GREN-DE 70-1466

THE PRESSURE DEPENDENCE OF THE ELASTIC CONSTANTS OF DENTAL AMALGAM*

by

Dale E. Grenoble† and J. Lawrence Katz
Laboratory for Crystallographic Research
Department of Physics and Astronomy
Rensselaer Polytechnic Institute
Troy, New York 12181

*Contribution #40: This investigation was supported by U.S.P.H.S. Training Grant No. 5TI-DE-0117-07 and Research Grant No. 5PO1-DE-2336-03 from the National Institute of Dental Research, N.I.H., and by the National Aeronautics and Space Administration through Rensselaer's Interdisciplinary Materials Research Center.

†Present address: Department of Dental Materials, School of Dentistry, University of Southern California, Los Angeles, California 90007. This paper is based on a thesis submitted to the Department of Physics and Astronomy, Rensselaer Polytechnic Institute, in partial fulfillment of the requirements of the Ph.D. degree and was presented before the Dental-Medical Materials Group of the A.I.M.E. in Las Vegas, May, 1970.

Submitted to the Journal of Biomedical Materials Research.

NOV 21971

ABSTRACT

The pressure dependence of the elastic constants of dental amalgam has been examined in the 0-50 kilobar range using a solid media, pressure apparatus coupled with an ultrasonic interferometer. Computer analysis of the measured longitudinal and shear ultrasonic wave velocities yields the pressure dependence of the bulk, shear and Young's moduli and Poisson's ratio. Samples were prepared with varying compositions from micro-cut and spherical dental alloys. The elastic behavior of these samples can be directly related to the sample structure and composition as well as to the manipulation during preparation. In addition, an estimate can be made of the volume concentration of porosity.

INTRODUCTION

The examination of the response of a solid body to a deforming force can be separated into three parts: elasticity-spontaneously reversible deformation; anelasticity-time dependent reversible deformation; and plastic flow-irreversible deformation. Although several investigators have examined the viscoelastic behavior of dental amalgam,[1,2] there has been no definitive study of its purely elastic response to external forces. This three part series of papers will examine this elastic behavior, separated from the flow characteristics, of amalgam.

The early measurements of the elastic constants of amalgam, using tensile measurements produced values which are now seen to be much too low.[3] These measurements were made at sufficiently low strain rates that the amalgam samples were probably able to flow during the experiments. Since ultrasonic techniques measure the elastic response at extremely high strain rates compared to the rate of flow in amalgam, the purely elastic behavior can be studied.[4,5]

Dental amalgam can be considered a quasi-isotropic, quasi-homogeneous material since it is a random, polycrystalline mixture of several phases. Although an isotropic material requires only two independent elastic constants to describe its elastic behavior, the four quantities, bulk, shear and Young's moduli and Poisson's ratio, are generally used. These elastic constants can be determined by measuring the velocities of propagation of ultrasonic waves in samples whose lengths are long compared to the wavelength of the radiation. In elastic, isotropic media there are only two solutions to the equations of motion[6] - a pure longitudinal wave and a pure shear wave. These two velocities can be expressed in terms of the bulk and shear moduli as follows:

$$v_\ell = \sqrt{\frac{K + 4/3G}{\rho}}$$

$$v_\tau = \sqrt{\frac{G}{\rho}}$$

v_ℓ = velocity of the longitudinal wave
v_τ = velocity of the shear wave
K = bulk modulus
G = shear modulus
ρ = density

Expressing the elastic constants in terms of these velocities:

$$K = \rho(v_\ell^2 - 4/3\ v_\tau^2) \qquad E = \frac{9KG}{3K+G} = \rho v_\tau^2 \frac{3v_\ell^2 - 4v_\tau^2}{v_\ell^2 - v_\tau^2}$$

$$G = \rho v_\tau^2 \qquad \nu = \frac{3K-2G}{2(3K+G)} = \frac{v_\ell^2 - 2v_\tau^2}{2(v_\ell^2 - v_\tau^2)}$$

where E = Young's modulus and ν = Poisson's ratio.

SAMPLE PREPARATION

Dental amalgam samples were prepared from spherical and micro-cut commercial dental alloys with 45%, 50% and 52% Hg by weight. The samples were triturated for 15 seconds in a Wig-L-Bug and condensed in a 1 cm. diameter, cylindrical, steel die at pressures of 1000-3000 psi. This pressure range was chosen since mercury was not expressed from the samples, and hence, the mercury content was known.

EXPERIMENTAL TECHNIQUE

High pressures were applied to samples using an uniaxial, solid media press. The specimen was placed between two tungsten carbide pistons, see Figure 1, and was constrained laterally by two pyrophyllite gaskets and a tungsten carbide girdle. The gaskets and girdle prevent radial extrusion of the specimen and reduce the radial pressure gradient over the sample. Since the stresses in the specimen can be separated into hydrostatic and deviatoric components, the

apparatus was calibrated in terms of the hydrostatic pressures needed to induce known phase transitions in different materials.[7] From the calibration curve, the hydrostatic component of the applied pressure could be determined as a function of applied force, measured using a strain gauge load cell.

A 35 megahertz, quartz, x-cut transducer was bonded on the back of one piston and a 30 megahertz, quartz, y-cut transducer was bonded on the back of the other piston; Dow 276-V9 resin was used as a bonding agent. The transducers were maintained free from external stresses by drilling holes in the steel blocks used to apply pressure, see Figure 1. Electrical contact was made with the transducers through these holes. No bonding agent was used between the piston and specimen faces since the applied pressure forces the pistons and specimen together firmly enough to produce good acoustic contact.

It is difficult to resolve ultrasonic pulses from small specimens, and inaccurate time measurements result from transit time techniques. Therefore, an interference technique was used to determine the ultrasonic velocities. This technique was first developed by McSkimin[8] and was adapted to this pressure apparatus by Ahrens and Katz[9] and Gilmore,[7] Figure 2. The transducers were excited, one at a time, by a pulsed oscillator at frequencies in the region of 1/2 the resonant frequency so that the transducer output was approximately independent of small frequency changes. The resulting ultrasonic pulses were reflected internally from the piston-specimen faces, and when the pulse duration was made longer than twice the transit time within the specimen, interference occurred between reflections. The carrier frequency was varied until reflections from the two interfaces were π radians out of phase with each other, and destructive interference between the overlapped reflections was observed on the oscilloscope, Figure 3. At this frequency the internal reflections within the sample were all in phase but were π radians out of phase with the internal anvil

reflections, Figure 4.

