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Contract: N00014-85-K-0222

Work Unit: 4327-555

Scientific Officer: Dr. Richard S. Miller

Technical Report No. 11

CAVITATION IN MODEL ELASTOMERIC COMPOSITES

by

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October, 1987

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REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER Technical Report No. 11	2. GOVT ACCESSION NO. 11-10-111	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) Cavitation in Model Elastomeric Composites	5. TYPE OF REPORT & PERIOD COVERED Technical Report	
	6. PERFORMING ORG. REPORT NUMBER	
7. AUTHOR(s) K. Cho and A. N. Gent	8. CONTRACT OR GRANT NUMBER(s) N00014-85-K-0222	
9. PERFORMING ORGANIZATION NAME AND ADDRESS Institute of Polymer Science The University of Akron Akron, Ohio 44325	10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS 4327-555	
11. CONTROLLING OFFICE NAME AND ADDRESS Office of Naval Research Power Program Arlington, VA 22217-5000	12. REPORT DATE October 1987	
	13. NUMBER OF PAGES 29	
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)	15. SECURITY CLASS. (of this report) Unclassified	
	15a. DECLASSIFICATION/DOWNGRADING SCHEDULE	
16. DISTRIBUTION STATEMENT (of this Report) According to attached distribution list. Approved for public release; distribution unrestricted.		
17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)		
18. SUPPLEMENTARY NOTES Submitted for publication in: Journal of Materials Science		
19. KEY WORDS (Continue on reverse side if necessary and identify by block number) Adhesion, Bonding, Cavitation, Composites, Elastomers, Fracture, Inclusions, Strength, Triaxial Stress		
20. ABSTRACT (Continue on reverse side if necessary and identify by block number) 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 fiber 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 per cent of the steel endpiece diameter, then the stress required to form a cavity was greater than this, and it increased rapidly as the rubber thickness was reduced further. (Key words: )

## 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



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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

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

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  $10^{-15} \text{m}^3$ , 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 two rigid parallel cylinders, when it is subjected to a tensile load in the direction of their centers.

## 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, Figure 1a, and

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

for a layer bonded between two parallel rigid cylinders, Figure 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  $\delta$  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  $\underline{\sigma}$  denotes the mean applied stress (10). With spherical end-pieces close together the observed and calculated strains  $\underline{e}_m$  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 2X (10).

### 3. Experimental details

#### (i) Preparation of test-pieces.

Four sizes of steel balls were used to construct test-pieces having spherical end-pieces, Figure 1a. The diameters were 6.35 mm, 9.50 mm, 18.8 mm 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 mm to 50 mm, were used to construct test-pieces having cylindrical end-pieces, Figure 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 6 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 mold 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  $\underline{E}$  of  $0.96 \pm 0.05$  MPa.

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

- (ii) 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 50X. The displacement  $\underline{\delta}$  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  $\underline{-P_m}$ .

Some experimentally-determined relations between tensile load, represented by the mean applied stress  $\underline{\sigma}$ , and the corresponding displacement  $\underline{\delta}$ , are shown in Figure 2. They are seen to be approximately linear, up to maximum tensile strains of about 80 per cent, and values of the tensile stiffness  $\underline{K}$  obtained from their slopes were in good agreement with those obtained previously for similar test-pieces subjected to small compressions (10). Thus, the assumption of linear elastic behavior 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  $10 \mu\text{m s}^{-1}$ , 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

##### (i) Formation of cavities

Photographs of representative cavities are shown in Figures 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, Figure 3, and sometimes midway between them, Figure 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.

(ii) Conditions for cavitation

Values of the critical dilatant stress  $\underline{-P_m}$  for cavity formation, determined as described above from the measured critical loads or displacements, are given in Tables 1 and 2 for specimens with spherical and cylindrical end-pieces, respectively. They are also plotted in Figures 5 and 6 against the ratio  $\underline{h/D}$  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 per cent 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 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 per cent of the end-piece diameter, the stresses necessary to form a large cavity became considerably greater than those for thicker rubber layers, Figure 5, as much as four times greater when the layer thickness was only about 2 per cent 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.

