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Equipment for Ultra-High-Pressure Work

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The paper reports on general purpose equipment for ultra-high-pressure work for basic and applied research as well as for production applications. The author expresses the hope that the opportunities offered to research workers in the field of ultra-high pressure by the new types of equipment and greatly increased sizes of test specimens will lead soon to many new industrial applications of the ultra-high-pressure technique in the field of chemistry, metallurgy, and related areas.

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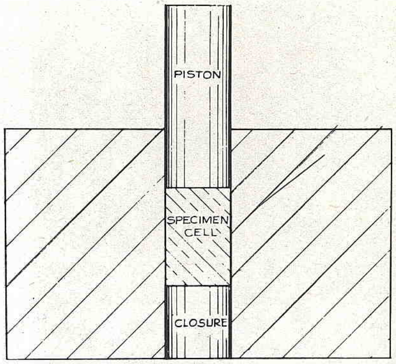


Fig. 1 Simple piston - cylinder system

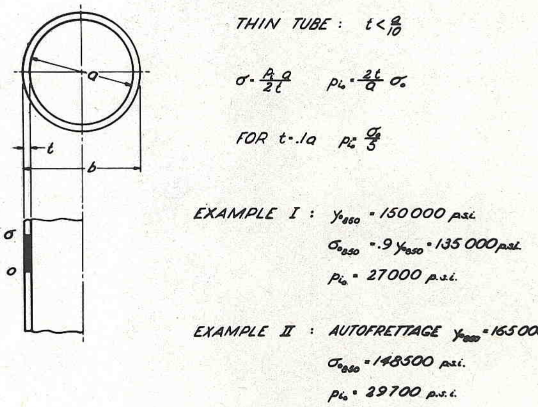


Fig. 2 Stress conditions in a thin tube

The year 1909 was when the scientists began to formulate their ideas about the structure of atoms. That was also the year when an inquisitive young scientist in Cambridge, Mass., decided to find out what happens to matter when it is subjected to extremely high pressure.

Fifty years later, having written over 300 papers on the effects of ultra high pressure on matter as well as two books considered throughout the world as "high pressure bibles," having been honored by a number of scientific institutions and having received many awards and prizes (among others, the 1946 Nobel Prize for Physics) Percy W. Bridgman retired from his laboratory in 1959.

A review of the ultra-high-pressure field at the time of his retirement would show to his satisfaction a steadily growing interest in ultra-high-pressure research. Since then further progress has been made in the development of the equipment for and techniques of ultra-high-pressure work.

In reporting on the present state of art I will confine myself to the general purpose equipment for basic and applied research as well as for production operations. Time limitations force me to omit reporting on specialized equipment for x-ray and optical work and for some other special purposes.

GENERATING HIGH PRESSURES

The most obvious method of generating high pressures is shown in Fig. 1: The hydraulically operated piston advances within the cylinder and compressing the substance placed within the cylinder generates substantial pressures within it. The maximum pressures obtainable in a cavity of this type depend on the strength of the materials used and the stresses developed in the cylinder and piston.

Fig. 2 shows that the pressures generated within a thin-wall cylinder are limited to a fraction of the permissible stress in the cylinder wall. Referring to operations at room temperature and assuming that the thin-wall cylinder is made of steel with some ductility, the range of pressures obtainable in such a device would be well below 50,000 psi.

One would assume that making the wall of the cylinder thicker and thicker would result in an unlimited increase of pressures that can be contained within the cavity. Unfortunately, this is not the case. As shown in Fig. 3, only the inner layer of a thick-wall cylinder close to the ID is fully utilized from the viewpoint of stress. The farther away we move from the cylinder axis, the less is the utilization of the material. It is therefore useless to make the OD of a thick-wall cylinder bigger than three or four times the ID.

A thick-wall cylinder of these dimensions allows the generation of internal pressures in the range of approximately 45 per cent of the permissible stress in the cylinder wall. Even an infinitely thick cylinder will not allow pressures in excess of 0.58 of the permissible stress. Consequently, a thick-wall cylinder made of steel and operating in the elastic range could not be used for pressures beyond 80,000 to 100,000 psi.

