from Equation 3.

It should be noted that only stresses arising from the restraints at the bonded surfaces are considered in this procedure for determining the triaxial tensile stress $-P_m$; simple tensile stresses are neglected. They should be relatively small, however, for specimens with closely-spaced end-pieces.

4. Experimental results

4.1. Formation of cavities

Photographs of representative cavities are shown in Figs 3 and 4, for specimens with spherical and cylindrical end-pieces, respectively. The cavities appeared suddenly when the critical load was reached and grew rapidly to a large size. They formed in the general area of maximum tensile strain and maximum hydrostatic tension, sometimes near one of the end-pieces, Fig. 3, and sometimes midway between them, Fig. 4. Similar observations were reported previously for a rubber block containing two spherical inclusions [5].

It was noticed that the formation of cavities was somewhat time-dependent; that is, when a load slightly smaller than the value taken here to be the critical one was applied and maintained for several seconds, a cavity would often appear in the same way as in steadily-increasing loading. This feature is tentatively ascribed to time dependence of the tensile modulus and tear strength of the silicone rubber used.

4.2. Conditions for cavitation

Values of the critical dilatant stress $-P_m$ for cavity formation, determined as described above from the measured critical loads or displacements, are given in Tables I and II for specimens with spherical and cylindrical end-pieces, respectively. They are also plotted in Figs 5 and 6 against the ratio h/D of the end-piece separation to diameter of the sphere or cylinder. The horizontal broken lines in each of these figures represent the theoretically-predicted result, Equation 1.

Experimental results for rubber layer thicknesses greater than about 5% of the end-piece diameter are seen to be in reasonably good agreement with the theoretical cavitation stress for a highly elastic solid containing a precursor void. This is the case for layers bonded to either spherical or cylindrical end-pieces and for a wide range of end-piece diameters. As the relations for dilatant stress are rather different in these two cases, Equations 2 and 3, the general agreement

Figure 4 Cavity formed in a silicone rubber layer bonded between two steel cylinders at a tensile strain $e_m = 0.08$. Initial separation $h = 0.43$ mm, diameter $D = 9.55$ mm.

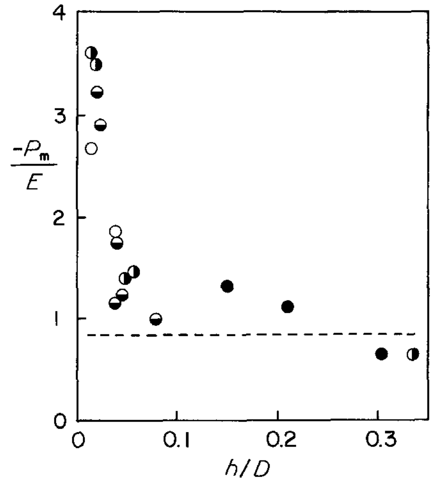

Figure 5 Dilatant stress $-P_m$ for cavity formation in a silicone rubber layer bonded between two steel spheres against ratio h/D of initial separation distance h to sphere diameter D. (●) $D = 6.35$ mm, (◒) 9.5 mm, (○) 18.8 mm, (◑) 49.3 mm. The horizontal broken line represents the predicted result from Equation 1.

found to hold strongly suggests that the observed cavities arise from the proposed mechanism of unstable elastic expansion of precursor voids under the action of a critical dilatant stress.

At extremely small thicknesses of the rubber layer, less than about 5% of the end-piece diameter, the stresses necessary to form a large cavity became considerably greater than those for thicker rubber layers, Fig. 5, as much as four times greater when the layer thickness was only about 2% of the end-piece diameter. This feature was found to hold for specimens with spherical end-pieces having a wide range of diameter, suggesting that it was not simply a function of the layer thickness itself but of the ratio of thickness to end-piece diameter. It is reminiscent of the previous observation that cavitation is more difficult to bring about near small spherical inclusions [5]. Once again, it appears that an additional restraint on cavity formation is operative in small volumes of rubber near highly-curved surfaces, in addition to the simple elastic resistance that governs the process in larger samples.

TABLE II Effect of separation distance h on the cavitation strain e_m and dilatant stress $-P_m$ for specimens with cylindrical end-pieces of diameter $D = 9.55$ mm

Length L (mm)	h (mm)	h/D	F (N)	δ (mm)*	e_m*	$-P_m/E$†
12.5	0.43	0.045	25	0.034	0.08	0.84
	0.91	0.095	66	0.255	0.28	1.35
	1.21	0.127	77	0.440	0.36	1.27
25.5	1.00	0.105	114	0.161	0.161	0.70
	1.36	0.142	122	0.395	0.29	0.90
	1.51	0.158	127	0.476	0.32	0.86
50.0	0.85	0.089	170	0.150	0.18	0.91
	0.95	0.099	206	0.209	0.22	1.02
	1.16	0.121	212	0.286	0.25	0.90
	1.24	0.130	216	0.317	0.26	0.87

*Calculated from F.
†Calculated from e_m using Equation 3.

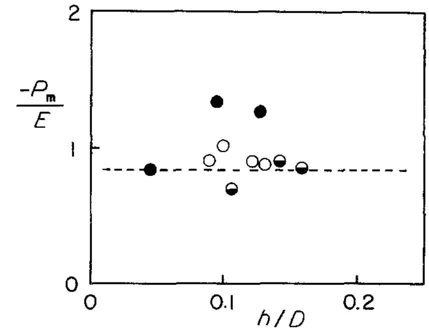

Figure 6 Dilatant stress $-P_m$ for cavity formation in a silicone rubber layer bonded between two parallel steel cylinders against ratio h/D of initial separation distance h to cylinder diameter D. $D = 9.55$ mm, L = (●) 12.5 mm, (○) 25.5 mm, (◒) 50 mm. The horizontal broken line represents the predicted result from Equation 1.

5. Conclusions

Large cavities have been found to form in layers of rubber bonded between rigid spheres or cylinders when the assembly is put into tension. The critical tensile loads and deflections are in generally good agreement with a simple fracture criterion: that a critical level of the local dilatant stress $-P$, of about $5E/6$, is reached. This is the theoretical value at which unbounded elastic expansion of a (hypothetical) precursor void would take place. Thus, it appears that visible cavities occur as a result of such a process, and form when and where the critical dilatant stress level is attained. However, when the rubber layer is extremely thin, less than about 5% of the end-piece diameter, then cavitation requires substantially higher stresses. For a regular arrangement of spheres on a cubic lattice, this close proximity corresponds to a high volume concentration of filler particles, of more than 45%.

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