##### 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  $\underline{P}$ , of about  $\underline{5E/6}$ , 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 per cent 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 per cent.

#### Acknowledgements

This work forms part of a program of research on highly-filled elastomeric composites supported by the Office of Naval Research (Contract N00014-85-K-0222). A grant-in-aid from Lord Corporation is also gratefully acknowledged.

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Table 1: Effect of separation distance  $h$  on the cavitation strain  $e_m$  and dilatant stress  $-P_m$  for specimens with spherical end-pieces of diameter  $D$ .

<u>D (mm)</u>	<u>h (mm)</u>	<u>h/D</u>	<u><math>\delta</math> (mm)</u>	<u><math>e_m</math></u>	<u><math>-P_m/E</math></u>
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

Table 2. Effect of separation distance  $\underline{h}$  on the cavitation strain  $\underline{e}_m$  and dilatant stress  $\underline{-P}_m$  for specimens with cylindrical end-pieces of diameter  $\underline{D} = 9.55$  mm.

<u>Length L</u> (mm)	<u>h</u> (mm)	<u>h/D</u>	<u>F</u> (N)	<u><math>\dot{\epsilon}</math></u> (mm) <sup>a</sup>	<u><math>\underline{e}_m</math></u> <sup>a</sup>	<u><math>\underline{-P}_m / E</math></u> <sup>b</sup>
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

a: Calculated from  $\underline{F}$ .

b: Calculated from  $\underline{e}_m$  using equation 3.



Figure Captions

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

Figure 2: Experimental relations between mean tensile stress  $\underline{\sigma}$  and ratio  $\underline{e}$  of displacement  $\underline{\delta}$  to initial separation  $\underline{h}$  for rubber layers bonded between two steel spheres.

Figure 3: Cavity formed in a silicone rubber layer bonded between two steel spheres at a tensile strain  $\underline{e}_m = 0.23$ . Initial separation  $\underline{h} = 0.43$  mm, diameter  $\underline{D} = 9.50$  mm.

Figure 4: Cavity formed in a silicone rubber layer bonded between two steel cylinders at a tensile strain  $\underline{e}_m = 0.08$ . Initial separation  $\underline{h} = 0.43$  mm, diameter  $\underline{D} = 9.55$  mm.

Figure 5: Dilatant stress  $\underline{-P}_m$  for cavity formation in a silicone rubber layer bonded between two steel spheres vs ratio  $\underline{h}/\underline{D}$  of initial separation distance  $\underline{h}$  to sphere diameter  $\underline{D}$ .  $\underline{D} = 6.35$  mm, ● ; 9.5 mm, ⊖ ; 18.8 mm, ○ ; 49.3 mm, ⊕ . The horizontal broken line represents the predicted result from equation 1.

Figure 6: Dilatant stress  $-P_m$  for cavity formation in a silicone rubber layer bonded between two parallel steel cylinders vs 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.