It must be pointed out that the quoted yield strength applies to "reasonable size" workpieces

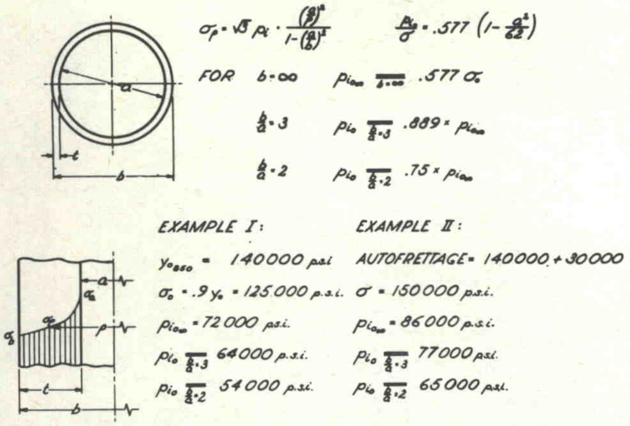


Fig. 3 Stress conditions in a thick tube (elastic range)

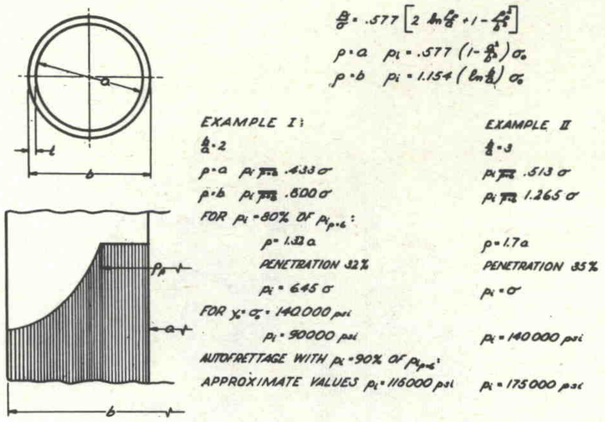


Fig. 4 Stress conditions in a partially plastic thick tube

with a cavity diameter of 5 to 6 in. Larger workpieces show a substantially lower yield point and consequently the pressures stated above cannot be generated in thick-wall cylinders of more than 5 to 6 in. ID.

Proper arrangements of the cylinder and rams would allow exceeding the yield point in certain limited areas of the cylinder as shown in Fig. 4. Partially plastic thick-wall cylinder can be made to store more strain energy than a thick-wall cylinder in a purely elastic state. Allowing a plastic zone of approximately 50 per cent of the total wall thickness results in the possibility of containing pressures up to 125,000 psi. Proper care must be taken in this design to limit the length of the test cavity to a fraction of the total length of the thick-wall cylinder in order to eliminate the effect of the ends.

Compound Cylinders

The next step in the development of arrangements for higher and higher pressures consists of the use of compound cylinders. Fig. 5 shows the stress distribution in a two-layer compound cylinder with a shrink fit of reasonable interference between the two tubes. A review of the stresses in the compound cylinder shows immediately that the pressures which could be generated in such a cylinder are substantially higher than the pressures which can be generated in a simple thick-wall tube. The increase is equivalent to the compressive stresses generated at the ID of the cylinder when at rest.

A compound cylinder operating in the elastic range allows the containment of pressures in the range of 150,000 to 175,000 psi.

If partially plastic conditions are permissible, then the pressures can be increased to a range of 200,000 to 225,000 psi, Fig. 6.

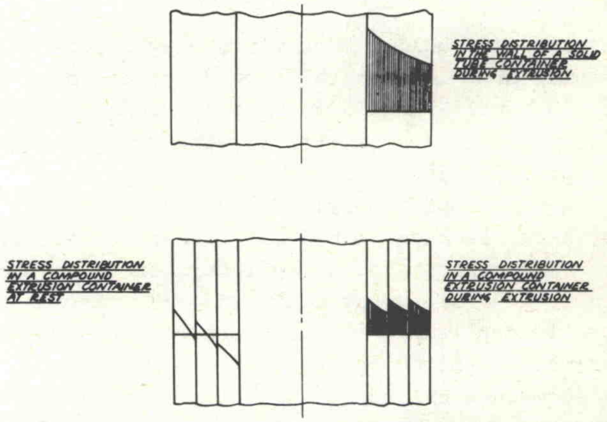


Fig. 5 Extrusion container stress distribution

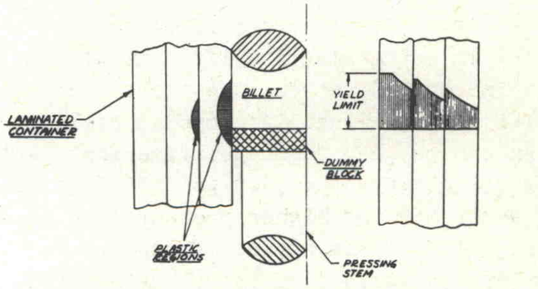


Fig. 4 Plastic deformation of container when stressed beyond elastic limit

Use of Materials Other Than Alloy Steel

Until now, we have been reviewing cylinders made of alloy steel. Configurations of this type can be made with cavity diameters of 4 to 5 in. and even larger. The introduction of stronger materials, in particular tungsten carbide, allows a further increase of generated pressures. However, the fabrication of large tungsten-carbide