This destructive interference, seen as a minimum in the overlapped pulse pattern, puts no restrictions on the values of the phase changes at the two boundaries except that the difference is π radians at a particular set of frequencies. It is evident from Figure 4 that destructive interference occurs whenever

$$2\ell = n\lambda$$

or when

$$2\ell = \frac{nv}{F}$$

ℓ = sample length
λ = wavelength
v = velocity
F = frequency

This can be rewritten as: $F = nF_o$ where $F_o = v/2\ell$ is a fundamental frequency corresponding to $2\ell = \lambda$. However, F_o is most easily determined by measuring the frequency difference between two successive minima:

$$F_o = F_n - F_{n-1}$$

In order to compute the ultrasonic velocities from these fundamental frequencies, it is necessary to know the length of the specimen at each pressure. Although the sample length can be measured during the experiment, it is possible to calculate the sample length once the changes in fundamental frequency with pressure and the initial sample length and density are known. The frequency differences between the fundamental frequency versus pressure curves for increasing and decreasing pressures provides information about the permanent deformation of the sample length during the pressure cycle. This calculation is more sensitive to small length changes than a direct measurement since the sample length is expressed in terms of an integral number of wavelengths of the radiation.

Since the elastic properties, as well as the density and dimensions, of an elastic solid change when pressure is applied, it is necessary to simultaneously consider the changes in the specimen's ultrasonic wave velocities,

length and density with pressure in order to calculate the changes in the elastic constants. Six coupled, first order, differential equations describing the geometry of the system, the elastic properties of the gasketing materials and the wave velocities, density, and length of the specimen as functions of pressure, and a computer program to evaluate them have been developed by K. L. Dunn and R. S. Gilmore of this laboratory.

RESULTS

Figures 5, 6 and 7 present the behavior of the bulk, shear and Young's moduli of four different dental amalgam samples as a function of pressure. The data for the spherical amalgams, prepared with 45% (●), 50% (+) and 52% (▲) Hg, are represented by solid data points and solid lines. A micro-cut amalgam, 50% Hg, is presented for comparison and is shown by open data points (○) and a dashed line. The observed linear behavior is consistent with that measured both in ductile and brittle materials.[7] It is evident from these measurements that Young's modulus for dental amalgam tends to decrease with increasing mercury content, Figure 7. This behavior holds at atmospheric pressure as well. The atmospheric pressure value of each elastic modulus is obtained from these pressure dependent measurements by a back extrapolation based on a least squares analysis (written by K. L. Dunn of this laboratory). These values are listed in Table 1.

Dickson and Oglesby[4] demonstrated this same behavior in the Young's modulus of dental amalgam measured directly at atmospheric pressure using a transit time, ultrasonic technique. This behavior can be related to the phase composition of amalgam, which is discussed in detail in the third paper of this series[10] and to porosity which is discussed below.

TABLE 1

The Bulk, Shear and Young's Moduli and Poisson's Ratio of Several Dental Amalgams of Ideal Density at Atmospheric Pressure

(All Moduli in 10^{12} dynes/cm^2)

Composition	Bulk Modulus	Shear Modulus	Young's Modulus	Poisson's Ratio
45% Hg Spherical Alloy	0.805	0.281	0.750	.35
50% Hg Spherical Alloy	0.752	0.258	0.695	.35
52% Hg Spherical Alloy	0.750	0.248	0.672	.35
50% Hg Micro-cut Alloy	0.750	0.253	0.680	.35

The experimental accuracy of these results is difficult to assess. Although the precision and reproducibility of the ultrasonic interferometer are very high, within ±1%, there are several factors which affect the accuracy of the measurements. The initial sample length and density inside the apparatus are only known to about ±5%. The pressure calibration was developed in terms of solid state phase transitions whose transition pressures are known only within ±1% to ±3%. In addition, errors may be introduced during the numerical evaluation of the differential equations used in the high pressure calculations; the behavior of the numerical integration technique at a discontinuous volume change is not fully known. Therefore, although the scatter of values of repeated measurements is small, the absolute errors may be of the order of ±5% or higher.

DISCUSSION

The curves of the elastic moduli versus pressure for the spherical amalgam samples show slight changes in slope in the neighborhood of 20 kilobars. These slope changes were initially observed in the ultrasonic velocity measurements and are naturally reflected in the elastic constants. Similar slope changes have been observed in two of the constituent phases of dental amalgam (γ-Ag_3Sn and γ_2-$HgSn_{7-8}$).[11] Changes of slope of this type may arise due to any one or combination of several factors; the discussion of these effects has been deferred to the second paper in this series.[11]

Another type of slope change can be observed in the elastic moduli of the micro-cut alloy at lower pressures, Figure 8. This deviation from linearity at low pressures is typically seen in porous materials.[7] Pores, or voids, are present in dental amalgam samples prepared by trituration and

condensation. At pressures above the yield strength of amalgam, the specimen flows by plastic deformation; porosity is removed, and the ultrasonic velocity measurements are made on the material at ideal density. However, as the pressure is decreased, the sample expands, and porosity may be reintroduced in the forms of microfissures and cracks due to fracturing on stress release.

Studies of other materials have shown that the Young's modulus of a material decreases significantly with the introduction of small volume fractions of porosity. These studies fall into two general classes: those in which the pores are closed (often a spherical geometry is assumed for simplification); and those in which the pores are interconnected in a rather complex geometry. Mackenzie[12] has derived expressions for the elastic constants of a solid in which closed spherical pores are contained. Coble and Kingery[13] in their experiments on sintered alumina, have shown that the effective Young's modulus decreases with increasing porosity as predicted by Mackenzie, and can be estimated by $E = Eo\,[1 - (1 + A)\,V + AV^2]$ where V = the relative volume fraction of porosity, Eo is the modulus at ideal density, and A = 0.9 for a material where $\nu = 0.30$.