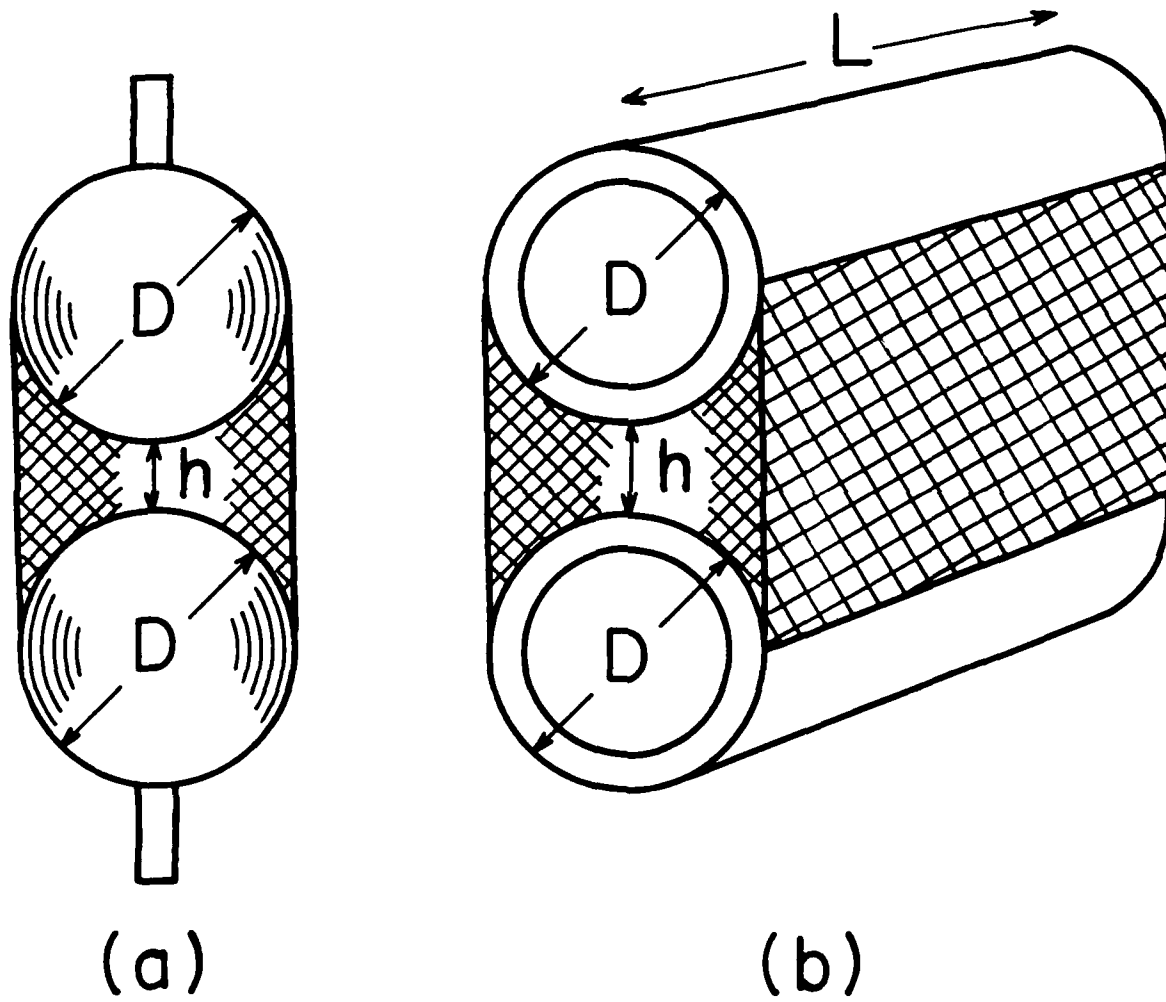


FIGURE 1

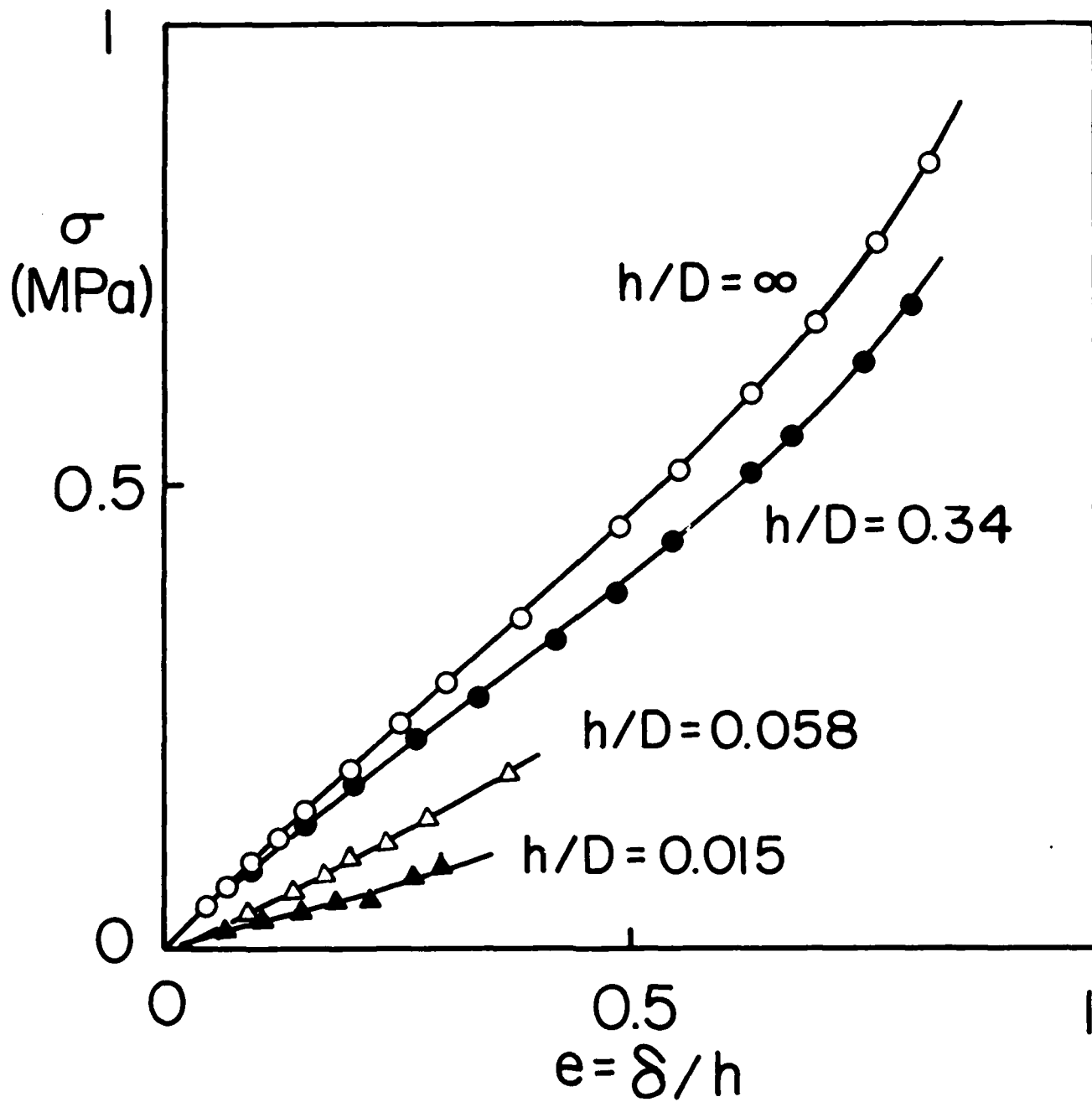


