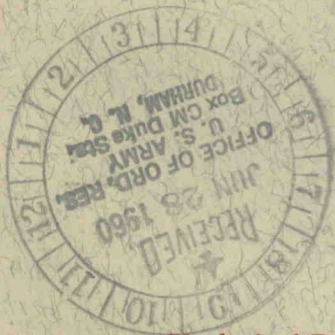


*H. J. Hall*

*High Pres. Apparatus*

# Some Developments in High-Pressure Technique



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by

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Cambridge, Massachusetts

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## SOME DEVELOPMENTS IN HIGH-PRESSURE TECHNIQUE

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Abstract: Various research problems requiring high-pressure techniques have been investigated in the Laboratory for Insulation Research during the past five years. The report here presented summarizes some of the technical experience gained in the course of this work.

### Introduction

Industrially, high-pressure techniques are important for chemical synthesis in gases, liquids, and solids; a notable example is the synthesis of diamond. Scientifically, interest in high pressures includes problems such as seismic wave transmission through the earth and the states of matter in dense stars. In the latter case the densities are many orders of magnitude greater than those of minerals in the earth and the intensities of pressure are such that they could not conceivably be reproduced in the laboratory. Bridgman<sup>1)</sup> has developed techniques for producing pressures as high as 100,000 and for certain measurements 400,000 atm; at these pressures the density of some substances doubles.

In solid-state theory, the significance of knowing the properties of crystals at high hydrostatic pressures lies in the dependence of the electronic wave functions on internuclear distances: Cooling a crystal will decrease the

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\* Present address: National Carbon Co., Research Laboratories, Cleveland 1, Ohio.

1) P. W. Bridgman, "The Physics of High Pressure," Bell and Sons, London, 1949.



internuclear distance without requiring complicated pressure contraptions, but at the same time it changes the statistical occupations of energy states. In barium titanate at  $130^{\circ}\text{C}$ , for instance,<sup>2)</sup> decreasing the internuclear distances by applying pressure at constant temperature lowers the dielectric constant, whereas decreasing the internuclear distances identically by cooling at constant pressure raises the dielectric constant. For a one-inch long rod of iron, a change of length of 0.001 inch can be produced either by a temperature change of  $70^{\circ}\text{C}$  or by a pressure change of 5000 atm.

### Solids and Gases as Pressure Media

Bridgman<sup>3)</sup> measured the variation in conductivities of metals at pressures as high as 100,000 atm by a very simple device. The two truncated cone sections of Fig. 1 represent the jaws or anvils of a hydraulic vise. The metal-strip resistance to be measured is embedded in a plastic disk squeezed in this vise. The material of the disk is a soft substance (e.g., silver nitrate); it is kept from oozing out of the vise by means of a pipestone washer surrounding it. Pipestone is like a finely grained sandstone and crushes under the squeeze, but it packs together tightly and does not ooze out. A thin metal disk pressed in a two-jaw vise has a sufficiently sharp superconductivity transition at 30,000 atm to allow a recording of the shift in the transition temperature.<sup>4)</sup>

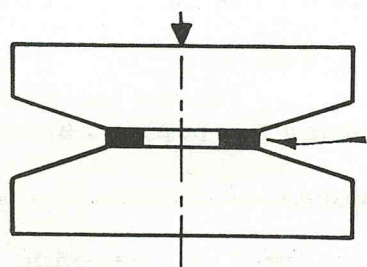


Fig. 1.

Pipestone ring Double anvil for 100,000 atm.  
(After Bridgman.<sup>3)</sup>)

2) W. J. Merz, Phys. Rev. 78, 52 (1950).

3) P. W. Bridgman, Proc. Am. Acad. Arts Sci. 81, 165 (1952).

4) P. F. Chester and G. O. Jones, Phil. Mag. [7] 44, 1281 (1953).



Hall, one of the group that developed the diamond-synthesis techniques, has extended the technique of Fig. 1 to a vise consisting not of two but of four jaws pressing on a tetrahedrally-shaped sample.<sup>5)</sup>

In experiments such as these, where pressure is created in a relatively plastic solid, very high pressures can be reached. Under such conditions, however, it would be difficult to measure the conductivity properties of a semiconductor because the potential electrodes would be torn adrift by the plastically flowing solid pressure medium. Almost any single crystal to be studied electrically or magnetically would be broken, unless it were unusually strong or of compact geometry (e.g., spherical). For the majority of experimental measurements on crystals it is therefore necessary to use a fluid compressive medium that does not become too viscous at high pressures.

Bridgman<sup>1)</sup> has shown how to compress pentane to a pressure of 30,000 atm, but pentane would decompose at high temperatures. Further, as shown by the approximate freezing curve in Fig. 2, pentane would not cover much area in the low-temperature parts of the P-T diagram. For these reasons, the two gases kept in mind in the design of techniques described here have been argon and helium. Argon, extrapolating Bridgman's data,<sup>1)</sup> probably freezes near 15,000 atm. Helium<sup>6)</sup> is the most difficult gas to seal tightly in a pressure vessel. Bridgman found that metallic surfaces had to be polished very smooth

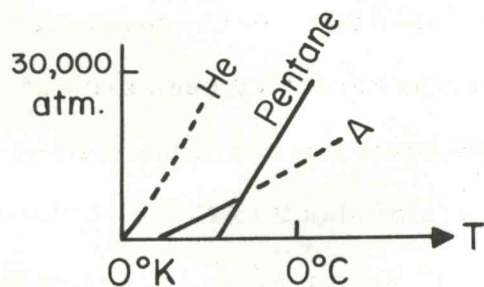


Fig. 2.

Approximate melting curves for pentane,<sup>1)</sup> argon,<sup>1)</sup> and helium.<sup>6)</sup> (dashed lines do not represent data but suggest areas to the right of which the fluid is not frozen.)

5) H. Tracy Hall, *Sci. American* 201, No. 5, 61 (1959).

6) E. R. Grilly and L. R. Mills, *Annals of Physics* 8, 1 (1959).



in sealing areas and that helium diffuses through tool-steel vessels at pressures above 8000 atm (which is the initial elastic limit of a very thick-walled cylinder of hardness about Rockwell 45 - 48 C). No one has seriously considered using helium at pressures higher than this, but the approach must be along two lines: better steels or different metallurgical grain characteristics of stainless steels or beryllium copper, and temperature reduction of the cylinder in which the driven piston generates the pressure, to reduce the diffusion rate of the helium.

### Limits Imposed by Metals

#### Hardness and ductility

A typical vessel for high pressures might have a bore of 1/2 inch and an outside diameter of 4 inches. A very thick-walled vessel such as this can withstand an internal pressure far greater than the tensile strength of the metal. However, this is so only if the metal is sufficiently ductile to stretch internally. In the process of stretching, the cylinder gains in hardness by plastic deformation (cold work) and assumes a more favorable pattern of radial and tangential stress. It is necessary to "draw," by heating the vessel, from the quenched hardness of Rockwell 60 C to 40 - 50 C. The elastic limit of the vessel may then be increased by subjecting it to an application of internal pressure of 20,000 to 30,000 atm. In this way, Bridgman used tool-steel vessels for measurements up to 20,000 atm. Beryllium copper has recently been used at higher pressures;<sup>7)</sup> it is precipitation-hardenable to a maximum hardness of about Rockwell 40 C and more ductile than the vessels of about 45 C used by Bridgman; since it is softer, however, the bore will become scored more readily by the hard compressing piston. Work-hardened stainless steel tubing of grade

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7) D. M. Warschauer and William Paul, *Rev. Sci. Instr.* 29, 675 (1958); William Paul, G. B. Benedek, and D. M. Warschauer, *ibid.* 30, 874 (1959).



316 has been used at 20,000 atmospheres;<sup>7)</sup> although it is a non-hardenable alloy, it work-hardens to such a great extent that the autofrettage effect is very pronounced.

If pressure is applied to the outside of a pressure cylinder, the bore will stretch less for a given internal pressure. Bridgman forced an outer conical jacket (Fig. 3) over the lubricated conical surface of the cylinder. As internal pressure was increased or reduced, so was the forcing thrust; this allowed the jacket to be pushed on tighter or to slide off a little, keeping the effective radial pressure exerted by the jacket at about one third of the internal pressure. Even after repeated use with an internal pressure of 30,000 atm, the bore showed relatively little evidence of stretching. At this pressure, however, the binding of the packing rings against the bore began to cause trouble, a difficulty partly alleviated by copperplating the rings.

In industrial applications, cylinders are often strengthened by shrinking on one or more outer cylinders permanently about the inner cylinder,<sup>8)</sup> or by winding wire on under tension.

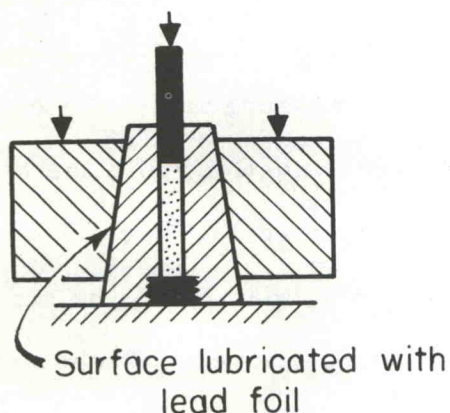


Fig. 3.

Schematic illustration of external support used by Bridgman<sup>1)</sup> for compressing pentane to 30,000 atm.

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8) D. M. Newitt, "The Design of High Pressure Plant and the Properties of Fluids at High Pressure," Clarendon Press, Oxford, 1940.



### The effect of temperature

If a pressure cylinder is to be heated or cooled in order to vary the temperature of the sample inside it, limits are set by the onset of creep in the metal at high temperatures and, for some metals, by embrittlement at low temperatures. Creep begins to occur above the recrystallization temperature of the principal element in the alloy (roughly  $350^{\circ}\text{C}$  for iron and  $200^{\circ}\text{C}$  for copper). Even at stresses very much lower than the elastic limit of the metal in short-time tests the metal will slowly stretch, or "creep," and eventually fracture. In precipitation-hardenable alloys, high temperature also results in a loss of short-time tensile strength because of overaging and agglomeration of the fine intermetallic precipitate that resists plastic flow. A steel cylinder at  $400^{\circ}\text{C}$  and a beryllium copper cylinder at  $300^{\circ}\text{C}$  could therefore be used at full pressure for only a relatively short time.

Low-temperature embrittlement begins at  $-100^{\circ}$  or  $-150^{\circ}\text{C}$  in many steels. Certain stainless steels are not subject to it and in beryllium copper both strength and ductility improve at low temperatures.

### The effect of gases

Hydrogen, in almost any form, under pressure in a hot steel cylinder will penetrate and attack the steel by chemical reaction with the carbon.<sup>9)</sup> In a vessel filled with mercury, sufficient pressure will break through the surface oxides and amalgamation will take place, damaging the strength of the vessel for pressures above a few thousand atmospheres. These are essentially chemical attacks, but there is also a form of embrittlement due to hydrogen under pressure in a cylinder at room temperature, occurring for pressures in excess of 2000 atm. Although this process is not considered "chemical," helium, the atomic size of which is about equal to that of the

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9) E. W. Comings, "High Pressure Technology," McGraw-Hill Book Co., New York, 1956.



hydrogen molecule, does not cause trouble up to about 8000 atm. Austenitic stainless steels, in contrast to martensitic stainless steels and tool steels, are not subject to this type of hydrogen embrittlement. In all cases, the rate of penetration of the gas, whether by diffusion through or between the grains, is bound to be very much less at low temperatures.

### Threads

It is a common experience that a threaded plug holding a very high pressure may be exceedingly difficult to unscrew even after the pressure has been released. The binding of threads in high-pressure cylinders is greatly reduced by lubrication with molybdenum disulphide powder (made into an oil paste for convenience), as first used by Lawson. Also, as most machinists know, the binding of threads is minimized if the two parts have different hardnesses or are of dissimilar materials.

The principal rules for a safe thread are: that the metal in the thread be sufficiently ductile to allow the thread to accommodate itself over the whole surface; that the threads strip by shear, for which one may safely take half of the stated tensile strength of the metal; that, if both parts of the thread are of similar strength, the thread will strip halfway between the crest and the root of the thread; and that Ordnance regulations, requiring a 5:1 safety factor, be observed. Thus, if the diameter and cylindrical length of a thread are  $D$  and  $L$ , and the tensile strength of the material is  $T$ , the maximum force applicable on the plug, within safety regulations, is  $(\frac{1}{2} T \times \frac{1}{2} \pi DL)/5$ . This means that a one-inch diameter screw holding a half-inch plug against 20,000 atm must be at least two inches long in order to have a 5:1 safety factor.