For the case of interconnected porosity, the decrease in Young's modulus with increasing porosity is much steeper. McAdam[14] has shown that the elastic behavior of sintered ferrous compacts as a function of porosity could be fitted by a smooth curve which has the equation $E = Eo\,(1 - V)^n$ where n = 3.4 and Eo and V are as before. In a more recent study on fibrous metals, Bol'shin and Fedotov[15] have shown that the same phenomenological expression holds for copper as well as for steel; they obtained n(Cu) = 2.6 and n(steel) = 3.6, in good agreement with McAdam.[14]

This effect is seen at lower pressure in the micro-cut alloy specimen, Figure 8; the measured ultrasonic velocities and densities, and the elastic

constants, fall below a linear extrapolation of the expected values for ideally dense samples. Spherical alloy amalgams did not exhibit evidence of porosity at low pressures after pressure cycling.

A theoretical estimate of the porosity present in the 50% Hg, micro-cut alloy, amalgam can be made by comparing the Young's modulus measured near atmospheric pressure with the values obtained by extrapolation from the high pressure values. In the case where closed pores in a continuous material are assumed, the effective Young's modulus can be estimated by using Coble and Kingery's form of the equation above since A = 0.9 also for a material where $\nu = 0.35$. Applying this equation to the 50% Hg, micro-cut alloy, one obtains a calculated porosity of 2.3%; assuming interconnected pores, and a value of $n = 2.6$ as for Cu, results in a calculated value of 1.8% porosity using McAdam's equation. These values compare well with the relative volumes of porosity measured in dental amalgam by Jorgensen and Kanai.[16]

Comparison of the Young's moduli measured in this experiment to those obtained by Dickson and Oglesby[4] shows that the values obtained for Young's modulus of amalgam in this experiment are about 10%-15% higher. Although some of this difference can be attributed to differences in sample composition and manipulation, a major factor is probably the presence of porosity in their samples since the effects of porosity were removed at high pressures in this study.

SUMMARY

The elastic constants of dental amalgam have been measured as functions of pressure in the range 0-50 kilobars. High pressure studies were needed so that the elastic behavior measured represented that of an ideally dense material.

Since the strain rate applied to specimens using ultrasonic techniques is very high, the responses measured in this experiment were indications of the purely elastic behavior of dental amalgam.

The atmospheric pressure values of the bulk, shear and Young's moduli and of Poisson's ratio of the amalgam samples were obtained by back extrapolation from the high pressure measurements. It was observed that these values decreased with increasing mercury content, and large decreases in the values of elastic constants were measured in samples containing porosity.

The elastic constants of amalgam exhibit linear increases with increasing pressure, however, some ultrasonic data indicated the possibility of changes of the slopes of the elastic constants versus pressure.

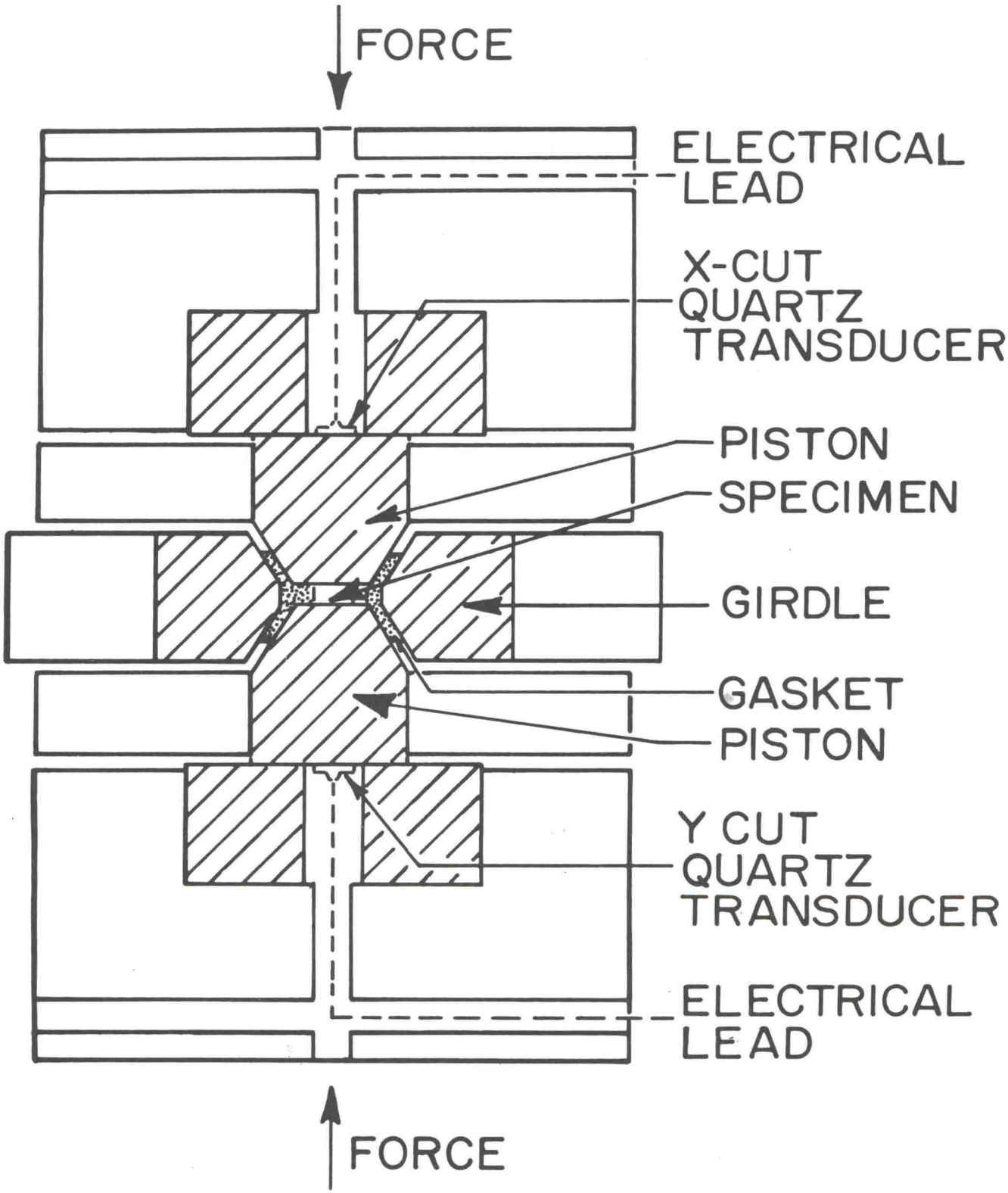
ACKNOWLEDGEMENTS

The authors would like to thank Mr. K. L. Dunn and Dr. R. S. Gilmore for their invaluable help in this project. Dr. Gilmore designed much of the experimental apparatus and technique; Mr. Dunn and Dr. Gilmore developed the method of analysis and Mr. Dunn wrote the computer programs used for the analysis.

