HIGH-PRESSURE APPARATUS

by

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Two types of ultrahigh-pressure, high-temperature apparatus are receiving rather extensive use at the present time. The first of these, the "Belt," utilizes conical "pistons" thrust into opposite ends of a properly shaped and gasketed chamber, that is, surrounded by a torroidal "belt" of supporting rings.[1] Figures 1 and 2 pertain to this apparatus. The second device is the "Tetrahedral Anvil Apparatus."[2,3] This device is geometrically the simplest of a series of related devices which made use of polygonal anvils that enclose regular polyhedra when fitted together about a common point. Pressure is generated by forcing the anvils against a polyhedral cell. Figures 3 and 4 pertain to this device.

Since these devices have already been described in Refs. 1, 2, and 3, they will not be discussed in detail here. Rather, three subjects pertinent to these and other high-pressure devices will be considered: (1) gaskets for obtaining relative motion of anvils or other pressure generating components, (2) solid pressure transmitting media, and (3) high tonnage hydraulic rams.

Gaskets

The use of a compressible gasket to obtain motion of anvils, conical pistons, or other components in high-pressure apparatus has become well established in the ten years since P.W. Bridgman reported his first anvil work.[4] Little has been done, however, to systematically determine the important variables that enable the gasket to perform its required function. An analysis of the functions performed by the gasket may help to indicate some fruitful areas for experimentation or innovation in this area.

The first function performed by the gasket is that of "yielding" to the thrust placed on it by a moving anvil or similarly functioning apparatus component. Yielding can occur through simple compression of the gasket material, by flow, or by a combination of compression and flow. The amount of yielding should be relatively large in order that the anvils might move a sufficient amount to compress the contents of the high-pressure chamber.

The second function of the gasket is that of "confining" (not yielding to the thrust of) the material being compressed by the advancing anvil. The tasks of yielding and confining are, of course, contradictory and must somehow be reconciled.

A third gasket function is that of "support." Just inside the inner edge of the gasket, the surfaces of the high-pressure apparatus components are subjected to the full pressure generated within the chamber. At the outer edge of the gasket, the apparatus components are subjected to only one atmosphere pressure. Ideally, the pressure exerted by the gasket against the components of the apparatus should gradually decrease from the full chamber pressure at the inner gasket edge to one atmosphere pressure at the outer edge. When this is the case, a sharp line of demarcation between chamber pressure and atmospheric pressure is avoided and consequently there will be no line of high stress concentration. If the gasket width and pressure gradient are judiciously chosen, the apparatus components will support each other and make it possible for the highly stressed portions of the components to withstand more load than would otherwise be possible. Pressure gradient patterns can be varied by choice of gasket materials and by changing the cross-sectional profile of the gasket from the inner to the outer edge.

The gasket functions of yielding, confining, and supporting enumerated above, must, in any given design, be accomplished without having the gasket absorb an inordinate fraction of the ram thrust available for operating the device. In the belt and tetrahedral designs, the thrust absorbed by the gaskets and by the internal friction of the solid pressure transmitting medium has been kept rather low. It amounts to only 10

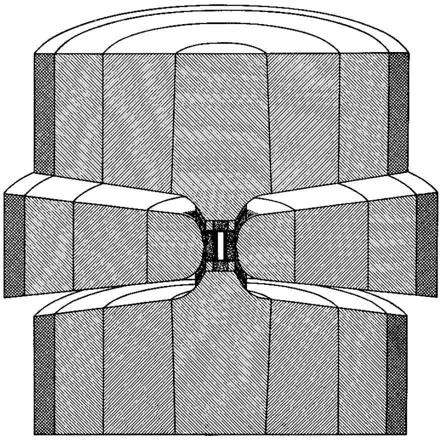

Fig. 1 The "Belt" ultrahigh-pressure, high-temperature assembly.