### Typical metals useful in high-pressure design

A bar of steel as it comes from the mill represents a particular composition of iron, carbon (0.1 to 1.5 %), and a variety of alloying and processing



elements that have been poured into an ingot, rolled into a rod and possibly annealed. If the bar is heated to  $1000^{\circ}$  or  $1200^{\circ}\text{C}$ , its structure will change to austenite, a face-centered cubic iron lattice containing carbon or carbide in solid solution. By quenching, the austenite is changed to martensite, a complicated structural aggregate which is very hard. In a heavy bar, however, the inside cannot lose heat fast enough and the austenite reverts back to the soft original state instead of becoming martensitic as on the outer surface. Alloying elements reduce the speed of transition from the austenitic to the soft state and thereby allow a heavy bar to harden more deeply. Alloying elements improve toughness (e.g., nickel) or wear resistance (e.g., chromium). They also vary the phase diagram: a fairly large amount of nickel stabilizes the austenite phase at room temperature. Austenite is very tough and finds its principal application in austenitic stainless steels (iron-chromium-nickel alloys), which contain little carbon but have a high degree of work-hardenability and can be hardened to about Rockwell 40 C if they contain age-hardening additions.

Pistons for producing very high pressures (cf., e.g., Fig. 26, p. 29) are made of a water-hardening or oil-hardening tool steel that is tough rather than especially wear-resistant. If they need to be stainless, they can be of a martensitic stainless steel such as 440 C. Such pistons are used at full hardness (Rockwell 60 C). In the apparatus of Fig. 3, Bridgman used tungsten carbide pistons. Pistons for precompressors (e.g., Fig. 21, p. 24) need be hard only on the surface. Nitrided steel has an especially high surface hardness (Rockwell 80 C).

Pressure cylinders in which the piston packing slides through the bore (e.g., Fig. 27) are made of oil-hardening tool steel that is tough, with a Rockwell hardness 40 - 50 C, much less than quenched hardness. They may

also be made of beryllium copper<sup>7)</sup> wrought alloy No. 25, age-hardened to Rockwell 40 C. Where the piston packing does not slide along the bore (e.g., Fig. 30, p. 35) pressure cylinders can be made of softer but very tough materials, such as 316 austenitic stainless steel (which is highly resistant to corrosion), beryllium copper alloy No. 25, or age-hardenable stainless steels. The same applies to sample vessels such as that at the lower end of Fig. 29, p. 33; this could be made of beryllium copper for magnetic measurements or of zirconium for neutron-diffraction studies of a sample under pressure.

Pressure tubing (e.g., the 4:1 diameter-ratio chrome-nickel-molybdenum tubing supplied by pressure-equipment manufacturers) can be used up to 7000 atm. Austenitic 316 stainless steel tubing, available in many sizes, will hold 20,000 atm.<sup>7)</sup> Parts such as tie-tubes (cf. Figs. 27, p. 30, and 30, p. 35) and cylinders for precompressors (cf. Fig. 21, p. 24) can be made of tool steels available in the form of hollow bars or they can be bored from solid rods of a deep-hardening steel available in moderately hard and yet machinable condition. Tie-tubes hold the pressure systems of Figs. 27 and 30 (pp. 30 and 35) together and also act as burst shields; they should therefore not be harder than Rockwell 35 or 40 C and may even be made of unhardened stock such as commercial mechanical tubing. If they are made of a tool-steel hollow bar, air-hardening grades should be used, since these have the least deformation on heat treating.

#### Approach to a high-temperature gas system

If measurements are to be made at temperatures above 350° - 400°C, the steel cylinder cannot be used at full pressure. Superalloys would have to be used and the vessel would have a relatively short life, determined by the creep



characteristics of the metal. The alternative is to put a heating coil inside the bore. The inner wall of the cylinder must be thermally insulated from the heating coil to prevent the temperature of the metal from exceeding  $300^{\circ}$  -  $350^{\circ}\text{C}$ . A nonporous insulating jacket must be used, since pores become filled with the heat-convecting compressed gas, and it must be possible to remove heat from the inner wall of the cylinder at a rate sufficiently high to compensate for the relatively poor thermal insulation. The gas must be inert and quite free from impurities; Yoder<sup>10)</sup> used specially purified argon. At room temperature argon probably freezes at about 15,000 atm, but if the vessel walls are allowed to heat up to  $150^{\circ}$  or  $200^{\circ}\text{C}$  (cf. Fig. 2), the argon should still be fluid at a very much higher pressure. It will of course freeze at the ends of the cylinder, but frozen argon is relatively plastic<sup>1)</sup> and will not impede the motion of the compressing piston. If the electrical inlet posts in the other end of the cylinder are properly constructed, the frozen argon should not affect them.

There are many points of view concerning the type of vessel suitable for this purpose. Figure 4 illustrates one approach, based on Bridgman's method of Fig. 3. The central part of the cylinder can be easily cooled by a

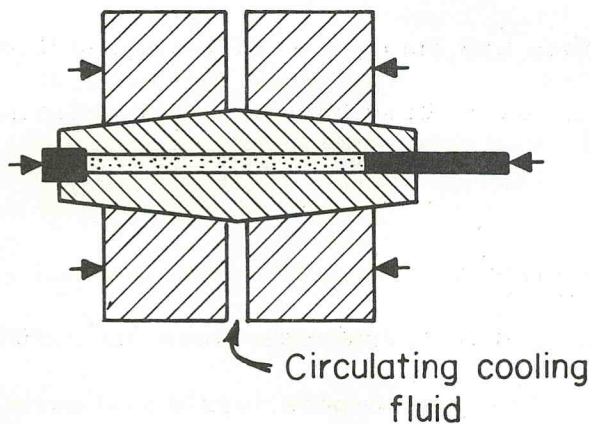


Fig. 4.

Possible adaptation of Bridgman method (Fig. 3) for use with an internal heater.

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10) Hatten S. Yoder, Jr., Trans. Am. Geophysical Union 31, 827 (1950).

liquid circulating between the two jackets that apply the externally supporting pressure. In addition to the radial compression exerted by the supporting jackets, there is also a very large longitudinal compression in the central part of the cylinder. The latter part is therefore under a more nearly hydrostatic externally supporting compression, which greatly reduces the shear stresses that are principally responsible for flow and fracture.

An approach to a low-temperature gas system

For low-temperature studies, it is necessary to use helium (cf. Fig. 2), as first reported by Bridgman<sup>11)</sup> for pressures up to 8000 atm at liquid-air temperature. A metal, such as beryllium copper, that is not subject to low-temperature embrittlement is required. Bridgman found that at 10,000 atm helium went through a tool-steel cylinder. The metallurgical structure of beryllium copper or of austenitic stainless steel might be superior in this respect, but a more promising approach would be to operate the intensifier cylinder well below room temperature (Fig. 5). By thus reducing the diffusion rate of helium through the cylinder, the pressure limit should be considerably increased. This requires that the hydraulic rams that drive the pressure

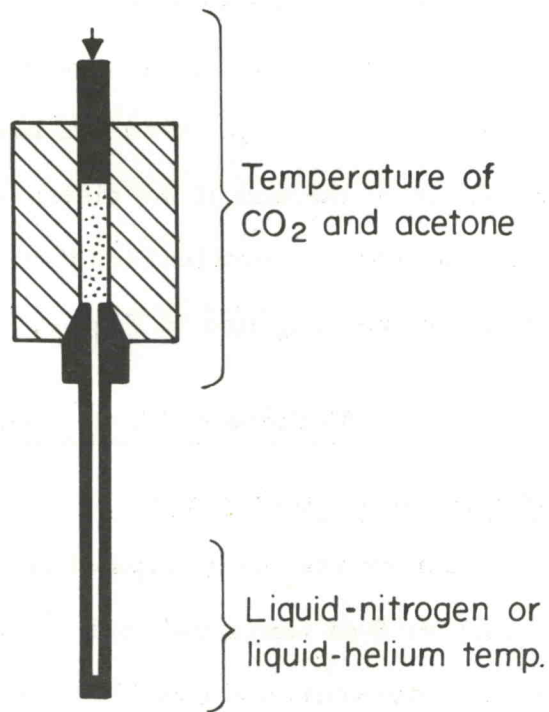


Fig. 5. Helium diffusion through a pressure vessel above 10,000 atm.

11) P. W. Bridgman, Proc. Am. Acad. Arts Sci. 67, 305 (1932); 68, 95 (1933).



piston and make the seals (e.g., in Fig. 30, p.35) be also at the lower temperature; they would have to be sealed with Teflon instead of rubber and powered with a compressed gas instead of oil. Electrical leads would be brought in at the end of the piston as illustrated in Fig. 13 (p.19). In an experiment where electrical leads are not used, it may be permissible to go below the freezing line in Fig. 2.

### Methods of Making Seals. The Hydraulic Seal

#### Pistons and closure plugs

There are many ways of sealing moving pistons, as illustrated in detail in Chapter 5 of Comings' book,<sup>9)</sup> for instance. For compressing liquids and gases at pressures above 5000 atm, however, the seal invariably used is the Bridgman unsupported area seal (Fig. 6). The gasket is tightened by the force of the fluid being compressed; the pressure in the gasket is proportionally higher by the ratio piston-head area/gasket area. Metallic wedge rings prevent the gasket material from extruding; the best available gasket material<sup>12)</sup> is Teflon filled with 3% of molybdenum disulphide. Rubber O-rings make excellent high-pressure seals.<sup>13)</sup> It is a convenience for certain applications to

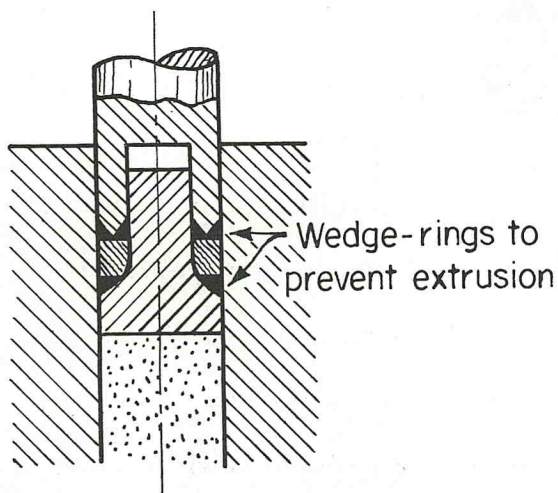


Fig. 6.

Bridgman's<sup>1)</sup> piston packing, showing antiextrusion rings used in Fig. 27 (p. 30).

12) H. A. Bowman, J. L. Cross, D. P. Johnson, J. D. Hill, and J. S. Ives, *Rev. Sci. Instr.* 27, 550 (1956).

13) A. W. Lawson, *ibid.* 25, 1136 (1954); W. B. Daniels and A. A. Hrushka, *ibid.* 28, 1058 (1957).

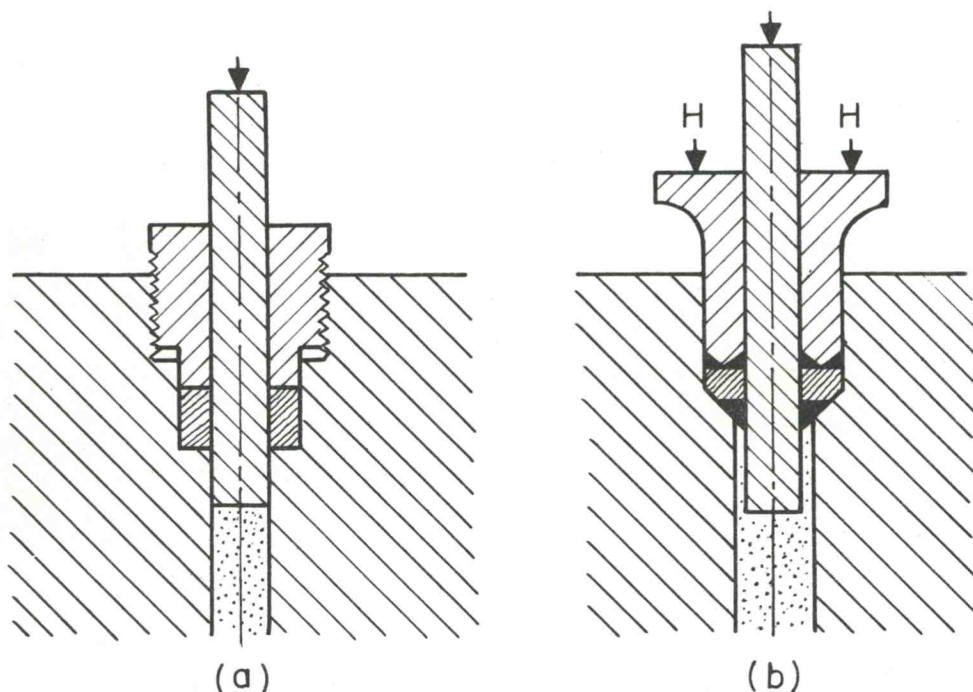


Fig. 7. Externally loaded seal for piston used in Figs. 22 (p.25) and 30 (p.35).

have the mushroom stem extended all the way through the piston (cf. Fig. 27, p.30).