FIGURE 2

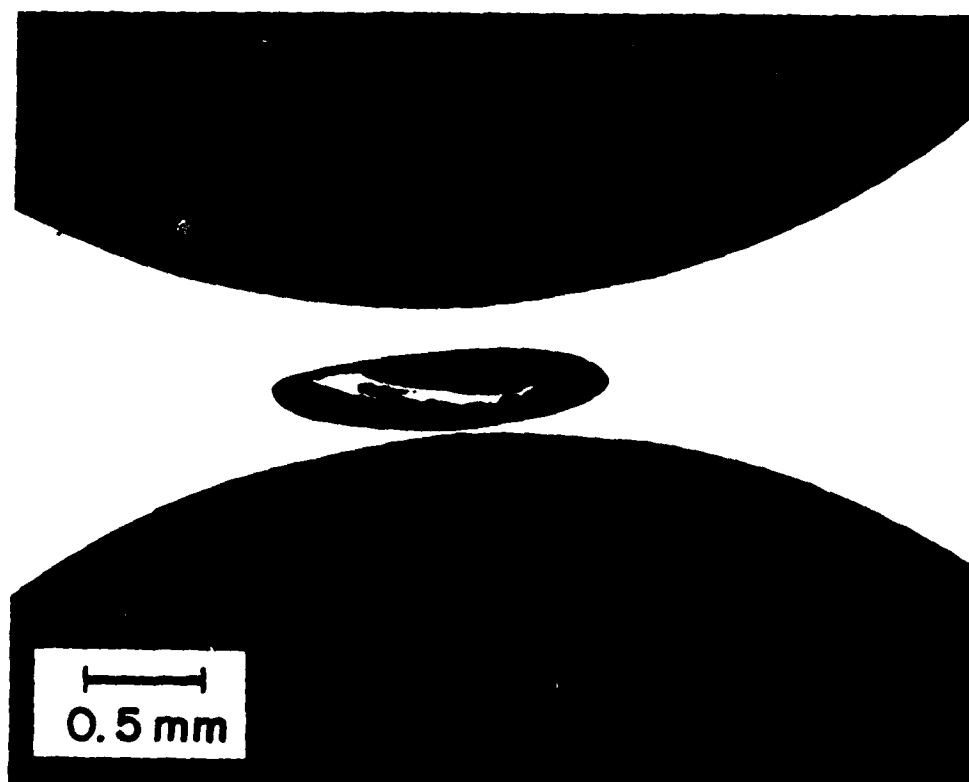