FIGURE 1

Diagram of the High Pressure Anvils

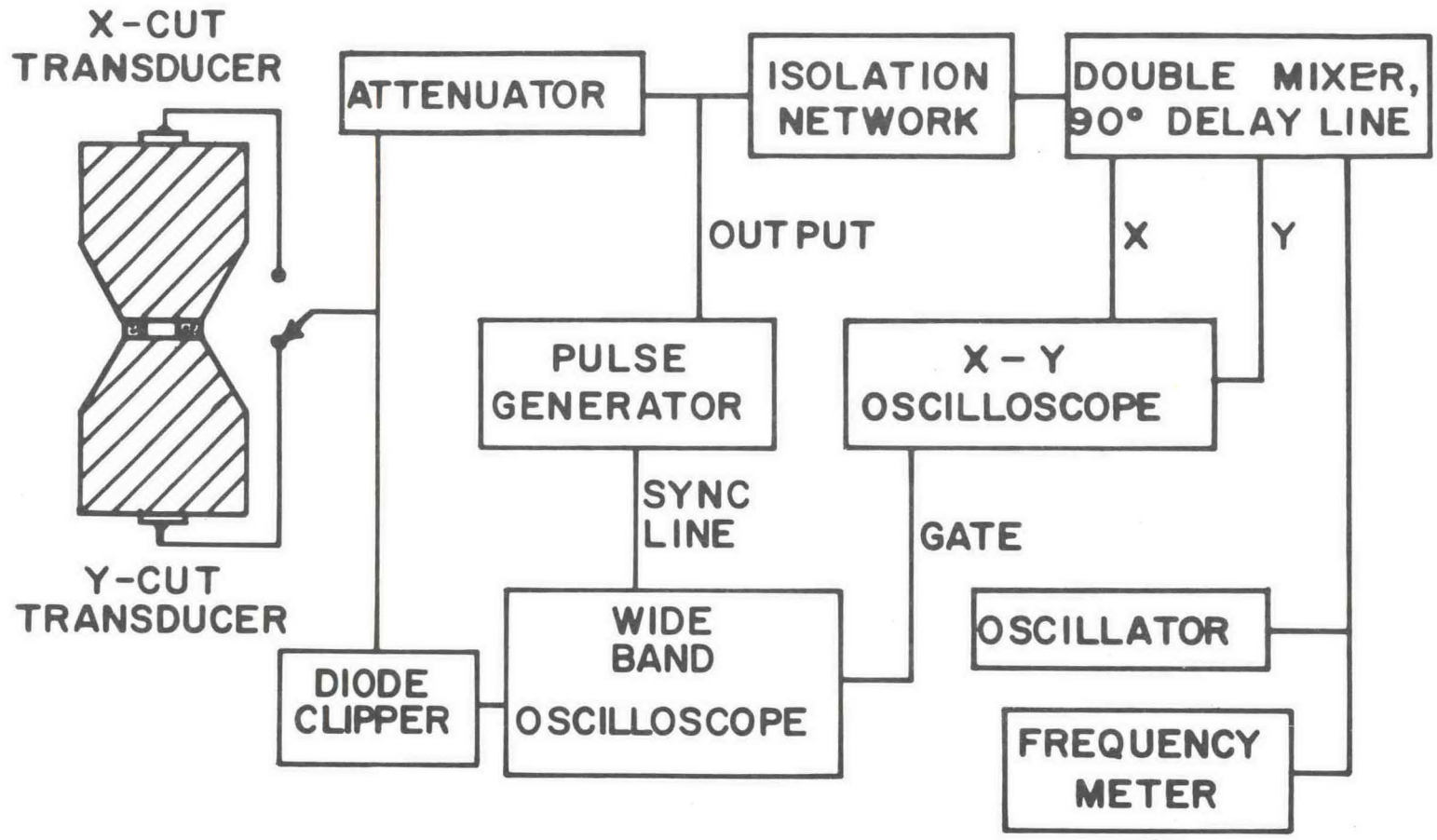
Retaining Ring, Sample and Gaskets



AFTER GILMORE (1968)

FIGURE 2

Block Diagram of the High Pressure Apparatus

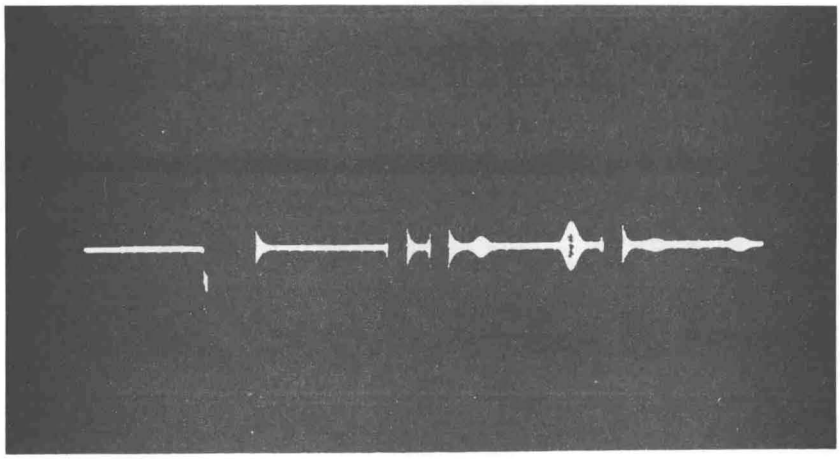


(AFTER GILMORE, 1968)