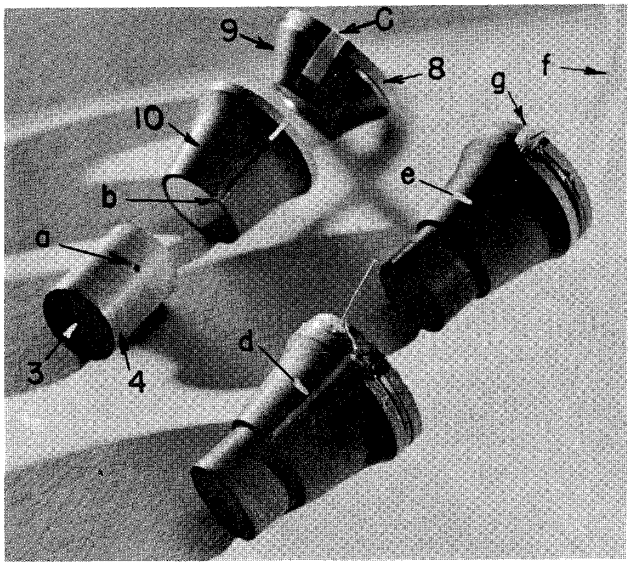

Fig. 2 Cell and gasket assembly for "Belt": (a) Hole for thermocouple in pyrophyllite cylinder 4; (b) slot for thermocouple lead in pyrophyllite gasket 10; (c) asbestos paper to insulate thermocouple lead from metal cone 9; (d) kaolin powder packed into slot b on top of thermocouple wire; (e) thermocouple slot; (f) insulating tubing over thermocouple leads; (g) daub of glue to anchor leads; (h) carbon heater-sample tube, 8 pyrophyllite gasket.

to 20 percent of the total applied thrust. These devices could probably be improved with respect to maximum pressure attainable and component lifetime by increasing the gasket-absorbed thrust to 50 percent of the total thrust. The additional thrust absorbed by the gaskets would be used to provide a more gradual pressure gradient from inner to outer edges of the gasket and to provide for additional component support.

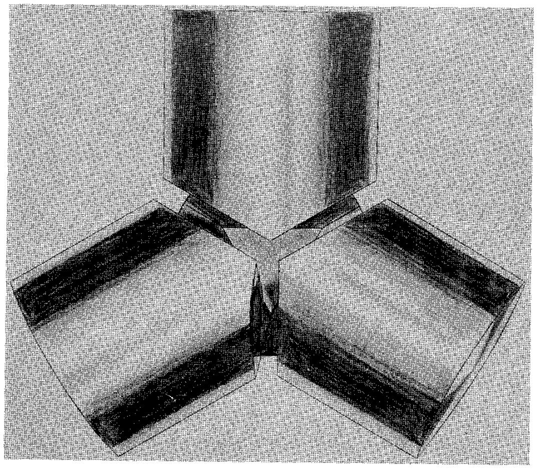

Fig. 3 Tetrahedral anvils.

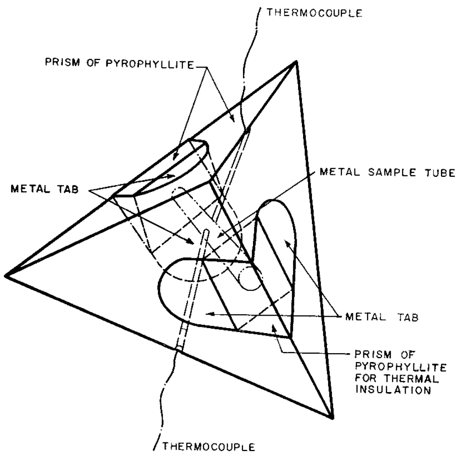

Fig. 4 Tetrahedral cell for use with tetrahedral anvils.

If yield to anvil advance were the only function to be performed by the gasket, the gasket material would, ideally, be extremely compressible and/or would flow very easily. The latter condition would be met by materials that have very low coefficients of internal friction. On the other hand, if confining the cell contents were the only function of the gasket, the gasket would ideally be composed of an extremely incompressible material that possessed great strength and a correspondingly high coefficient of internal friction.

Selection of a material to meet these opposing requirements must be made from materials of intermediate compressibility and internal friction. Universal, optimum values of compressibility and internal friction probably do not exist. Optimum values of these quantities probably depend on geometrical parameter associated with each apparatus design.