Figure 7a illustrates the usual way of packing pistons for relatively low pressures; many variations thereof are discussed by Newitt<sup>8)</sup> and Comings.<sup>9)</sup> It is not difficult, however, to tighten the gasket by a hydraulic thrust  $H$  (Fig. 7b) instead of taking up on a large screw. Figures 22 (p.25) and 30 (p. 35) illustrate an intensifier that makes use of this procedure.<sup>14, 15)</sup> An advantage over the seal of Fig. 6 is that the inside bore of the cylinder does not have to be accurately lapped or even smooth. The cylinder therefore need not be particularly hard and, since it requires very much less machine work, it can be considered expendable. An intensifier such as that of Fig. 30 (p.35) has

14) N. L. Allen, Tech. Rep. 107, Lab. Ins. Res., Mass. Inst. Tech., April, 1956.

15) P. W. Forsbergh, jr., Prog. Rep. No. XXIV, Lab. Ins. Res., Mass. Inst. Tech., January, 1959, p. 31.



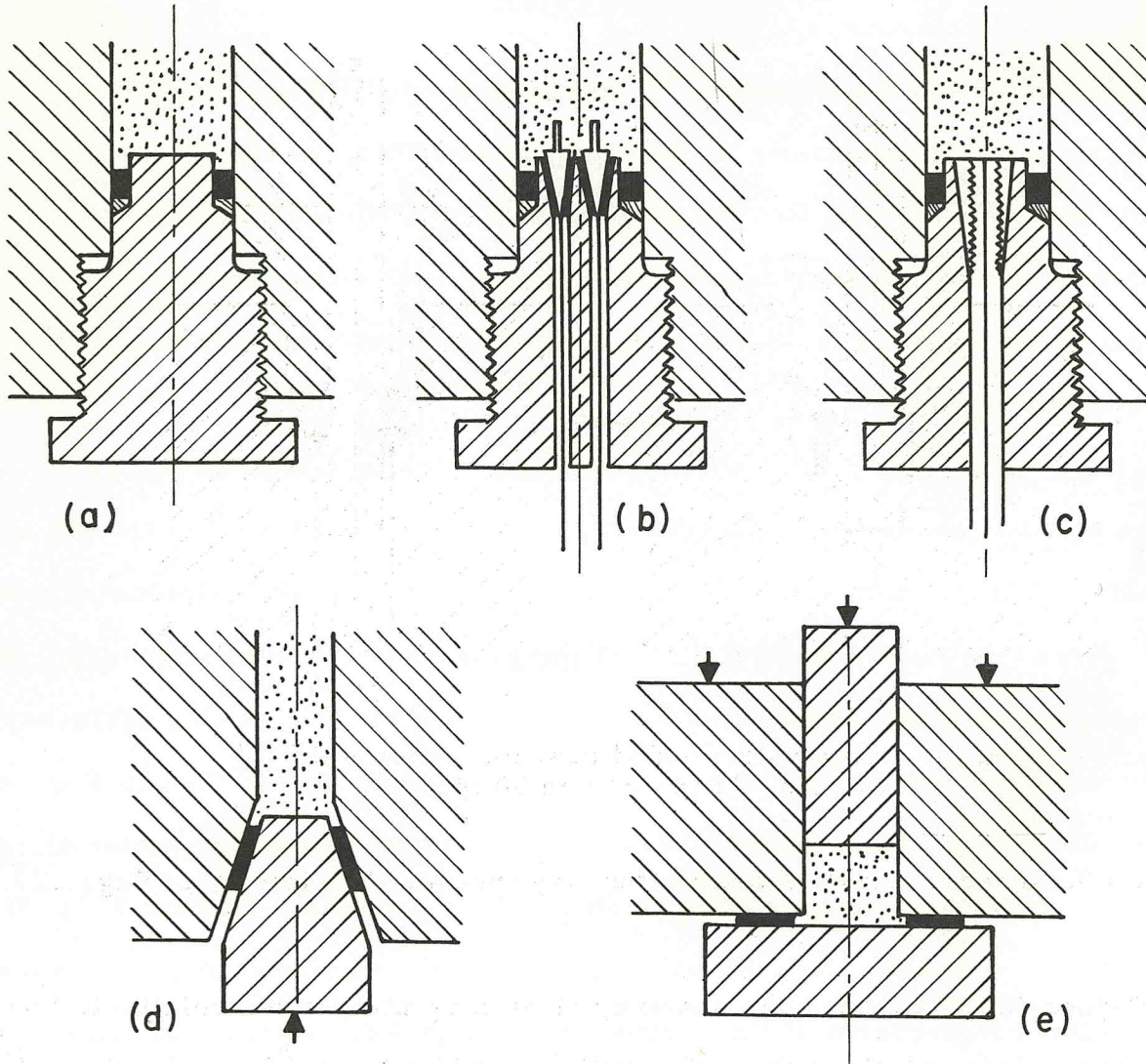


Fig. 8. (a) and (b) schematic of Bridgman's<sup>1)</sup> method for sealing a plug and electrical terminals in a plug; (c) method for bringing in a fine stainless steel tube;<sup>16)</sup> (d) and (e) plugs sealed by a simple gasket that is externally loaded (as used in Figs. 26, 27, and 30, pp. 29, 30, and 35); metal seal in (e) is analogous to Bridgman's pipestone seal in Fig. 1.

a few more parts than usual, but those that deteriorate with use (the piston and pressure cylinder) are of much simpler design.

Bridgman's method of sealing a closure plug in a high-pressure cylinder is shown schematically in Fig. 8a. Such a plug may be used to bring electrical terminals into the pressure vessel (Fig. 8b) by Bridgman's pipestone-cone method, or to connect a small tube of beryllium copper or 316 stainless by the

methods of Paul and Warschauer.<sup>16)</sup> The square ring in Figs. 8a, b, c is pushed into a V-shaped groove by the compressed fluid; as long as the ring has not flowed all the way into the groove, part of it remains unsupported and the corners of the ring bear against the cylinder wall with a pressure greater than that in the fluid. For 30,000 atm, several specially shaped rings have to be stacked together.<sup>1)</sup>

Figures 8d<sup>17)</sup> and 8e<sup>18)</sup> show a method of making a high-pressure closure that has proved very convenient in the systems of Figs. 26 (p. 29) and 27 (p. 30). A large hydraulic thrust compresses the conical or flat metal gasket to any given pressure; as in Fig. 1, if the gaskets are thin enough, say 0.020 inch, they cease flowing after they have been formed to the sealing surfaces and thereby work-hardened. The sealing area is much larger than in Figs. 8a, b, c and the gasket can be plated with a very thin layer of a softer material, such as copper, to fill up tiny scratches on the sealing surfaces. In Fig. 27 (p. 30), the large thrust making the seal adds a longitudinal component of compression to the radial compression of the shrunk-on outer jacket, a beneficial situation already described on p. 5.

#### Electrically insulating seals

Bridgman's method (Fig. 8b) consists of a conical metal plug insulated from the closure plug by a conical shell of pipestone, a material discussed in connection with Fig. 1. This seal holds pentane at 30,000 atm.<sup>1)</sup> For sealing a gas, the pipestone has to be soaked in rubber cement or, for low temperatures, in silicone oil.<sup>7)</sup> Alternatively, the pipestone cone may be followed

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16) William Paul and D. M. Warschauer, Rev. Sci. Instr. 27, 418 (1956);  
D. M. Warschauer and William Paul, ibid. 28, 62 (1957).

17) P. W. Forsbergh, jr., and G. J. Wolga, Tech. Rep. 111, Lab. Ins. Res.,  
Mass. Inst. Tech., October, 1956.

18) T. P. Janusz, Tech. Rep. 150, Lab. Ins. Res., Mass. Inst. Tech.,  
April, 1960.



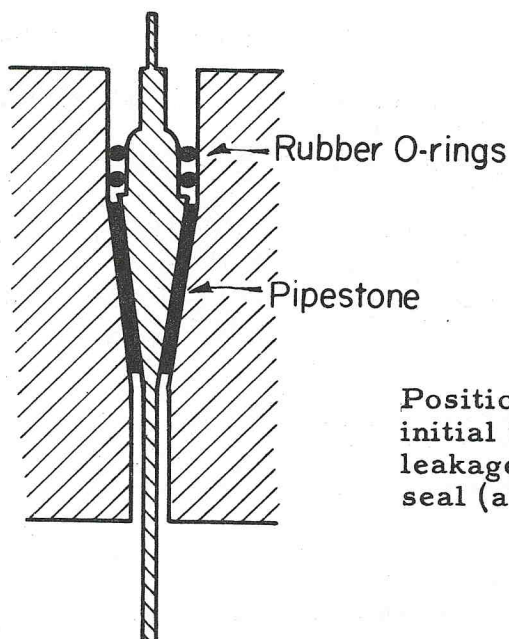


Fig. 9.

Position of rubber O-rings to ensure initial tightness and freedom from gas leakage through pipestone in Bridgman seal (as used in Fig. 27, p. 30).

by one or more tightly compressed rubber O-rings (Fig. 9); the latter are at first slid onto the thinner part of the stem above the conical plug, after which an initial application of pressure forces them into the tighter space. To ensure tightness further, oil or grease may be put above the O-rings after they have been seated; at high pressure oil and grease harden but by that time they have worked themselves into the smallest leakage paths, such as fine scratches. In making the conical insulating shell, the pipestone is cemented onto the electrode cone and then turned down to the required thickness (about 0.008 inch).

Figures 23 (p. 26) and 24 (p. 27) illustrate two coaxial pressure cells for dielectric measurements at high frequencies.<sup>19, 20)</sup> Here the insulator is a carefully machined piece of fused quartz. The conical center conductor and the outer conductor, in which the quartz cone is seated, are of Invar. The Invar pieces are plated with silver or gold and carefully lapped to the

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19) M. G. Vallauri and P. W. Forsbergh, jr., Rev. Sci. Instr. 28, 198 (1957).

20) W. B. Westphal, E. B. Littlefield, and P. W. Forsbergh, jr., Prog. Rep. No. XXI, Lab. Ins. Res., Mass. Inst. Tech., June, 1957, p. 9.

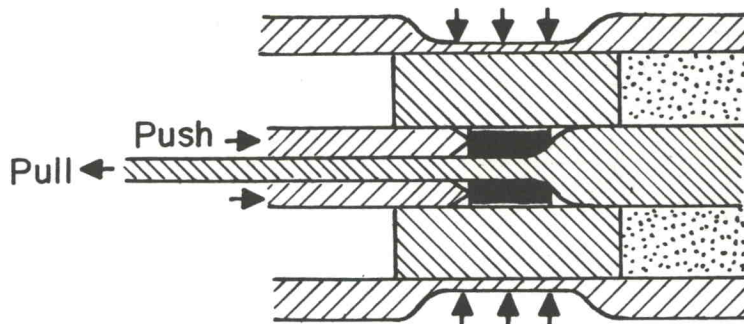


Fig. 10. Possible use of friction to hold and seal a cylindrical window in a coaxial transmission line.<sup>21)</sup> Seal on o.d. made by hydraulic pressure. Seal on i.d. made by pressure in Teflon exerted by a pull rod (as in Fig. 27, p. 30); a thin silver sheath encases Teflon for electrical continuity.

quartz cone. The electrical characteristics of a conical seal such as this are impossible to calculate theoretically. A cylindrical plug held in place by frictional force resulting from a hydraulic pressure around the outer conductor and inside the inner conductor (Fig. 10) has been suggested by Littlefield.<sup>21)</sup>

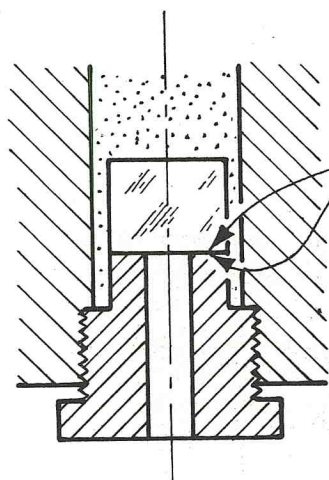
The best window design<sup>22)</sup> for a high-pressure vessel is a thick disk of quartz or sapphire (or germanium for infrared studies<sup>7)</sup>) having one optically flat face placed against the mating surface on the closure plug (Fig. 11). The

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21) E. B. Littlefield, private communication.

22) T. C. Poulter, Phys. Rev. 35, 297 (1930).





Optically flat  
surfaces

Fig. 11.

Poulter<sup>22)</sup> method of sealing a  
window.

stresses causing windows to fail at high pressures depend on the geometry and relative compressibilities of the plug and the window.