FIGURE 3

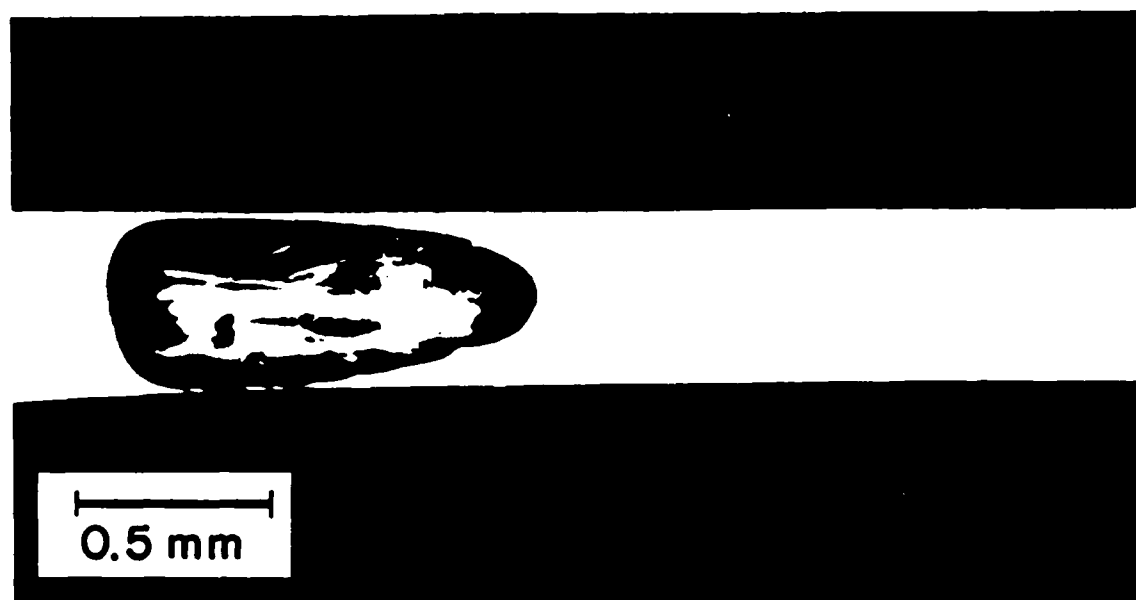


FIGURE 4