As a start towards providing information of potential usefulness in gasket design, internal frictions of several substances have been measured by use of Bridgman's shear apparatus.[5,6] Measurements at the higher pressures could not be made on materials with high coefficients of friction such as rouge (Fe_2O_3) because the force required to move the rotating anvil is so great that the carbide anvil is sheared and broken before the thin layer of rouge is sheared. Some of the data collected are displayed in Table I. Most ultrahigh-pressure gaskets in use today have coefficients of friction in the range of 0.25 to 0.50.

TABLE I

Coefficient of Friction of Some Materials at 24,200 Atmospheres

Ferric Oxide Powder	0.71	Aluminum Hydroxide Powder	0.39
Zinc Oxide Powder	.58	"Micro-Cell" Earth Powder	.37
Pumice Stone Powder	.52	Calcium Hydroxide Powder	.27
Chromic Oxide Powder	.50	"Permagel" Clay Powder	.18
Pyrophyllite Powder	.25	Boric Acid Powder	.14
Pyrophyllite Natural Block	.47	KCl Powder	.12
"Attasol" Clay Powder	.47	NaCl Powder	.12
Lead Dioxide Powder	.46	Mica Sheet	.07
Manganese Dioxide Powder	.45	Boron Nitride Powder	.07
Titanium Dioxide Powder	.45	Graphite Powder	.04
Molybdenum Trioxide Powder	.42	Molybdenum di-sulfide Powder	.04
Tin Oxide Powder	.41	Silver Chloride Powder	.03
Boron Carbide Powder	.40	Indium Sheet	.01

In order for the gasket to confine the material within the chamber, it must inherently possess great strength or else it must transmit the expulsion force imposed upon it by the chamber contents to the anvils or other apparatus components. Apparently, thin sheets of many substances subjected to heavy loads perpendicular to the sheet develop high strengths in directions parallel to the sheet. In spite of this, the gasket may be expelled or extruded by the expulsion force. This may be due to too low a coefficient of internal friction of the gasket material or to too low a coefficient of sliding friction between the gasket material and the apparatus components or both. The coefficient of sliding friction can be increased by roughening the anvils, by using a gasket material with a higher coefficient of internal friction, or by interposing a thin layer of very high friction material between the gasket and the anvil. The latter procedure is used in the tetrahedral anvil device. In this instance a thin layer of rouge is applied to the surface of the pyrophyllite tetrahedron.

In order for the gasket to successfully confine the chamber contents, it has been necessary to use the gasket material in relatively thin sections. This, of course, limits the relative motion of apparatus components and imposes limits on the chamber size for a given pressure. Additional relative motion was obtained in the belt design by using a sandwich gasket in which a thin sheet of steel was interposed between two sections of pyrophyllite. Appropriate geometrical considerations in the apparatus design, in this case, also increase the relative motion of the pressure generating components.

There may be more elaborate composition-type gaskets constructed of multiple layers of materials with widely differing strength and frictional properties that would perform the tasks required of the gasket more effectively than does the sandwich gasket currently used in the belt.

Pressure Media

An effective way to subject a substance to high-temperature simultaneously with high pressure is to enclose the substance in a thin-walled electrically heated capsule to which pressure is transmitted by a solid medium. Ideally, this solid pressure transmitting medium must meet the requirements enumerated below. It must:

1. Transmit pressure hydrostatically.
2. Have very low compressibility.
3. Have very low thermal conductivity.
4. Have very low electrical conductivity.
5. Have very high melting point and the melting point should increase with increasing pressure.
6. Be chemically inert.
7. Be thermally stable.

These requirements must be met at both ordinary and high temperatures. In some instances additional special requirements must be met as would be the case where x-ray transparency is necessary for diffraction work at high pressure and high temperature.

Of course it is impossible to find a solid pressure transmitting medium wherein all these requirements are fully and simultaneously met. Consequently, a compromise must be effected. At the present time, pyrophyllite, talc, and hexagonal boron nitride are being widely used as pressure transmitting materials in high-pressure, high-temperature work. Each of these materials has its virtues and drawbacks. None of them transmits pressure as hydrostatically as would be desirable. It seems certain that no one has, up to the present time, conducted a systematic investigation to discover materials that better meet the requirements enumerated above than do the materials now commonly used. Such research would indeed be important to the advancement of the high-pressure art.