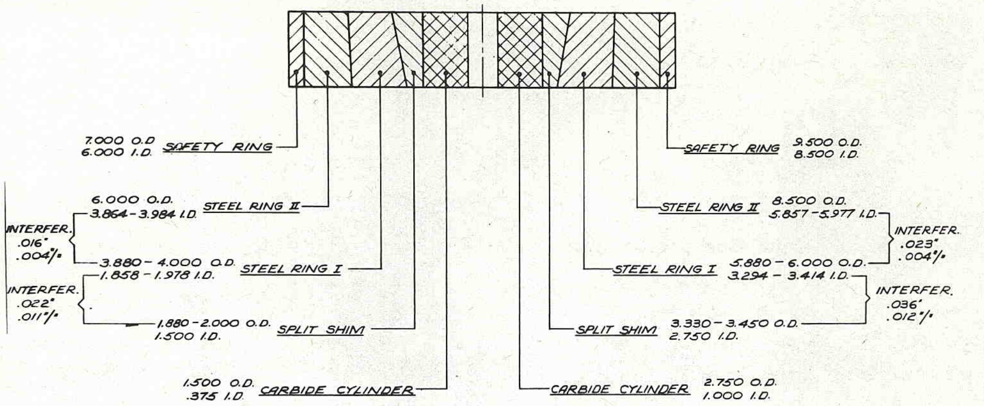


Fig. 7 Compound cylinder - H. T. Hall

rings is difficult. Such rings show a substantial drop in physical properties as compared with the small rings and therefore tungsten-carbide cylinders are limited to sizes in the range of 1 to 2 in. ID.

A simple piston-cylinder system shown in Fig. 1 can be made with a tungsten-carbide cylinder. The piston and the enclosure are also preferably made of tungsten carbide. The device as shown is applicable to the pressure range of approximately 250,000 psi.

Compound cylinders utilizing tungsten carbide expand the range very substantially. Fig. 7 shows a compound cylinder consisting of three basic rings; the inner tungsten-carbide ring and two steel rings. A shrink fit is obtained by tapering the ID and OD of steel ring 1 and the ID of steel ring 2. In order to reduce the difficulties in assembling a split shim with a tapered OD is introduced between the carbide cylinder and the first steel ring. A compound cylinder of this design can be used for repetitive application of pressures of up to 500,000 psi (35 kilobars).

Generation of higher pressures is possible but the life of the device is rapidly reduced to a few cycles at 750,000 psi (50 kilobars).

Two improved arrangements are shown in Fig. 8. The arrangement shown on top of the figure indicates an axial restraint the objective of which is to counteract the axial tension stresses generated in the carbide cylinders under the influence of the Poisson effect. The auxiliary pressure 2 is proportional with the ram pressure 1 so that the tension stresses due to the application of pressure 1 are counteracted by an appropriate level of pressure 2.

The corelation between pressure 1 and pressure 2 can be properly controlled on the way up. However, it is difficult to maintain the proper relation between pressure 1 and pressure 2 during the pressure release.

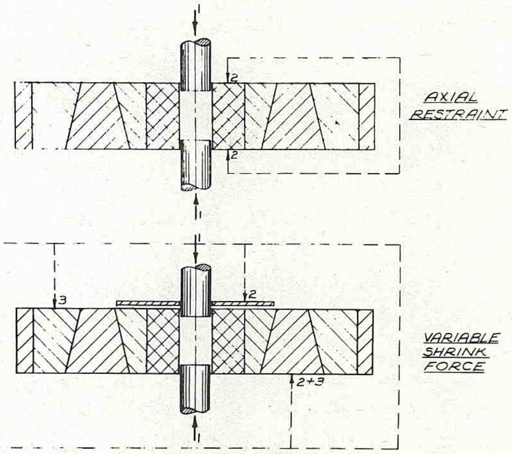


Fig. 8 Axially supported compound cylinders

At the bottom of Fig. 8, an arrangement is shown which increases the shrink force between the carbide and the two steel cylinders in proportion with the increase in ram pressure. Pressures 2 and 3 are applied to rings moving downward while the combined pressure 2 plus 3 is applied to the intermediate ring moving it upwards thus increasing the shrink fit when pressure 1 is rising. Again, this arrangement works rather satisfactorily on the way up but fails invariably on the way down.