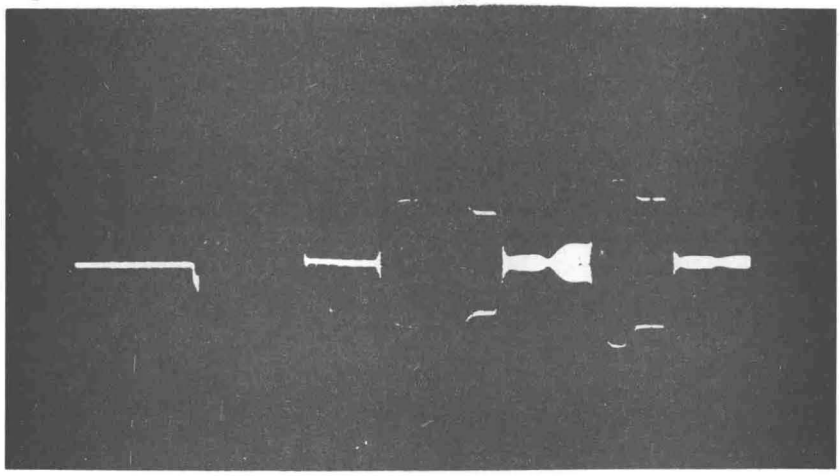
FIGURE 3

Photographs of Reflections and Interference at High Pressure

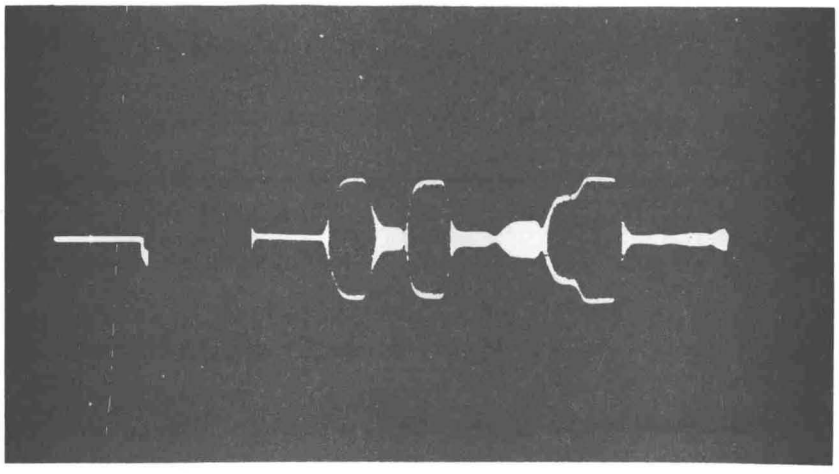
(NaCl sample used for illustrative purposes)



1 - main pulse
2 - reflection from first specimen - anvil boundary
3 - reflection from second specimen - anvil boundary



reflections overlapped - maximum

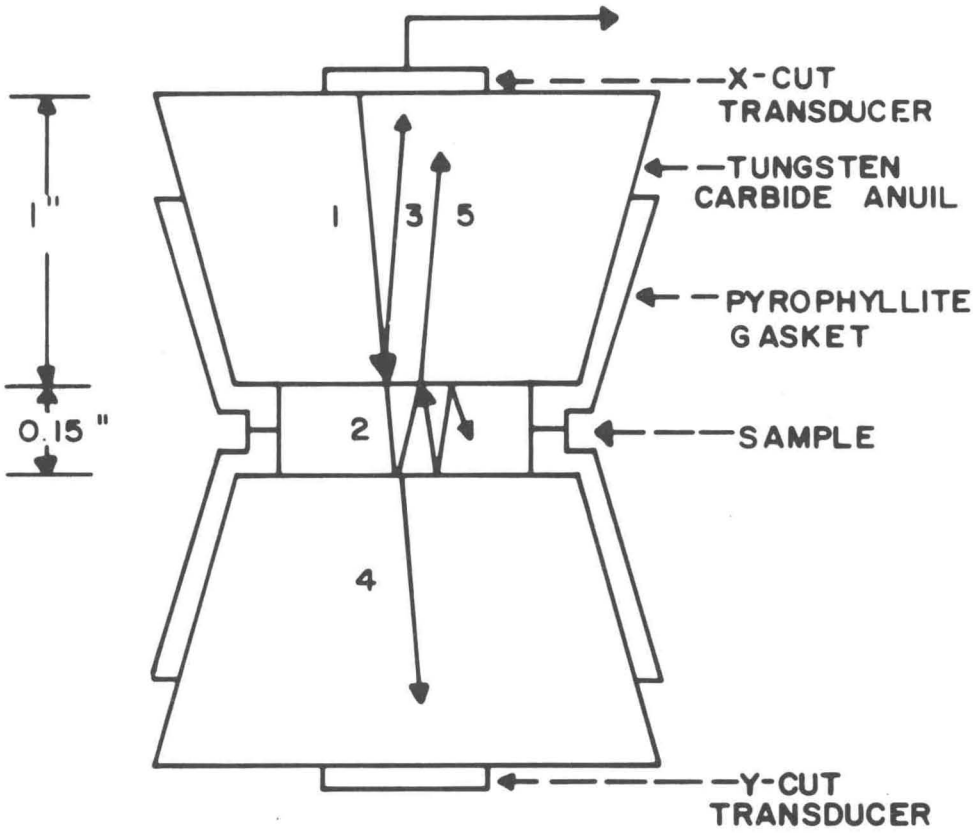


reflections overlapped - minimum

FIGURE 4

Schematic of Ultrasonic Reflections

Within the Anvils and Specimen



1 - MAIN PULSE (DRAWN AT ANGLE FOR CLARITY)