It seems reasonable that hydrostatic solids would possess low coefficients of friction as measured in Bridgman's shear apparatus. Some semi-quantitative information on this point has been obtained,[7] but a great deal of additional work would be desirable. Cho used the apparent pressure required to obtain the 24,500 atmosphere bismuth transition as a measure of how "hydrostatic" certain media are as regards pressure transition as a measure of how "hydrostatic" certain media are as regards pressure transmission. A simple piston-and-cylinder high-pressure apparatus was used in which a bismuth wire-silver chloride rod pressure sensing element was imbedded in the solid media.[8] His data for a specific geometry are summarized in the graph of Fig. 5 for compressed powders of AgCl, BN, MoS_2, and Fe_2O_3, and for naturally occurring block pyrophyllite. Data for Fe_2O_3 and ZnO could not be obtained directly but were obtained by extrapolating measurements on mixtures of these powders with MoS_2 powder to zero concentration of MoS_2. The coefficient of friction given in Fig. 5 is that measured by Bridgman's shear apparatus at a pressure of 30,000 atmospheres. The important feature of the graph is the significant change in slope that occurs at a coefficient of friction near 0.06. If the graph had continued in the direction of the dashed line, materials such as pyrophyllite would have little value as pressure transmitting media.

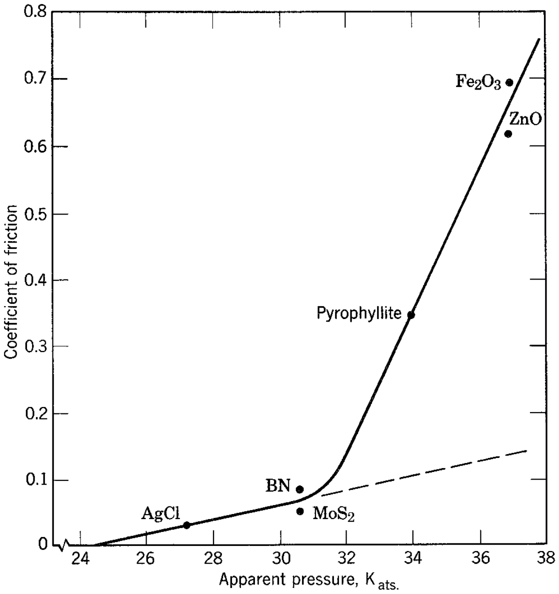

Fig. 5 Apparent pressure required for the Bi I → II transition to begin as a function of the coefficient of friction.

As already inferred, geometrical considerations are important in determining how well pressure is transmitted through solid media. Pressure cannot be transmitted into the apex of a cone of small solid angle by a material with a coefficient of friction of 0.5. Neither can pressure be transmitted effectively from the outside diameter to the inside diameter of an extremely thick-walled cylinder of high-friction material. Quantitative data on these effects are not available so studies in this area would be extremely welcome to the apparatus designer.

In addition to difficulties encountered in transmitting pressure through solid media because of high internal friction, effective transmission of pressure is sometimes prevented by high surface friction between moving apparatus components and the pressure transmitting medium. In some apparatus designs this problem is alleviated by interposing a film of low friction material between the pressure media and the apparatus components.

High Tonnage Rams

High-pressure apparatus which utilize internal furnaces for heating specimens to high temperatures must have relatively large amount of thermally insulating material surrounding the specimen in order to protect the apparatus. Ordinarily, the insulation volume if of the order of 15 times the specimen volume. Bearing this fact in mind, a simple calculation will show that, in the case of multiple anvil apparatus, hydraulic ram thrusts of the order of 1000 tons will be required to generate pressures of 100,000 atmospheres in a specimen of 1 cc volume. Significantly larger specimen volumes, such as might be desirable for commercial use, would require rams of many thousands of tons capacity.