Optically flat quartz disks have been used extensively for windows and electrical insulators.<sup>23)</sup> An example of their use is shown by the high-voltage pressure cell of Fig. 25 (p. 28), where the optically flat quartz seals impart dimensional stability and do not expose the compressed gas to soluble contaminants in rubber or other sealing materials. Waveguide windows can also be made in this manner,<sup>24)</sup> and an elegant quartz-sealed microwave cavity has been designed by Westphal (Fig. 12).<sup>25)</sup>

When piston seals are made, as in Figs. 7b and 30 (p. 35), electrical connections can be brought into the cylinder through the piston itself, as illustrated in Fig. 13. Insulating and metallic washers are placed in alternating sequence on the end of the piston, and wires are soldered to the edges of the metallic washers and taken out through a hole in the piston. Thus, any number of wires can be brought to a sample (e.g., in the lower end of the apparatus in Fig. 5).

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23) A. Michels and co-workers, numerous papers in *Physica*.

24) C. S. E. Phillips, *J. Chem. Phys.* 23, 2388 (1955).

25) W. B. Westphal, Prog. Rep. No. XVII, Lab. Ins. Res., Mass. Inst. Tech., June, 1955, p. 53.

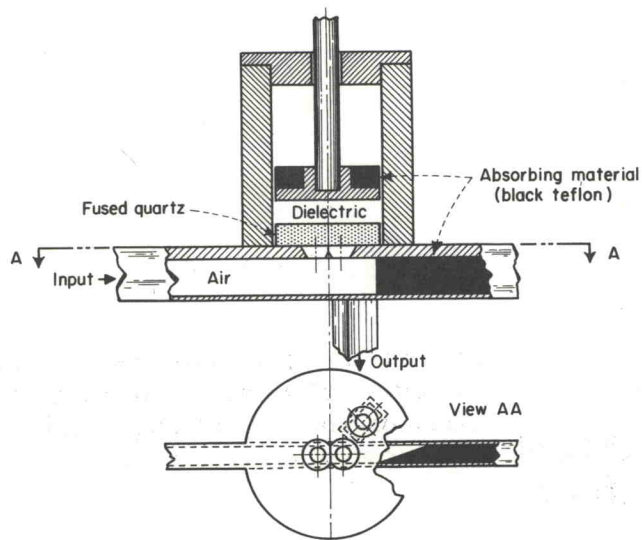


Fig. 12.

Possible application of quartz or sapphire disk seal to microwave cavity resonator.<sup>25)</sup>

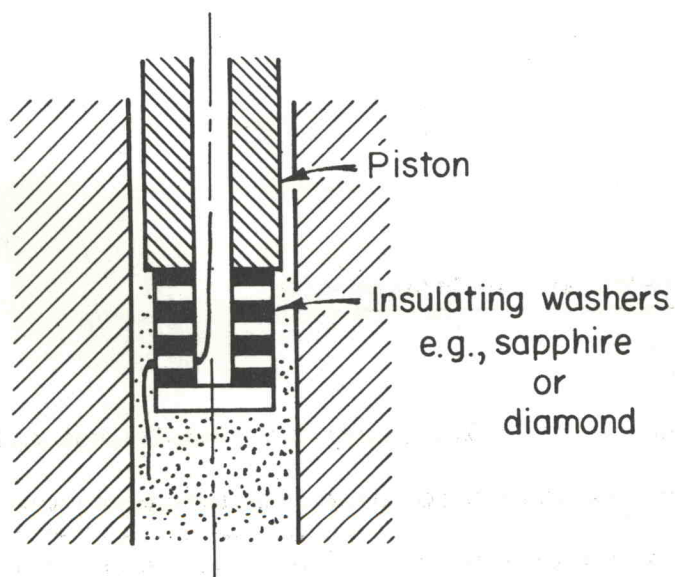


Fig. 13.

Possible application of disk seals to bring leads in through piston as sealed in Figs. 7.

Pressure seals such as used in Figs. 7 and 30 (p. 35) are suitable for the design of an optical absorption pressure cell having remotely controlled window separation (Fig. 14). This allows the path length for absorption to be controlled without taking off the pressure or removing the cell from the spectrophotometer. Furthermore, the windows can be brought exceedingly close together.



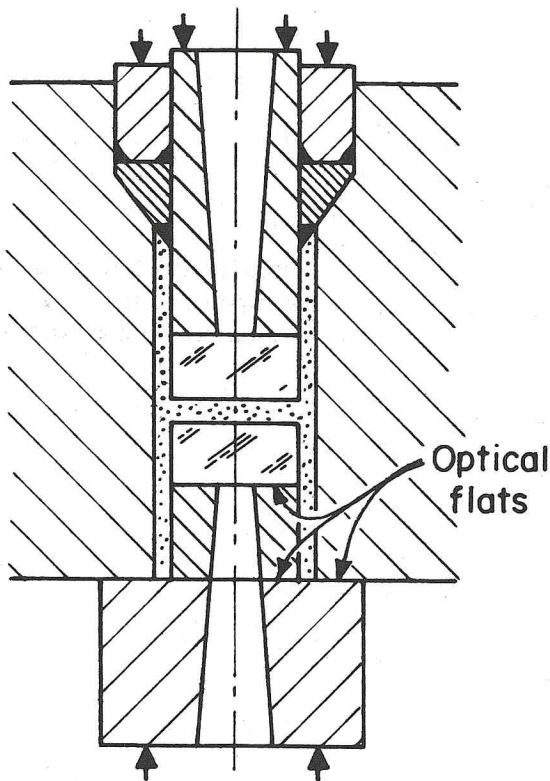


Fig. 14.

Possible application of sealing methods of Figs. 7 to make a controllable-clearance optical absorption cell.

Examples of High-Pressure Design

Two-dimensional pressure holders

A uniform circular disk subjected to radial pressure  $p$  on its cylindrical surface (Fig. 15) is under a uniform two-dimensional stress throughout the disk. Using Cartesian reference coordinates with the  $Z$  axis perpendicular to the disk, the stress system is represented by the tensor on the left of the following equation which is the sum of a hydrostatic pressure  $p$  and an equal mechanical tension  $p$  acting along the  $Z$  axis:

$$\begin{pmatrix} -p & 0 & 0 \\ 0 & -p & 0 \\ 0 & 0 & 0 \end{pmatrix} = \begin{pmatrix} -p & 0 & 0 \\ 0 & -p & 0 \\ 0 & 0 & -p \end{pmatrix} + \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

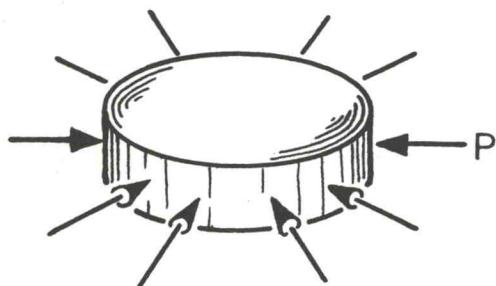


Fig. 15.

Production of two-dimensional pressure in a disk.

Brittle materials, such as crystals of barium titanate, cannot be subjected to much tensional stress, but they will support a large compressional stress because they do not flow plastically with the readiness of metals. Thus one can study the effect of an elongational elastic deformation on the properties of a brittle crystal without having to pull on it. The effect of nonhydrostatic stresses on disks cut in various crystallographic orientations gives much information about the equation of state of a crystal, as discussed, for instance, in a review on piezoelectricity published by the author.<sup>26)</sup>

Figure 16 illustrates a holder by means of which two-dimensional pres-

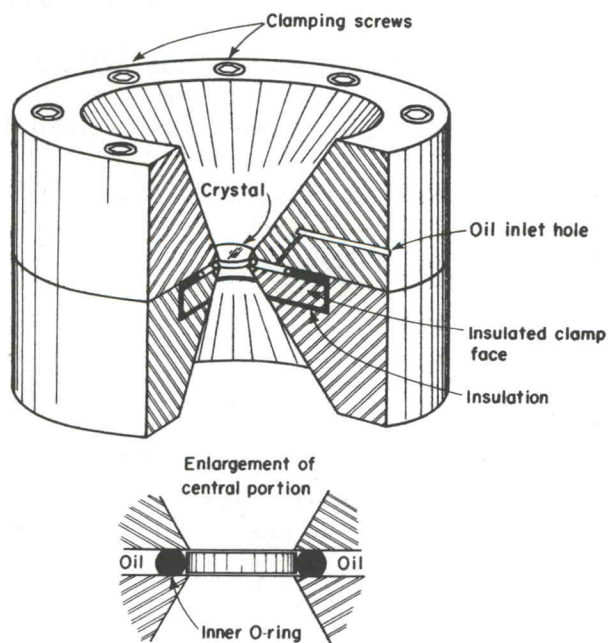


Fig. 16.

Two-dimensional pressure holder.

26) P. W. Forsbergh, jr., "Handbuch der Physik," Vol. 17, Springer, Berlin, 1956, p. 264.



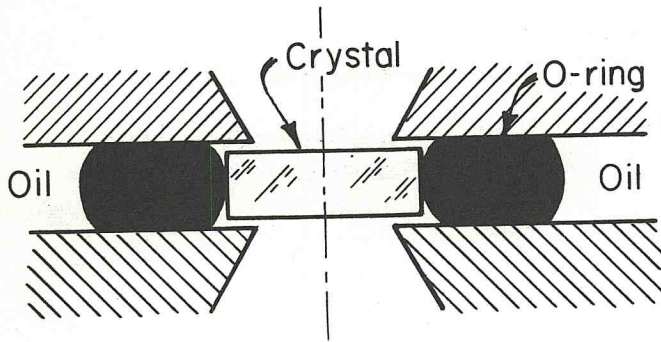


Fig. 17.

Two-dimensional pressure method used in Fig. 16.

sure<sup>27)</sup> can be applied. A rubber O-ring of a thickness 10 - 30% greater than that of the crystal disk is placed around the disk. The combination is then put between the two flat faces of a metal clamp (Fig. 17), which are brought together just enough to compress the O-ring and avoid squeezing the crystal faces. The clamp faces have holes through which to observe the crystal. Oil pressure is built up behind the O-ring. Soft rubber has a low modulus of shear and the oil pressure will therefore be transmitted very nearly hydrostatically to the cylindrical surface of the crystal disk. If the edges of the disk are not slightly rounded, small pieces of rubber may be nibbled off the O-ring and become wedged in the small clearance between the face of the disk and that of the clamp. This will create deviations from the purely two-dimensional stress pattern of the above equation. If the edges of the clamp hole are profiled (Fig. 18), such pieces of rubber work themselves out of the clearance.

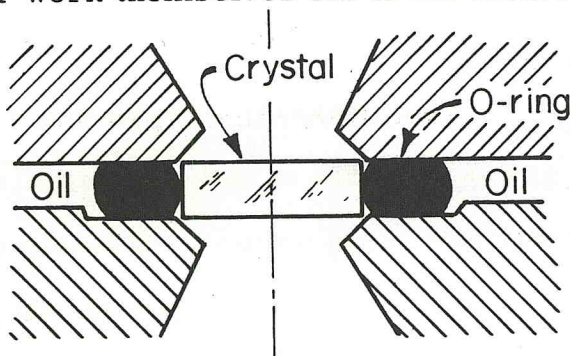


Fig. 18.

Contouring of jaws of two-dimensional holder for small crystals.

27) P. W. Forsbergh, jr., Phys. Rev. 93, 686 (1954). Note: Effect measured is spurious and treatment incorrect; two-dimensional technique has since been improved (Figs. 18 and 19).

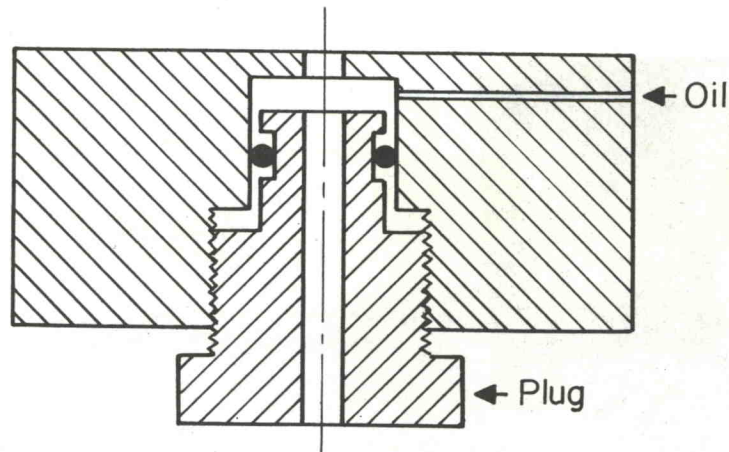


Fig. 19. Simplified two-dimensional holder.

An improved holder, used successfully on crystal disks of barium titanate only 0.020 inch thick and 0.040 inch or 0.080 inch in diameter is shown in Fig. 19. The base plug is held in a vise; the crystal disk is placed in the O-ring and the combination carefully placed on top of the plug. The body of the holder is then slowly screwed onto the plug until there is about a 0.0005 inch clearance on either side of the disk faces. If the disk is 0.020 inch thick, this clearance allows a perpendicular expansion of 5% when the two-dimensional pressure is applied.