2 - TRANSMITTED WAVE in SPECIMEN

3 - REFLECTED WAVE FROM UPPER SPECIMEN - ANVIL BOUNDARY

4 - WAVE TRANSMITTED INTO LOWER ANVIL

5 - REFLECTED WAVE FROM LOWER - SPECIMEN - ANVIL BOUNDARY

FIGURE 5

The Bulk Moduli, as a Function of Pressure,

for One Cut and Three Different Spherical Dental Amalgams

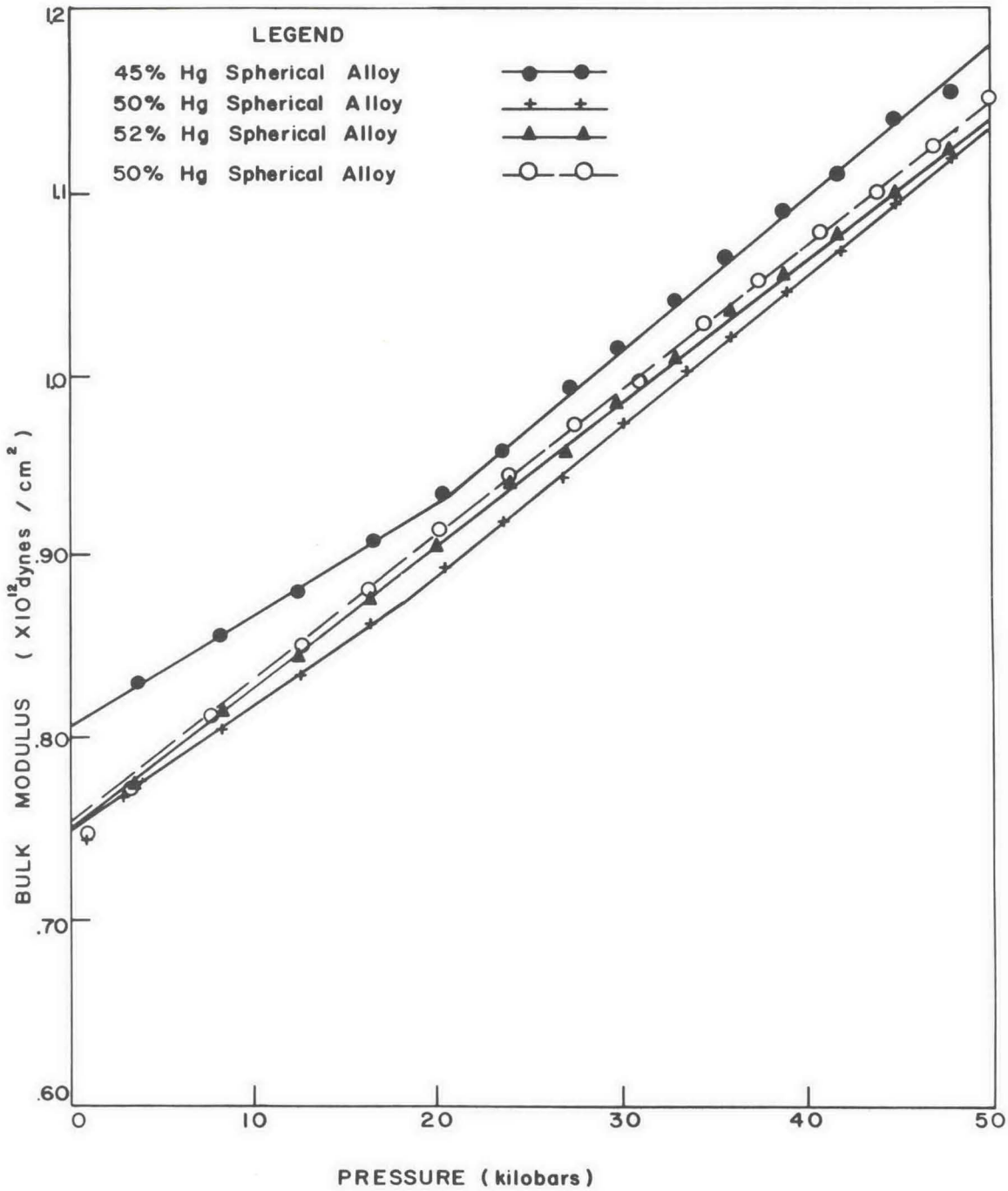

LEGEND
45% Hg Spherical Alloy
50% Hg Spherical Alloy
52% Hg Spherical Alloy
50% Hg Spherical Alloy
1.2
1.1
1.0
.90
.80
.70
.60
BULK MODULUS (X10^12 dynes / cm^2)
0
10
20
30
40
50
PRESSURE (kilobars)