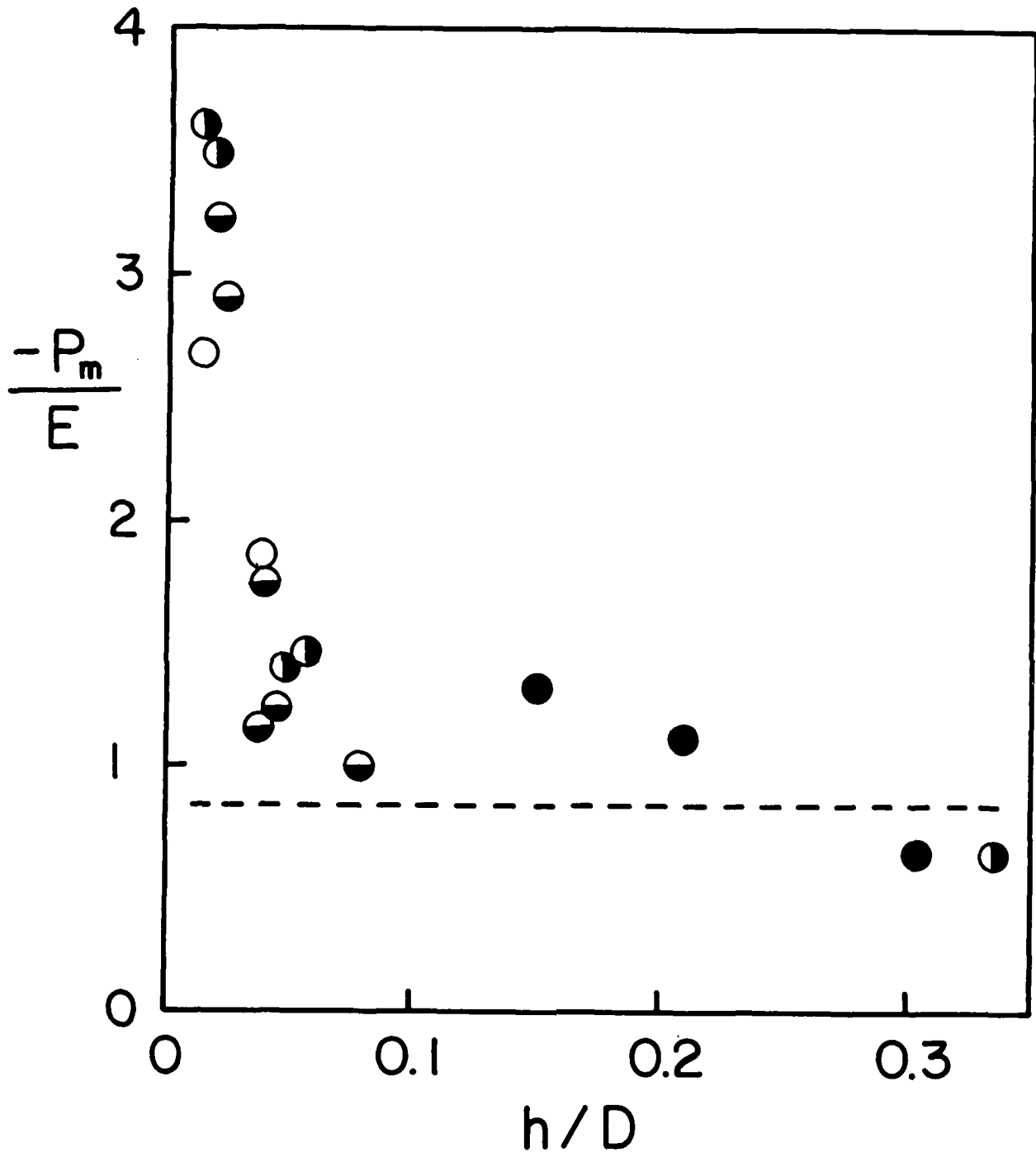


FIGURE 5

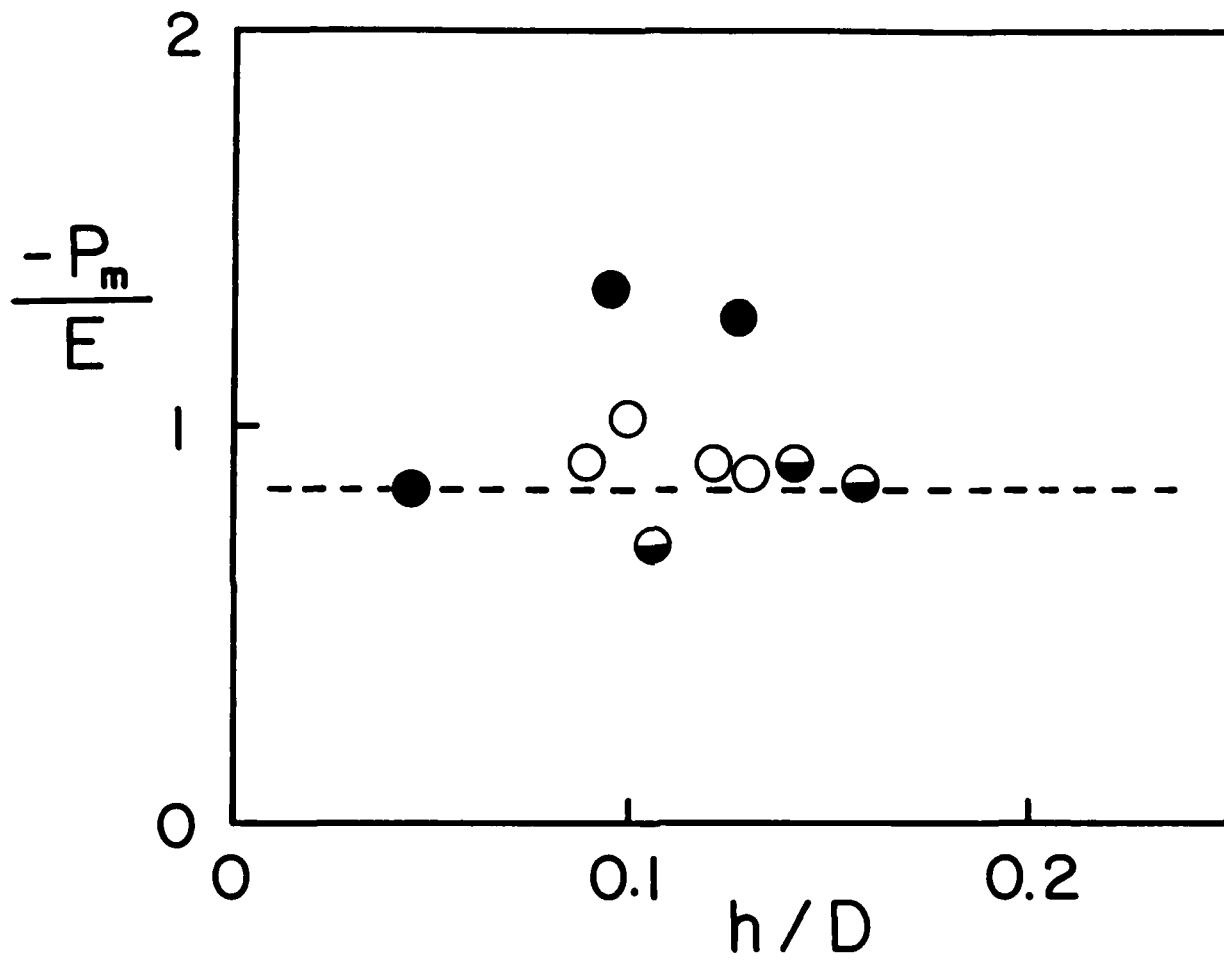


FIGURE 6



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