Figure 20 shows a rotatable two-dimensional pressure holder<sup>28)</sup> mounted in a Norelco X-ray camera.

### Precompressors

In order to minimize the volume of a high-pressure intensifier, such as in Figs. 27 (p. 30) and 30 (p.35 ), the gas is admitted at tank pressure (about 100 atm) into an auxiliary cylinder of relatively large volume. A piston in this cylinder, or precompressor (Figs. 21 and 22), displaces the gas into the high-pressure cylinder until 2000 - 3000 atm is reached. At this pressure the den-

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28) D. J. Epstein, P. W. Forsbergh, jr., and S. C. Abrahams, Prog. Rep. No. XV, Lab. Ins. Res., Mass. Inst. Tech., June, 1954, p. 28.



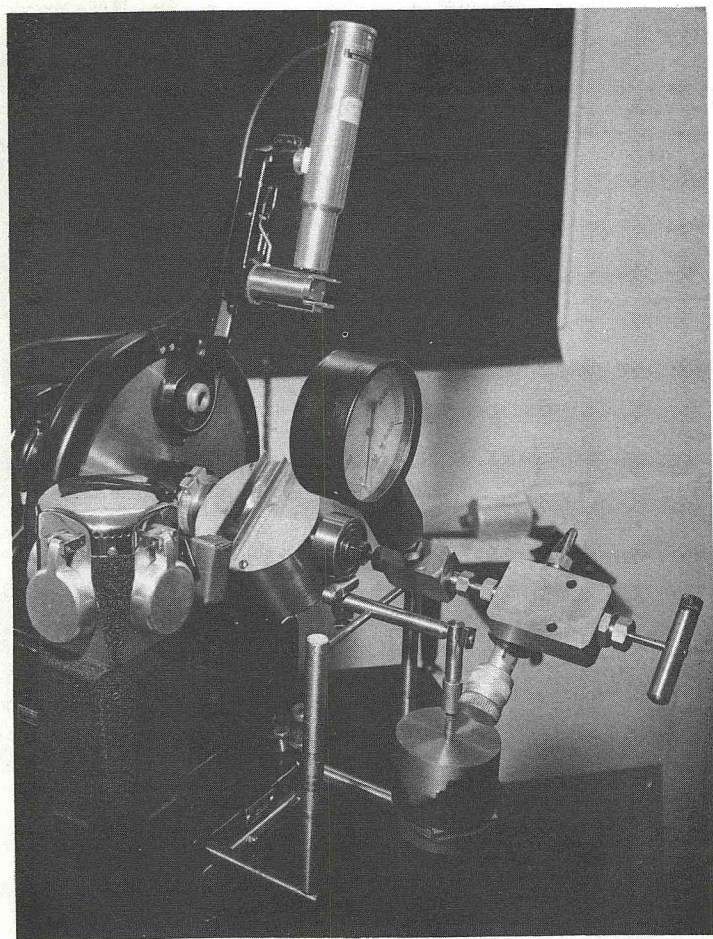


Fig. 20. Two-dimensional holder for X-ray diffraction work.<sup>28)</sup>

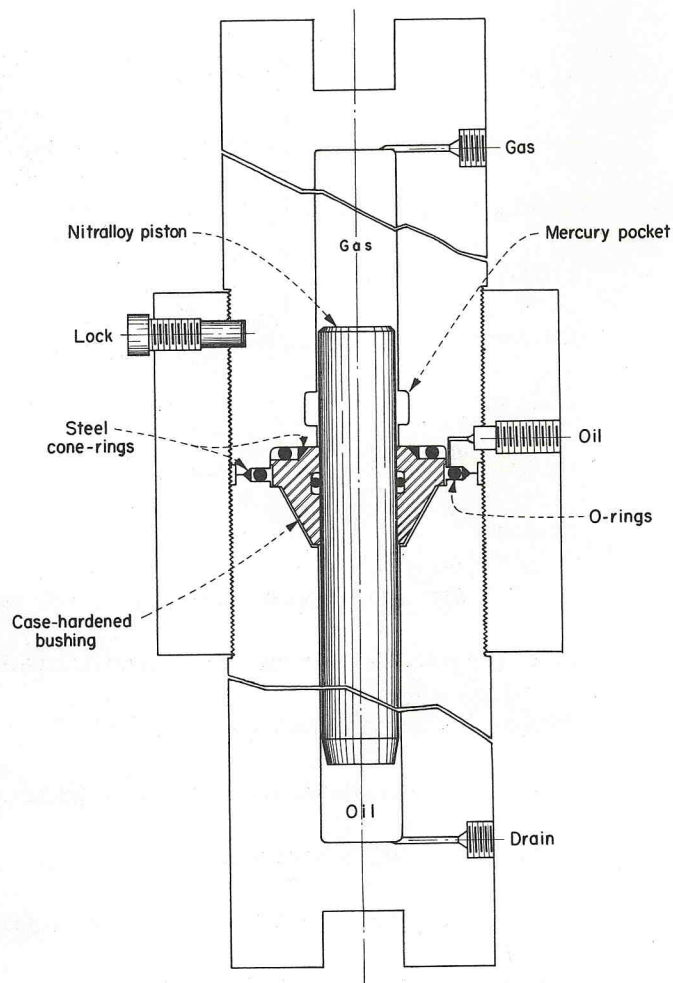


Fig. 21. Gas precompressor with bushing tolerance retained by the fluid pressure.<sup>19)</sup>

sity of the gas is comparable with that of ordinary liquids.

In Fig. 21, a Nitralloy piston is displaced by pumping oil in beneath it.<sup>19)</sup> The piston rides through a case-hardened bushing containing an O-ring that separates the gas from the oil. The bushing is surrounded by the same pressure on all sides; the very close tolerance to the piston is thereby retained regardless of pressures, as in the controlled-clearance piston used by Newhall<sup>29)</sup> for pressure balances. The friction is low enough for the piston to be displaced by tank pressure alone. The O-rings and triangular metal backing-rings are in-

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29) Donald E. Newhall, Ind. Eng. Chem. 49, 1993 (1957).

stalled so that an accidental sudden loss of pressure on either the gas or the oil side will not result in damaged seals; The gas and oil may also be throttled by constructions at the pipe fittings to prevent the piston from accidentally picking up so much speed as to hit the end of the cylinder with force.

Any precompressor packed with rubber or leather may cause contamination of the compressed gas. Traces of oil or other impurities will give rise to serious difficulties in optical-absorption and electric-conduction measurements in compressed gases. The chemically most resistant plastic is Teflon, which has a relatively low coefficient of friction even when not filled with a lubricant such as molybdenum disulphide. Figure 22 illustrates a precompressor having a Nitralloy piston and a hydraulically loaded Teflon seal of the kind illustrated in Fig. 7b (p. 13 ). It was built for use with the apparatus of Fig. 25 (p. 28).

Systems with lapped quartz seals

Figures 23 and 24 illustrate two coaxial pressure cells for dielectric

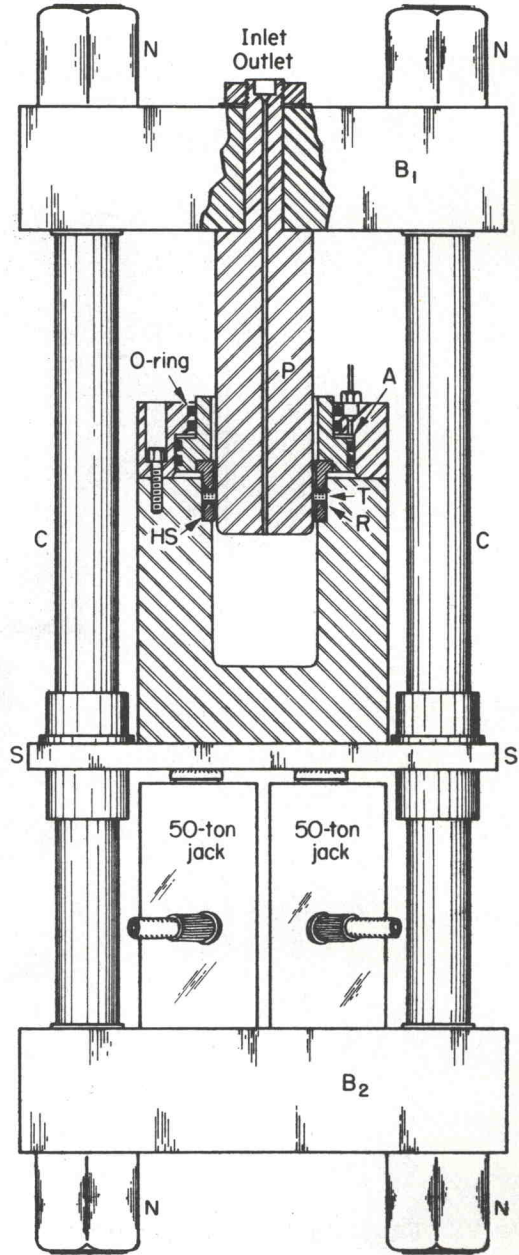


Fig. 22. Gas precompressor<sup>14)</sup> with piston sealed by external hydraulic load (as in Figs. 7).



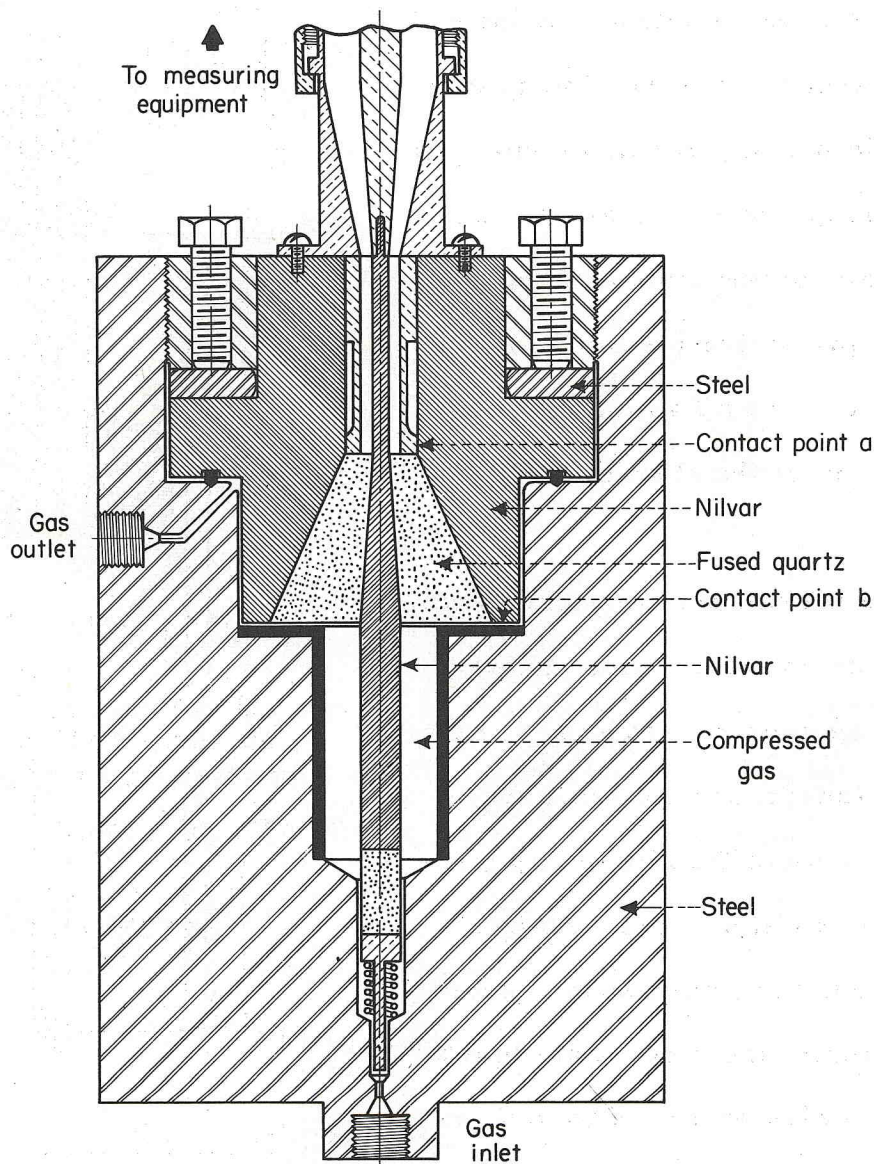


Fig. 23. Microwave coaxial cell for measuring dielectric constant and loss in compressed gases.<sup>19)</sup>

measurements at high frequencies. One is a line-terminating cell and the other a transmission cell. In the first,<sup>19)</sup> the quartz-cone seal discussed in the preceding part is held in an Invar closure head pressed (by tightening Allen bolts) onto a copper gasket in the conventional manner. It holds nitrogen at 1700 atm without appreciable leak, although at higher pressures the conventional copper gasket begins to leak. In the second,<sup>20)</sup> the seal is made by a hydraulic thrust

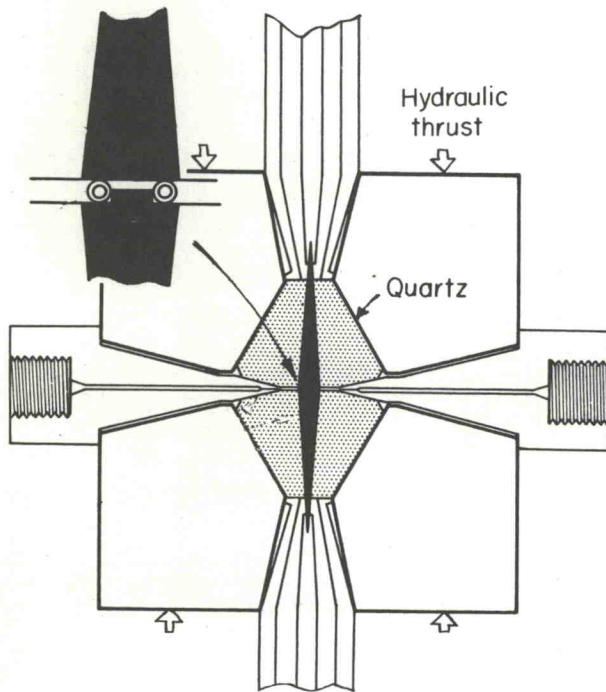


Fig. 24.