FIGURE 6

The Shear Moduli, as a Function of Pressure,

for One Cut and Three Different Spherical Dental Amalgams

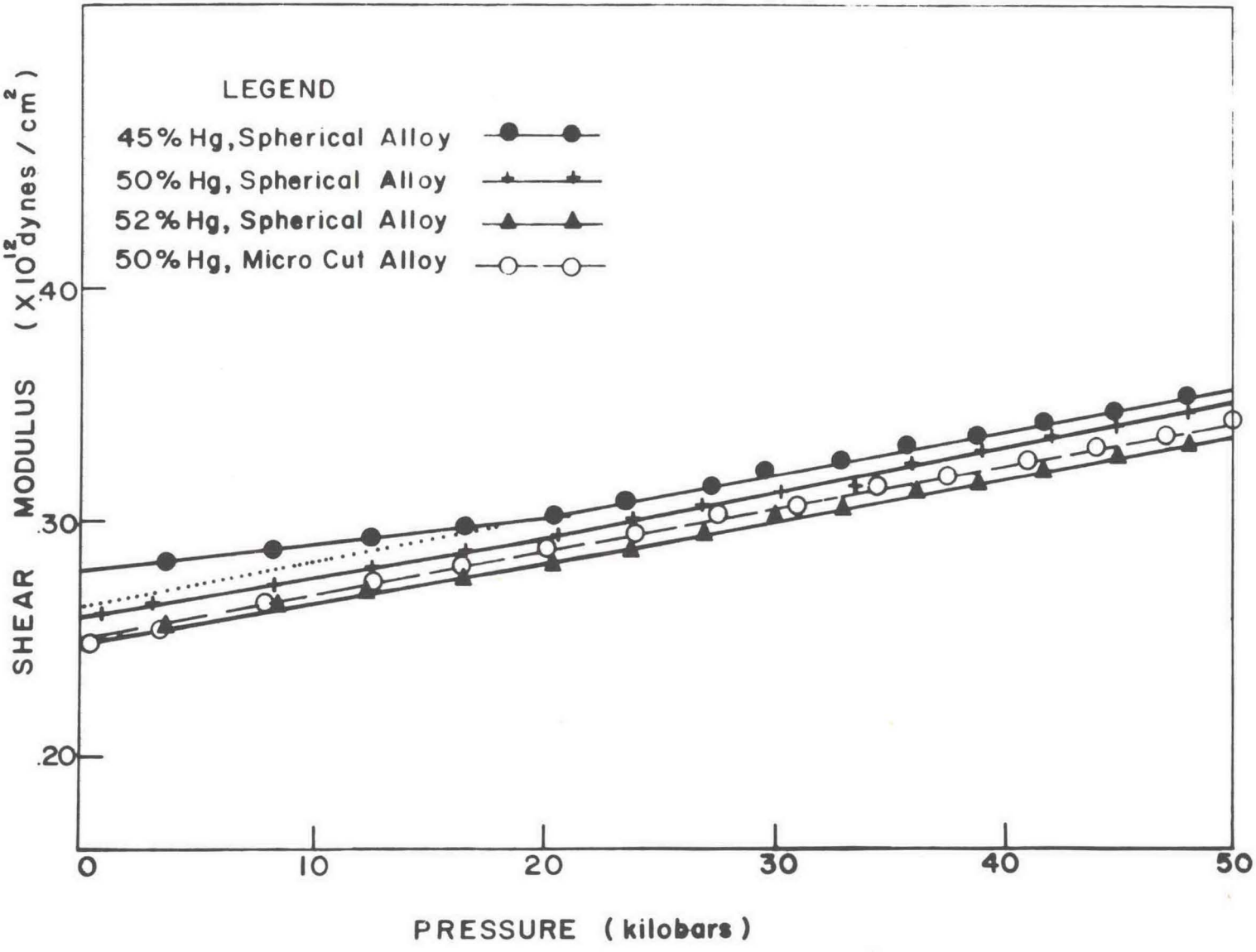

LEGEND
45% Hg, Spherical Alloy
50% Hg, Spherical Alloy
52% Hg, Spherical Alloy
50% Hg, Micro Cut Alloy
SHEAR MODULUS (X 10^12 dynes / cm^2)
.40
.30
.20
0
10
20
30
40
50
PRESSURE (kilobars)

FIGURE 7

The Young's Moduli, as a Function of Pressure,

for One Cut and Three Different Spherical Dental Amalgams

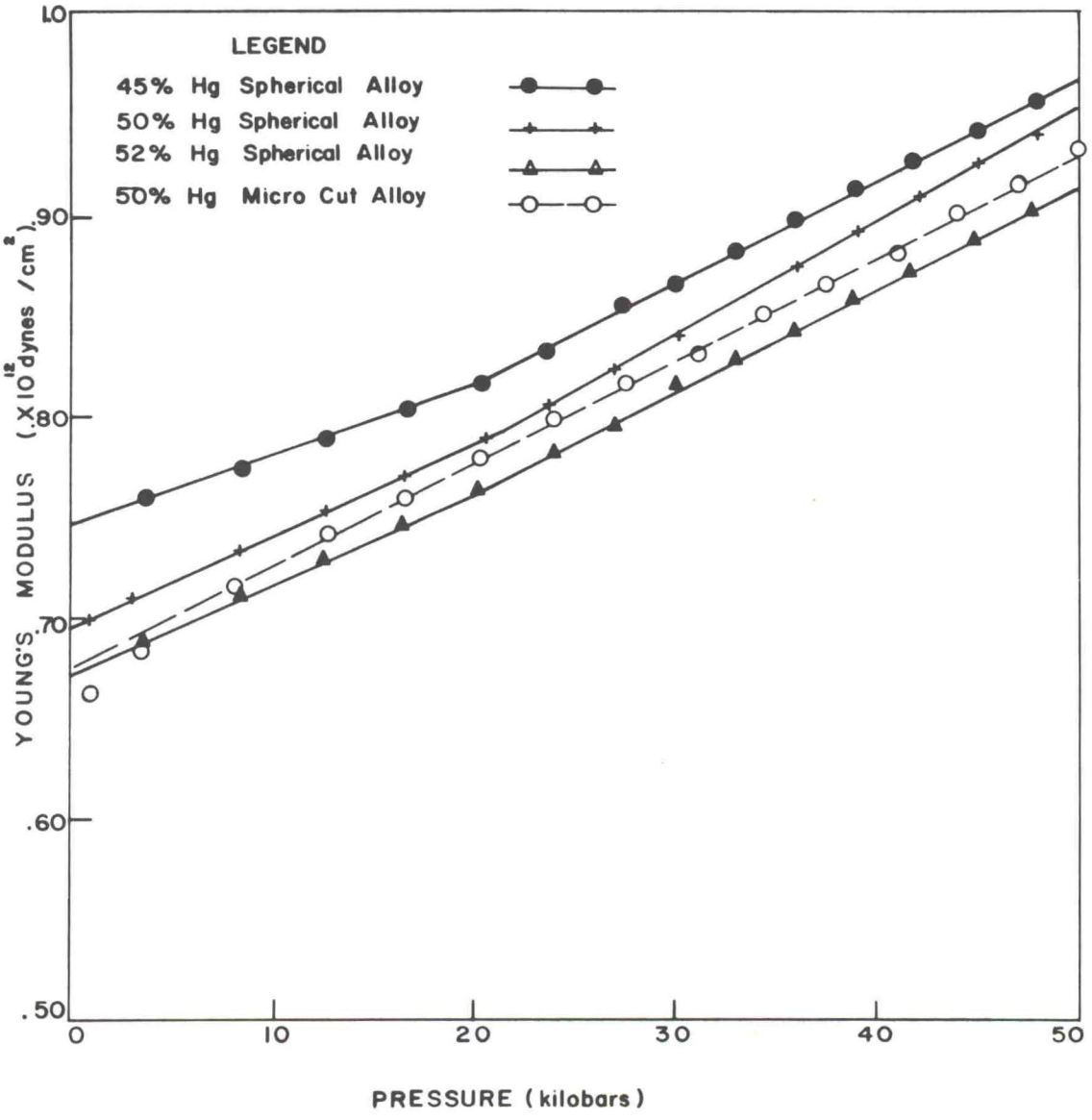
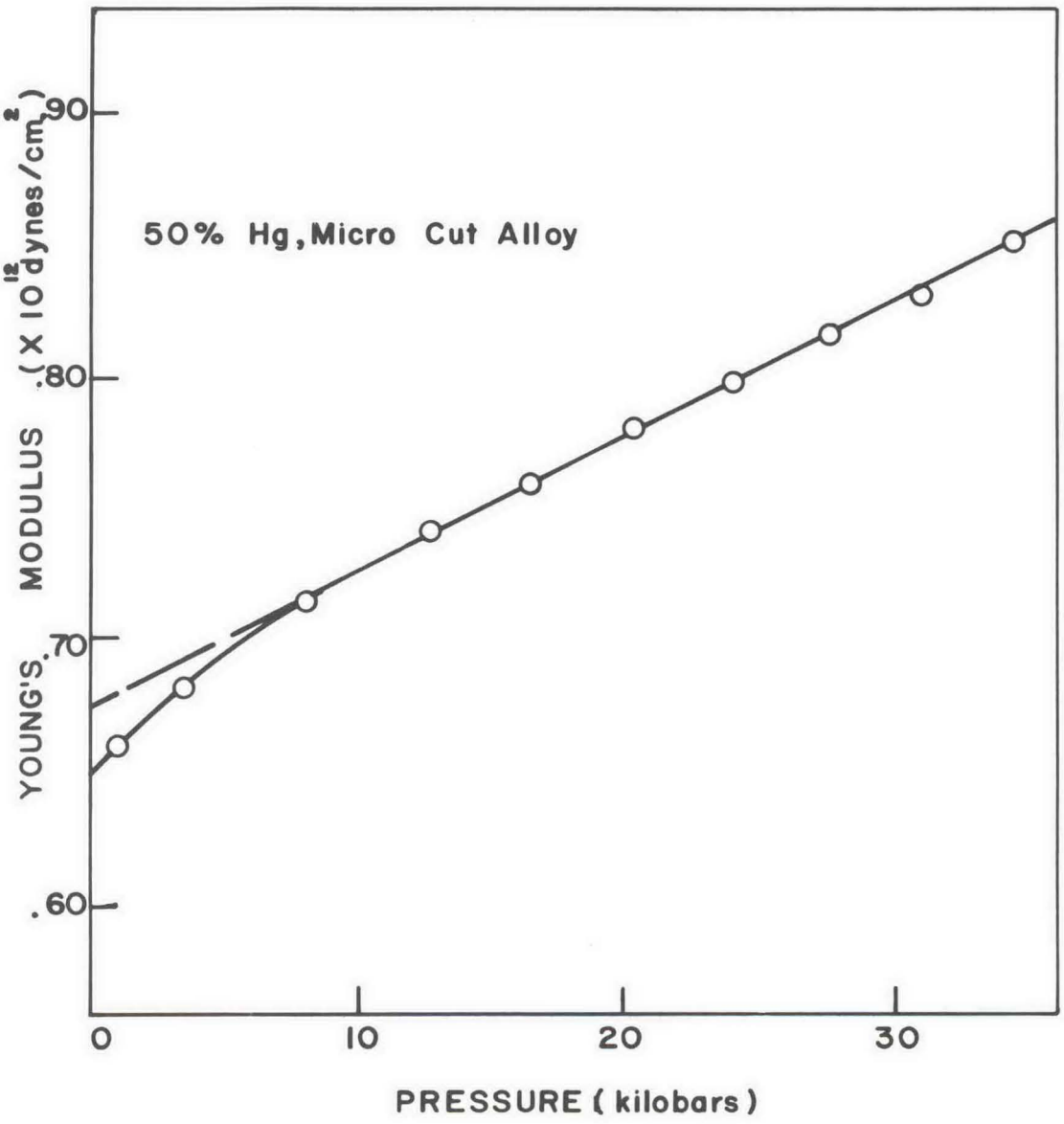