Fortunately, the low ram stroke (under load) requirements of multiple anvil and belt-type devices simplifies the problems that are connected with large-tonnage ram design. Because the amount of motion required of the ram under load is usually less than ¼ inch, simple "O" rings with backup washers can be used as piston seals. Furthermore, if clearance between piston and cylinder is kept low, oil pressures of

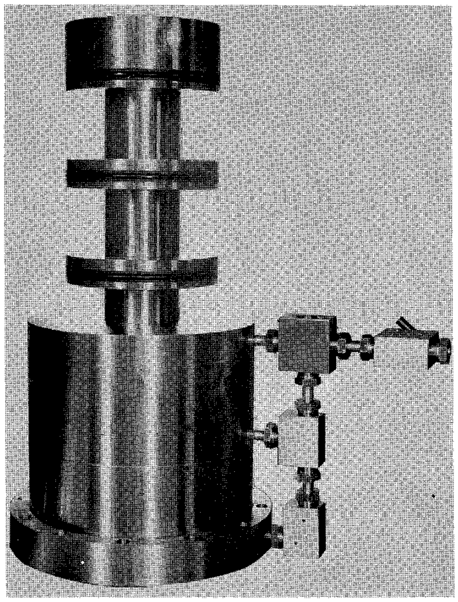

Fig. 6 Multiple-piston ram, partially assembled.

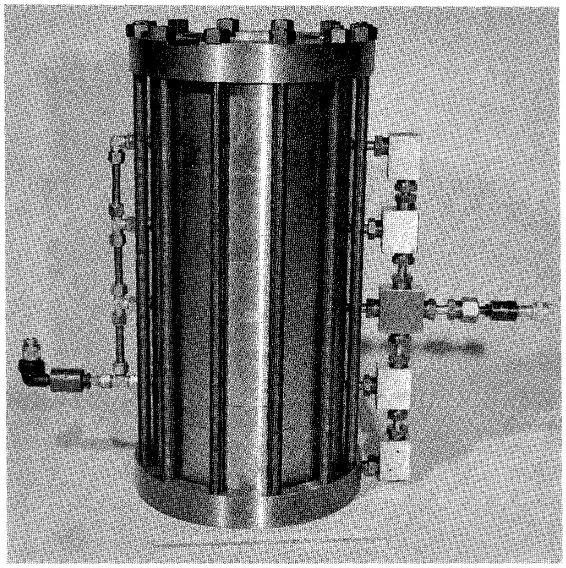

Fig. 7 Multiple-piston ram, assembled.

20,000 psi on 12-inch diameters can be used and indications are that both oil pressures and piston diameters may be increased beyond these figures without difficulty.

The cost of precision machine work required on hydraulic rams increases rapidly with increasing ram diameters. Furthermore, the number of shops with facilities capable of handling the work decreases rapidly with diameter in the larger sizes. With these facts in mind, a multiple-piston ram has been developed at Brigham Young University that may have considerable economic advantage over conventional single piston rams in the larger sizes. A photograph of the partially assembled ram is shown in Fig. 6. Three pistons are shown stacked on the central thrust column sections in the positions they will occupy in the completely assembled ram. Two pistons (not visible) are located in the completed assembly below. The pistons act in parallel. Oil pressure on each piston is the same. A short cylinder surrounds each piston. A plate with a central hole through which a section of the central column passes forms the bottom of each cylinder. The central hole is provided with an internal "O" ring seal. Oil enters each chamber through an opening in the plate. Oil entering below each of the five pistons is used to advance the ram. Oil entering chambers above the four lower pistons is used to retract the ram. The seal between the bottom of each short cylinder and the plate (with the exception of the top cylinder) is made with a thin sheet of copper. The pressure produced in the copper sheet by the ram sections above is sufficient to prevent leak. The top piston cylinder is gasketed with an "O" ring and the cylinder is kept in place by threaded rods and clamping plates. Each individual ram part is of extremely simple design thus ensuring economy of manufacture and freedom from local stress concentration in use.

The assembled ram is shown in Fig. 7. The total number of pistons in this ram is five. The inside and outside diameters of each cylinder are 8 and 12 inches, respectively. The effective piston area is the same as that of a single piston ram of 16-inch piston diameter. The over-all length of the ram is 27 inches. Total ram stroke is 2 inches. The design capacity of the ram is 3000 tons at 30,000 psi oil pressure. At the present time the ram has only been operated at a maximum load of 1750 tons. The cost of this ram is estimated to be less than one-third the cost of a single piston ram of the same capacity. (Note that a single piston ram, operating at the same oil pressure and utilizing the same quality steel, would be 24 inches outside diameter with a 4-inch-thick steel cylinder wall.)