Microwave transmission cell for dielectric studies of fluids more lossy than can be used in Fig. 23.

that squeezes the more obtuse faces of the quartz cones against a gold-plated stainless-steel spacer between them. Two tiny holes in the spacer allow the sample fluid to be brought in and flushed out. The dimensional stability of the assembly permits the Invar cones that form the inner conductor to be connected electrically by means of a small ring compressed between them in the process of assembly; the ring is made of fine tubing of silver- or gold-plated stainless steel.

#### Systems with optically flat quartz seals

The apparatus of Fig. 25<sup>14)</sup> exemplifies the use of optically flat quartz surfaces in insulating seals. The large standoff insulator that holds the high-voltage electrode is also made of fused quartz.

#### Systems with hydraulically loaded closures

Figures 26 and 27 illustrate systems with hydraulically loaded gaskets (cf. Figs. 8d and 8e, p. 14). In Fig. 26 this gasket was a piece of copper pipe spun into a cone and annealed. No machining tolerances had to be observed in



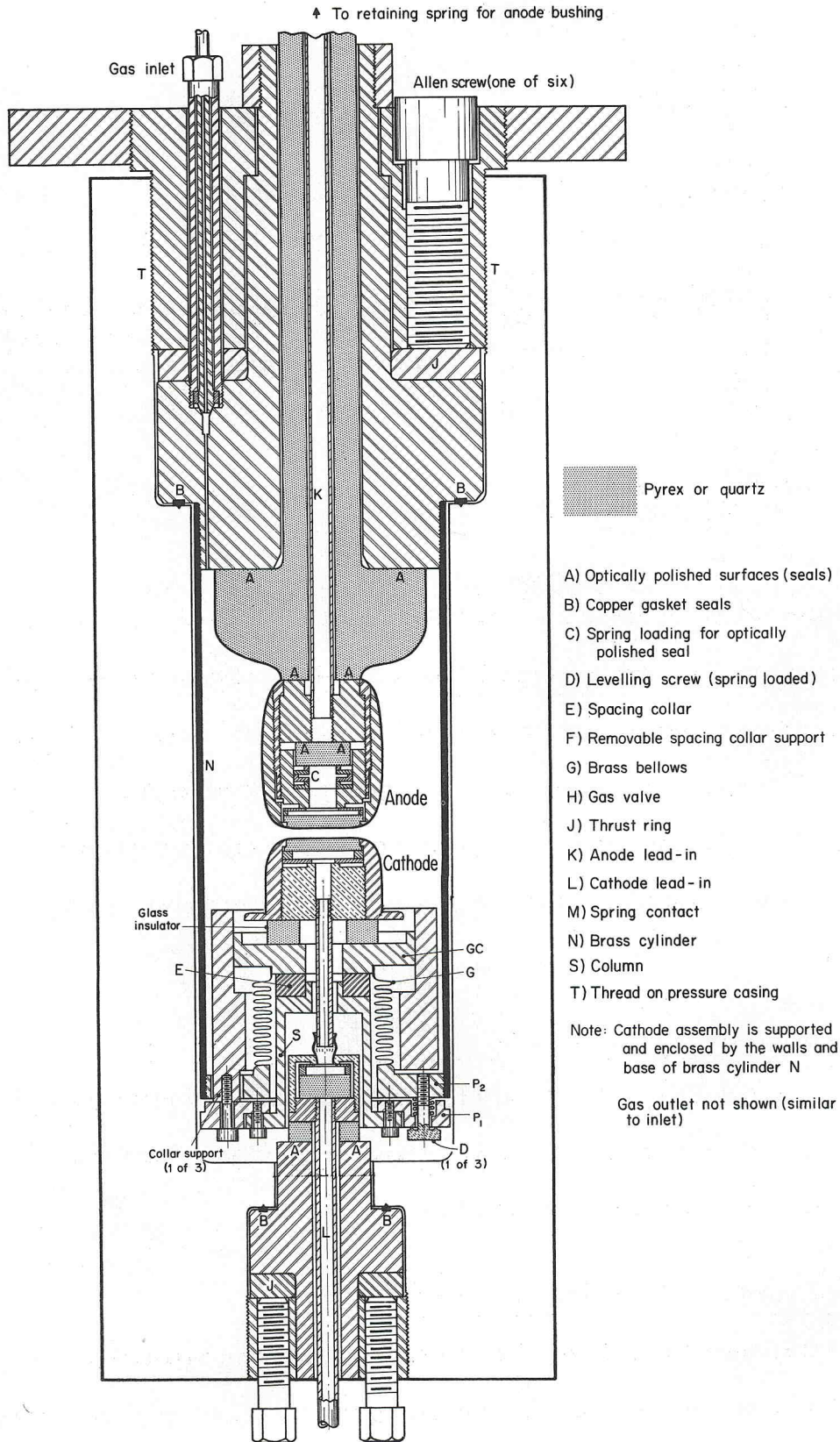


Fig. 25. High-voltage pressure cell for conduction studies in gases.<sup>14)</sup>

making the mating parts of the seal. The system held about 20,000 atm for 24 hours without any need for adjusting the hydraulic oil pressure that holds the seal and the piston. Because the cone angle was relatively small, the plug, after release of pressure, had to be removed with a hydraulic cork-puller. In Fig. 30 (p. 35) the same type of seal, using copper-plated iron or stainless steel, has a larger angle and the plug can be lifted out without the slightest sticking.

Figure 27 shows a system designed for adaptation to a variety of experimental purposes. Pre-compressed argon at 2000 atm, from the pre-compressor of Fig. 21, is brought into the cylinder via the dashed line beginning at 4. Hydraulic oil pressure on line 2 forces the pressure cylinder down onto the flat gasket (shown in detail in the inset). This seals off the cylinder from the precompressor and the pressure in the latter may be reduced if desired. Hydraulic oil pressure at 1 then drives the piston into the cylinder and the piston displacement is measured by a dial gauge at 3. The stem 3 allows the piston to be drawn out of the cylinder without losing the mushroom at its lower end. It also allows initial pressure to be set up in the piston packing before pressure is built up in the pre-compressor; this is accomplished by pulling on the stem with a hydraulic device that grips the stem with a built-in hydraulic gripper.

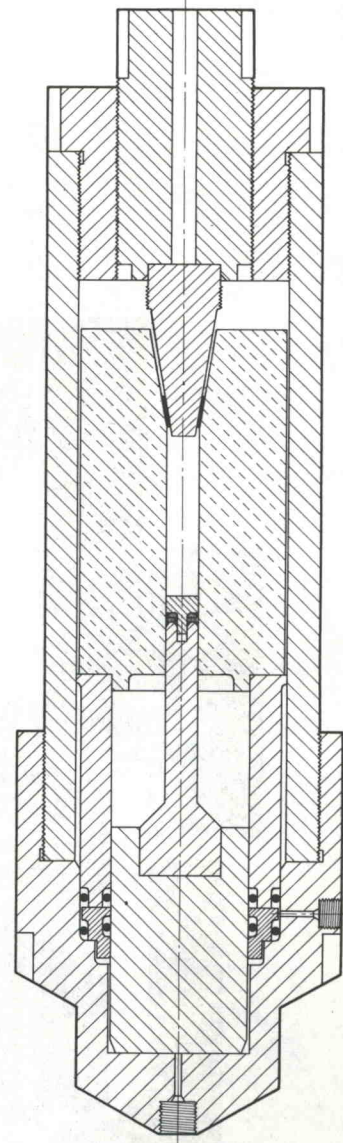


Fig. 26.

Vessel with plug sealed by external hydraulic load on conical gasket.<sup>17)</sup>



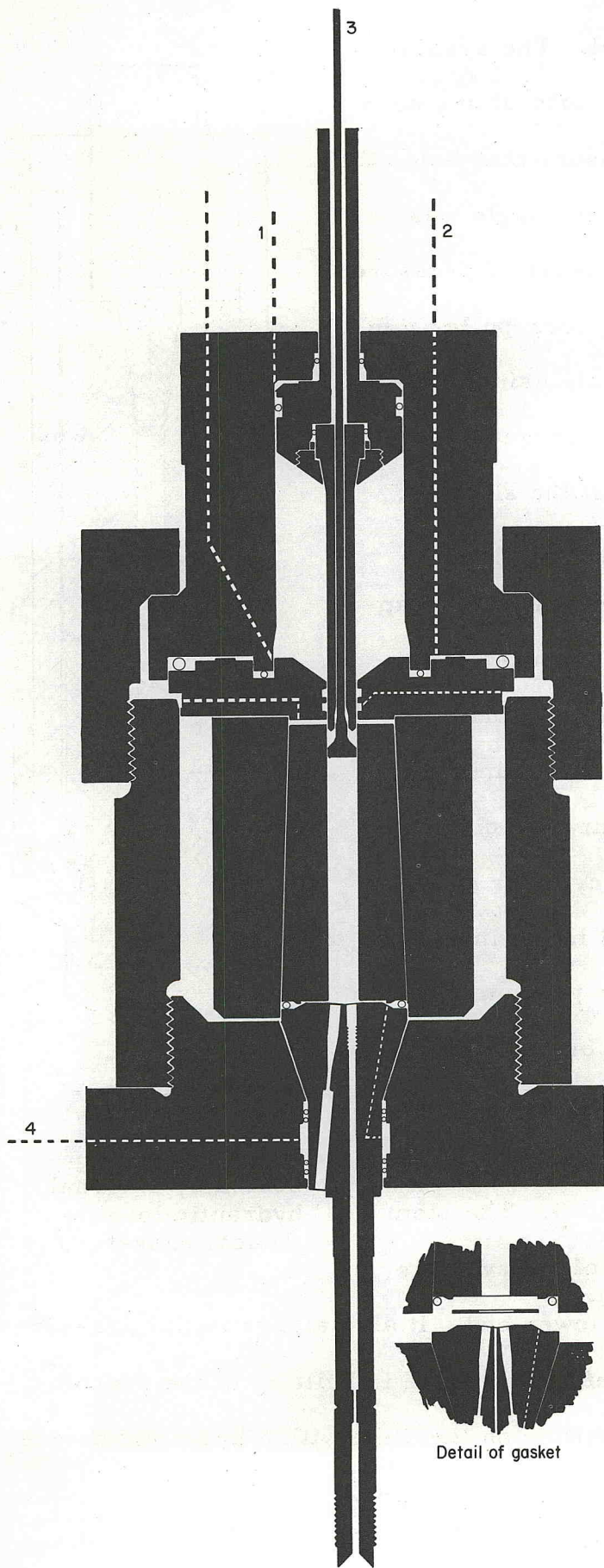


Fig. 27.