LEGEND
45% Hg Spherical Alloy
50% Hg Spherical Alloy
52% Hg Spherical Alloy
50% Hg Micro Cut Alloy
1.0
.90
.80
.70
.60
.50
YOUNG'S MODULUS (X10^12 dynes /cm^2)
0
10
20
30
40
50
PRESSURE (kilobars)

FIGURE 8

Young's Modulus vs Pressure for a 50% Hg, Micro-Cut Alloy

Showing the Back Extrapolation to Obtain an Atmospheric Pressure Value


50% Hg, Micro Cut Alloy
YOUNG'S MODULUS (X 10^12 dynes/cm^2)
.90
.80
.70
.60
0
10
20
30
PRESSURE (kilobars)

REFERENCES

1. P.L. Oglesby, G. Dickson, M.L. Rodriguez, R.M. Davenport and W.T. Sweeney, J. Res. Nat'l Bur. Standards, 72C, 203 (1968).

2. G. Dickson, P.L. Oglesby and R.M. Davenport, J. Res. Nat'l Bur. Standards, 72C, 215 (1968).

3. M.L. Rodriguez and G. Dickson, J. Dent. Res., 41, 840 (1961).

4. G. Dickson and P.L. Oglesby, J. Dent. Res., 46, 1475 (1967).

5. D.E. Grenoble, R.S. Gilmore and J.L. Katz, "Elastic Constants of Alloys and Amalgams," Abst., A.I.M.E., Los Angeles, Feb. (1967).

6. A.E.H. Love, A Treatise on the Mathematical Theory of Elasticity, Dover Publications, New York (1944).

7. R.S. Gilmore, "The Elastic Constants of Fifteen Materials as Functions of Pressure and Their Equations of State," Ph.D. Thesis, Rensselaer Polytechnic Institute (1968).

8. M.J. McSkimin, J. Acoust. Soc. Am., 22, 413 (1950).

9. T. Ahrens and S. Katz, J. Geophys. Res., 67, 2935 (1962).

10. J.L. Katz and D.E. Grenoble, "A Composite Model of the Elastic Behavior of Dental Amalgam," This Journal (197).

11. D.E. Grenoble and J.L. Katz, "The Elastic Constants of the Constituent Phases of Dental Amalgam," This Journal (197).

12. J.K. Mackenzie, Proc. Phys. Soc. (London), 63B, 2 (1950).

13. R.S. Coble and W.D. Kingery, J. Am. Ceram. Soc., 39, 377 (1956).

14. G.D. McAdam, J. Iron and Steel Inst., 168, 346 (1951).

15. M. Yu Bol'shin and S.G. Fedotov, Soviet Powder Metallurgy and Metal Ceram. 4(64), 77 (1968).

16. K.D. Jorgensen and S. Kanai, Acta Odont. Scand., 23, 501 (1965).