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[1] H.T. Hall, Rev. Sci. Instr., 31, 125-131 (1960). This article first appeared as General Electric Research Laboratory Report RL-1064, March, 1954. However, distribution of this report was rigidly limited.

[2] H.T. Hall, Rev. Sci. Instr., 29, 267-275 (1958).

[3] U.S. Patent No. 2,918,699.

[4] P.W. Bridgman, Proc. Roy. Soc. (London), A203, 1 (1950).

[5] P.W. Bridgman, Phys. Rev. 48, 825-828 (1935).

[6] G.R. Hyde, "Friction At Very High Pressure," M.S. Thesis, Brigham Young Univ. (1957).

[7] Yanglai Cho, "Propagation of High Pressure in the Solid State, " M.A. Thesis, Brigham Young Univ. (1958).

[8] Ref. (1), pp. 268-9.

DISCUSSIONS

by W.G. Field

Operation of a 600-ton version of the tetrahedral-anvil press presented difficulties in preventing breakage of thermocouple leads using the method previously described by Hall, that is to bring the leads out between anvils utilizing pyrophyllite tabs to hold the thermocouple leads in position. This difficulty may be due to greater movement of leads in this size press.

A method has been developed which allows the thermocouples to be installed simply and permit reliable operation over the complete pressure and temperature range of the equipment. This is accomplished in the following manner. Heating current is applied in the usual manner except that half-tabs are utilized and the current is, therefore, applied through only two heads. The tetrahedron has two additional slots cut at the exist point (as in Fig. 4) of the thermocouple leads. The leads are bent to lie on the bottom of this slot, covered with a pyrophyllite prism in the usual manner, and again bent to lie on the other two sides of the tetrahedron. No contact tabs are required. Contact is, therefore, through the heads to a contact ring and hence to the appropriate thermocouple lead material.

This technique introduces additional thermocouples. However, if the two heads are at the same temperature the e.m.f.'s due to these additional couples cancel. Examination of the tetrahedron geometry shows the two heads utilized for the thermocouple contacts are symmetrically located with respect to the heater and should, therefore, by at the same temperature, the anvils are also good heat sinks which assists in maintaining equal temperature.

This method has been utilized on some 30 experimental runs without a single failure over the entire pressure range. Tentative measurements comparing this method with another thermocouple utilizing Hall's technique showed temperature deviations of a few percent. The extreme simplicity, however, makes the method very desirable. The thermocouple reliability has also made it possible to utilize Servo techniques for controlling the temperature to ± ½°C as indicated by the thermocouple e.m.f.

by R.H. Wentorf

Wentorf: What was the diameter of the anvils used in the friction and shear measurements?

Hall: 0.375 inch.

Wentorf: Have measurements been made using anvils of several different sizes in view of the possible complexity of the pressure distribution on the working faces?

Hall: No.

by Alexander Zeitlin

As Dr. Hall's paper indicates, ultra-high pressure equipment, originally available only in sizes suited for basic research is now available in larger sizes suited for applied research.

A 2000/8000 ton tetrahedral unit can develop pressures of 100 kilobars (1,500,000 psi) in pyrophyllite tetrahedron with an edge length of 2 7/8 inches (total volume 2.80 cubic inches). Sizes of cylindrical specimens that can be used within such tetrahedron depend on compressibility of specimens and operating temperatures. For easily compressible specimens and high temperatures (above 2500°F) 10 percent utilization seems to be indicated. The dimensions of a corresponding cylindrical specimen would then be ½ inch diam. x 1 7/16 inches long. For a 50 percent utilization (specimens of small compressibility, temperatures below 2500°F) the dimensions of a cylindrical specimen will be 1 1/16 inches long.

For lower pressures, the dimensions would increase; for instance, for 50 kilobars (750,000 psi), the tetrahedron's edge would be 4 inches long (volume 7.5 cubic inches); a cylindrical specimen for 10 percent utilization would be 5/8 inch diam. x 2 ½ inches long; for a 50 percent utilization the dimensions would be 1 ½ inches diam. and 1 1/8 inches long.

These specimen dimensions would seem to open a wide range of new research potentialities.