Vessel with plug sealed by external hydraulic load on flat gasket. 18)

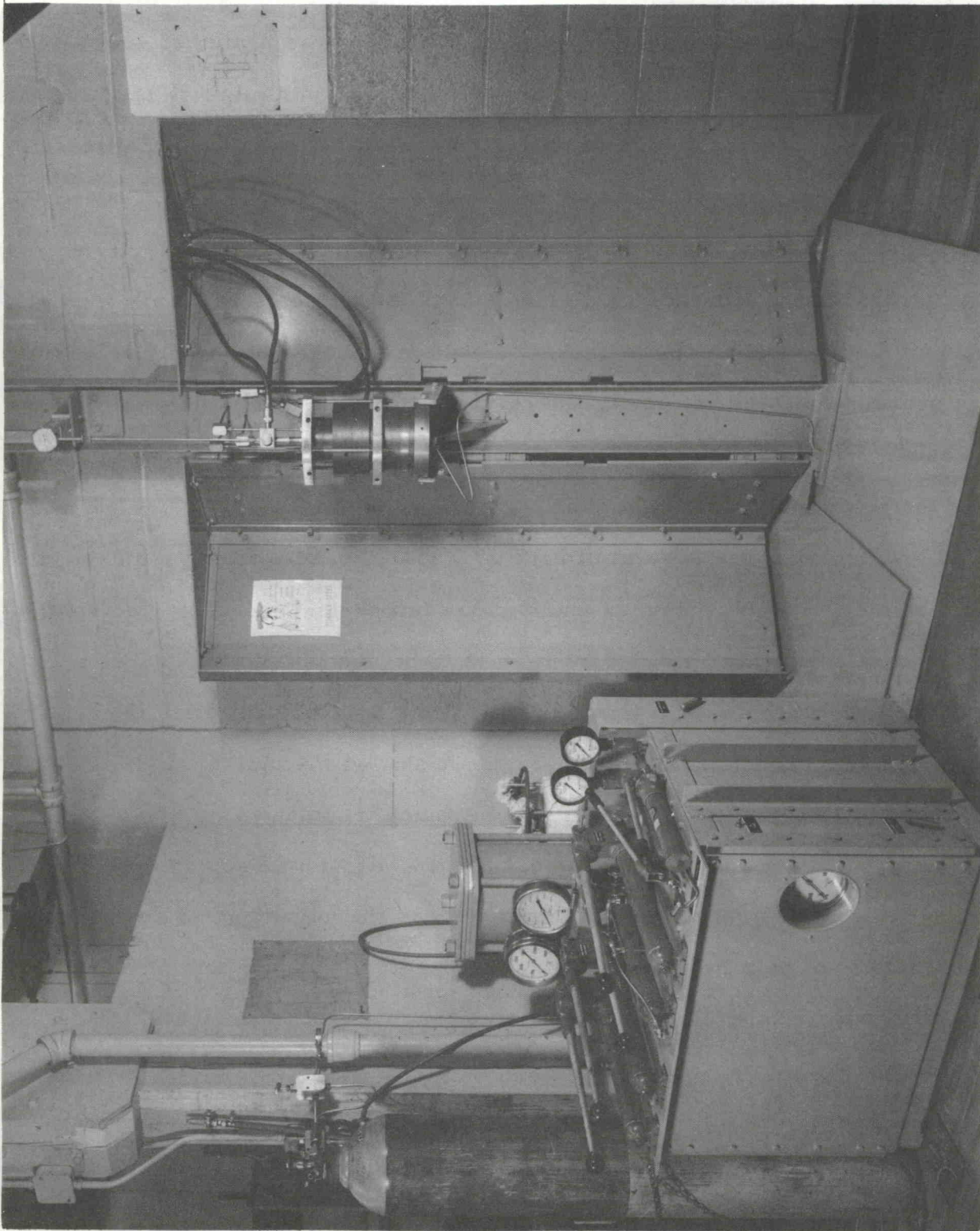


Fig. 28. Pressure intensifier of Fig. 27 with explosion-shield doors swung open. Foreground: console with hydraulic pumps for loading seals. Background: precompressor of Fig. 21, inside a burst shield. All pipes run under steel floor plate.



The inner part of the pressure cylinder was made of oil-hardening tough tool steel, heat-treated to Rockwell 53 C. The shrunk-on jacket was made of a hollow tool-steel bar (also oil-hardening tough steel); it was heated to 460°C and the inner part dropped into it, a stream of air being maintained in the bore to keep the bore surface cool. The resulting compound cylinder has a hardness 53 at the bore, decreasing with distance from the center.

The system of Fig. 27 is very easy to assemble and disassemble. The pressure cylinder has no threads or side holes. Positive seals are made every time, without having to replace the gasket. Seals are opened by simply releasing hydraulic pressure instead of moving the cylinder to a vise and using a large wrench. Figure 28 shows the intensifier and the console for the hydraulic pumps that make the seals and drive the pistons.

The lower part of the system of Fig. 27 was designed for length measurements of a sample at high pressures and various temperatures. For electrical-conductivity measurements, a different base is to be used (Fig. 29); wires are brought down to the sample through a long hole drilled in a solid rod of beryllium copper or age-hardening stainless steel. A skilled machinist can drill a 3/32-inch hole of 18 or 24 inches length without much trouble. This avoids making closures at the bottom, which would require a much larger sample cylinder. The smallness of the sample cylinder has the advantage of small heat capacity, which is of help in sweeping wide temperature ranges, and also that the sample cylinder can be placed in apparatus (such as pulsed magnets) in which the available space is small.

In Fig. 27 there is a thread for a throttling screw that controls the rate of passage of compressed gas from the cylinder in which the pressure is generated; this avoids crushing of the delicate device in the sample cylinder and restricts the outrush of gas in case the sample cylinder should burst. In Fig. 29

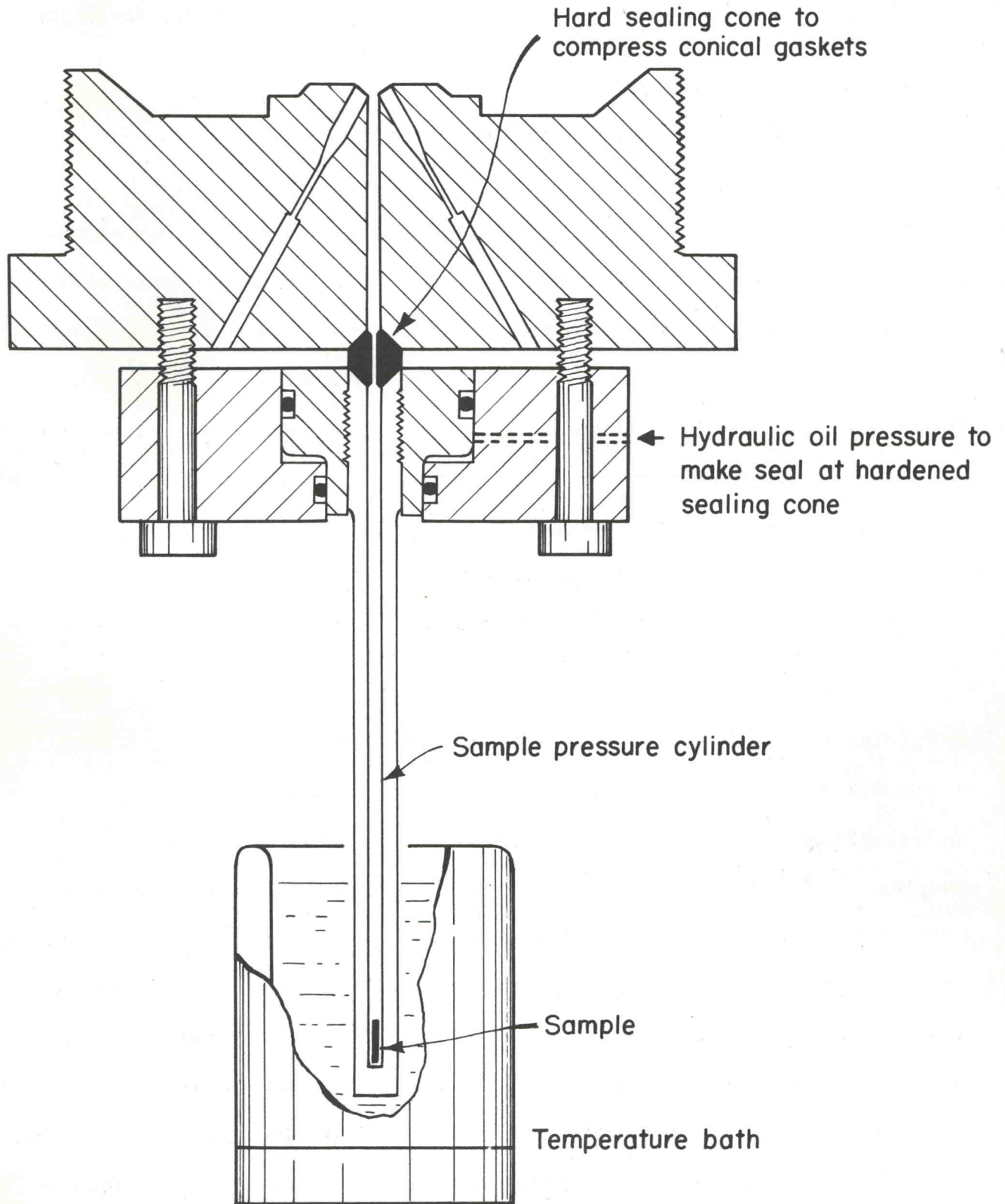


Fig. 29. Method of connecting sample tube to intensifier of Fig. 27 when dilatometer (Fig. 33, p. 38) is not in use.



it is possible to install a throttling cap over the electrodes for the same purpose.

### Systems with hydraulically loaded piston gaskets

Figure 30 illustrates an intensifier in which the piston slides through the packing as sketched in Fig. 7b (p. 13 ). The cylinder is of 17-4 PH but could equally well be of other materials. This intensifier has been tested successfully at 10,000 atm. The cylinder itself requires very little machine work and the bore is not even reamed smooth. The lower part of the assembly consists of a hydraulic device that pushes the piston in or pulls it out. The packing in this hydraulic device is of an unsupported-area type instead of being made with the usual plastic or leather cups found in commercial rams, and the design is such that there are no inside cylindrical surfaces requiring careful machining and finishing. The closure at the upper end of the pressure cylinder is of the type of Fig. 8d (p. 14 ) and the closure plug incorporates a bulk-modulus pressure gauge.<sup>30)</sup>

### Measurement of Pressure, Length, and Lattice Parameters

The most accurate measurements of pressure require a pressure-balance or free-piston gauge, a frictionless piston of known area loaded with known weights.<sup>1, 8, 9, 23, 29)</sup> A rough measure of pressure, sufficient for many studies in the solid state, can be obtained from the hydraulic oil pressure behind the pressure-generating piston in a system such as that of Fig. 27. The packing is of Teflon filled with 3% molybdenum disulphide.<sup>12)</sup> Figure 31 shows the piston displacement as a function of hydraulic oil pressure; this allows the correction for friction to be estimated. The usual way for measuring pressure, accurate to  $\pm 0.1$  %, is by the resistance change of a coil of manganin or gold-chrome wire.<sup>9)</sup>

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30) Donald E. Newhall, private communication.

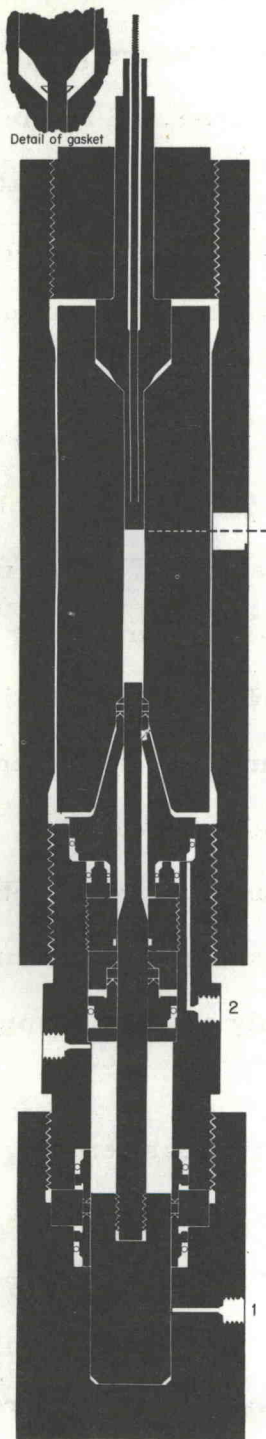


Fig. 30.

Intensifier in which seals are made according to Figs. 7b and 8b. A Newhall bulk-modulus pressure gauge is incorporated.

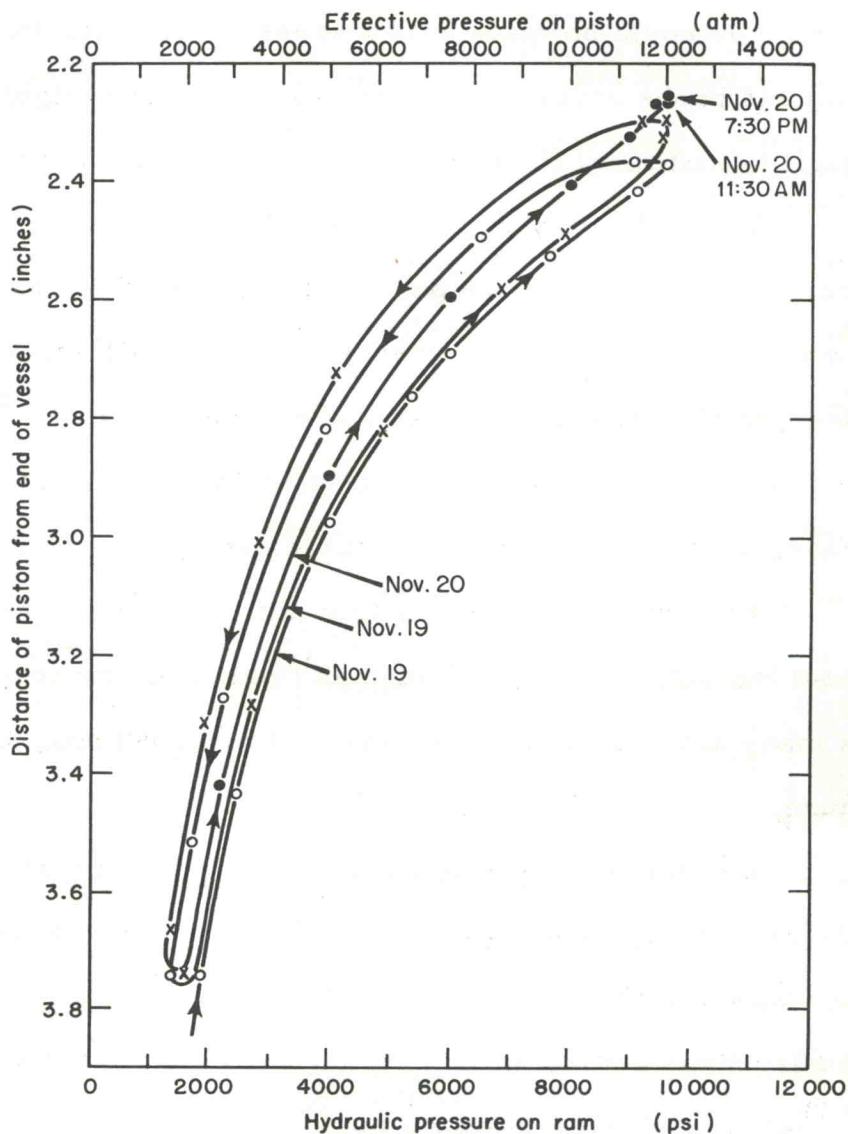


Fig. 31.

Piston displacement as a function of oil pressure in line 1 of Fig. 27. Argon initially at 2000 atm pre-compression pressure.



A very rugged type of pressure gauge has been developed recently by Newhall.<sup>30)</sup> This bulk-modulus gauge is incorporated in the closure plug of the intensifier in Fig. 30. Under pressure the probiscus in the upper end of the bore of the pressure cylinder contracts elastically and the stem inside it displaces upward. This displacement is measured by a pneumatic device. The system of Fig. 30 could be made with a pressure cylinder long enough to hold a bulk-modulus probe several feet long. Such a probe might have an accuracy and stability sufficient for a secondary standard of pressure in laboratory work.

From the point of view of solid-state theory, the significant variables are the changes in lattice parameters rather than the pressure. The use of X rays requires pressure vessels having beryllium windows<sup>31)</sup> (limited by the strength of beryllium) or diamond pressure vessels.<sup>32)</sup> Neutron diffraction is more promising because some metals, such as zirconium, are sufficiently transparent to neutrons for possible use as pressure vessels or windows.

For sufficiently symmetric lattices, the lattice parameter can be obtained from the volume alone; Bridgman<sup>1)</sup> has made measurements of volume change in many substances at pressures up to 100,000 atm, but only at room temperature.

For measuring length changes in single crystals at various temperatures and pressures, strain gauges are the most promising devices, although it is in principle possible to obtain length changes by integrating over measurements of elastic coefficients, at various temperatures and pressures, obtained by pulse techniques. A strain gauge, however, must be calibrated against a material on which absolute measurements have been made.

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31) R. B. Jacobs, Phys. Rev. 54, 325, 468 (1938).

32) A. W. Lawson and Ting-Yuan Tang, Third Annual Report to ONR on High Pressure Research, Institute for the Study of Metals, The University of Chicago, 1950, p. 64.

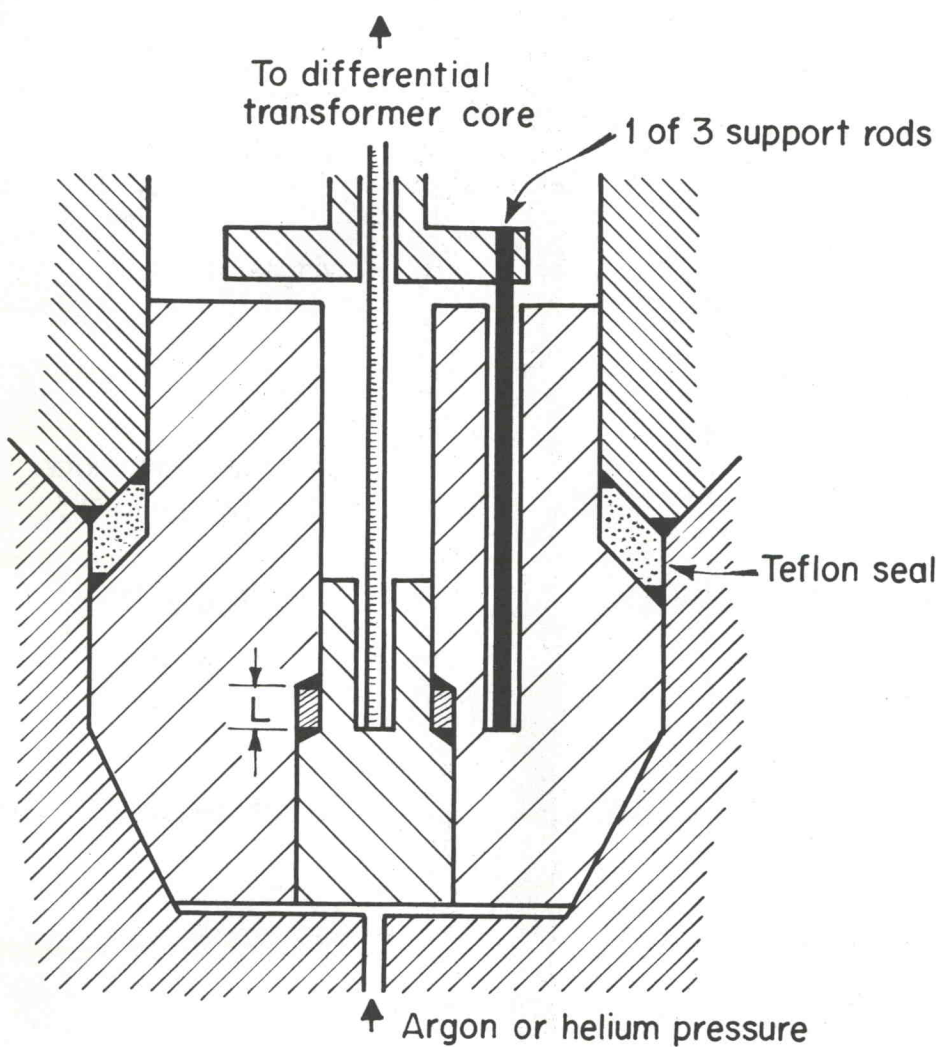


Fig. 32. Proposed method for measuring volume change at various pressures and temperatures in a soft-metal calibration sample of length  $L$ . Alternative is optical dilatometry.

An absolute measurement of volume at various temperatures could be made with a pressure cell as illustrated in Fig. 32, provided the metal measured is soft enough to be under approximately hydrostatic pressure. A cylindrical magnesium ring of length  $L = 1/4$  inch would shrink about 0.006 inch if the pressure in the magnesium were 10,000 atm. Correction for dimensional changes in the holder should account for less than 10% of the measured displace-



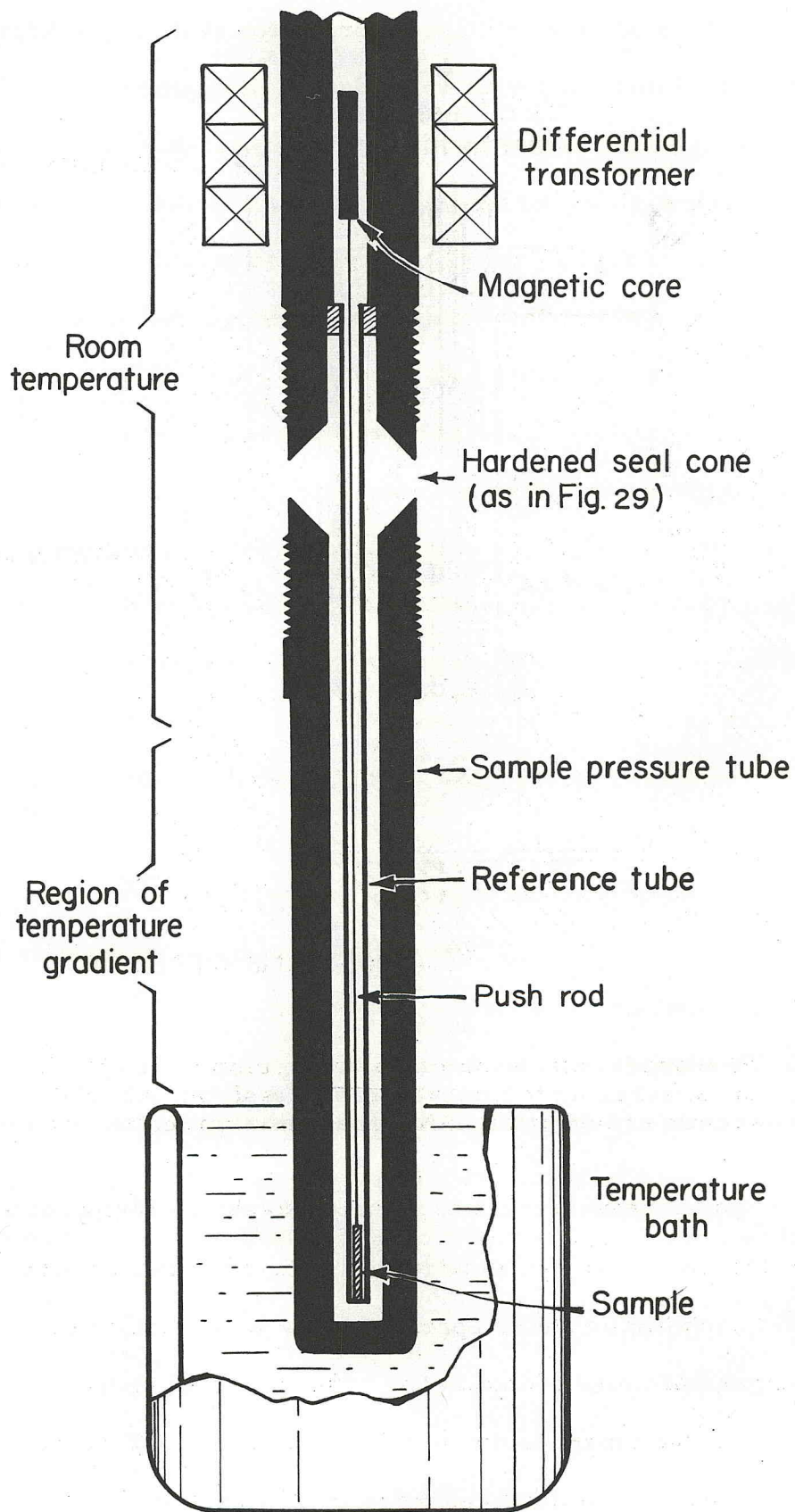


Fig. 33. Differential dilatometer<sup>18)</sup> for studying length changes at various pressures and temperatures.

ment. There will also be a frictional correction that can be kept at a minimum in a hardened die lubricated with molybdenum disulphide. A second way of measuring a change in length at low and high temperatures would be to use two tiny optical interference glasses above the sample, located in the bottom of a pressure tube such as in Fig. 29 (p. 33). One glass would rest on a ledge inside the pressure tube, the length of which can be measured externally. The high-pressure piston would be fitted with an observation window as in the upper part of Fig. 14 (p. 20). Optical interference was used successfully in the apparatus of Fig. 25 for measuring interelectrode separation.

The use of strain gauges may give rise to problems such as the limits of elastic deformation of the gauge element and the effect of compressed gas penetrating into the cement and disintegrating it on release of pressure. An alternative to strain gauges is illustrated in Fig. 33, where the upper pressure tube is that at the lower end of Fig. 27. Details are described in a report by Janusz.<sup>18)</sup> Briefly, the push rod and the reference tube in Fig. 33 should expand or contract equally in the region of temperature gradient. This is so if they are of the same metal and if the clearance between them is sufficiently small to ensure that corresponding points along their length are at equal temperatures. This appears to be the case in practice up to 150°C; at higher temperatures there are irregularities due probably to convection currents. These probably could be eliminated by having the tube horizontal instead of vertical; they should not occur for work at temperatures below room temperature.