Until now, we have been discussing the cylinder. Turning to the rams we find that straight rams as shown in previous illustrations cannot be operated satisfactorily at pressures beyond 500,000 psi. At the most 750,000 psi is obtainable with carbide rams but the useful life is reduced to a single cycle.

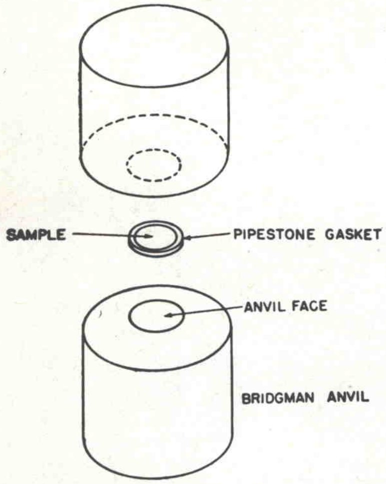


Fig. 9 Bridgman anvils

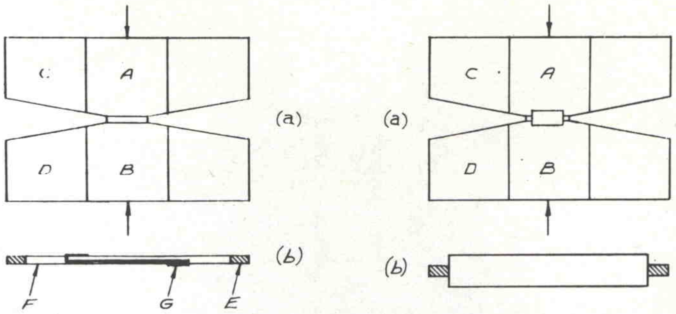


Fig. 10 Anvil apparatus and massive support principle

THE BRIDGMAN APPROACH

Realizing the shortcomings of all devices described previously, Bridgman suggested a somewhat different approach to the problem of high pressure equipment.

Fig. 9 shows the shape of the Bridgman anvil. The two rams or anvils have rather small faces from which the material spreads sideways rather fast so that the area of high stresses generated in the vicinity of the sample is limited to a small portion of the anvil. The sample itself is a wafer surrounded by a restraining gasket made of solid material which becomes viscous under pressure.

Fig. 10 shows the anvils designed as compound configurations with the shrink rings C and D increasing the range of pressures which can be developed between the anvils A and B. Very high pressures have been generated with the help of this device. The calibration of the device and the measurement of generated pressures is a subject of rather animated controversy between various research workers at present. There is a general agreement that pressures up to 250 kilobars (3,750,000 psi) can be generated in this device, while claims of individual research workers of having reached pressures of 500 and even more kilobars (7,500,000 psi) await confirmation by others.

Specimens processed between Bridgman anvils are rather small; the thickness is usually in the neighborhood of 0.010 in. The diameter of the wafer usually does not exceed 1/8 in. These small dimensions of the specimens prevent the use of the device for applied engineering investigation. However, certain basic investigations into the structure of matter are possible.

Bridgman's anvil device shows two features which became of extreme importance for the further development of high-pressure apparatus: One is the massive-support principle; the other is the use of pressure-transmitting material which becomes viscous under pressure.

Massive-Support and Plasticized Gasket Principles

The massive-support principle reduces the area of high stresses because it provides for rapid diffusion of stresses into larger cross sections which exercises a restraining force on the anvil surface.

The gasket which becomes viscous under the influence of the pressure is reduced in thickness simultaneously with the compression of the specimen. The excess material flows outward, thus leaving the area of high pressures and solidifying outside the anvil contact area thus forming a solid restraining ring around the anvil tips. The restraining force exercised by the extruded gasket assists in counteracting the bursting forces which are generated tangentially to the anvil surface.

Bridgman has tested a large number of materials and has indicated a number of substances which can be used as plasticized gaskets. The most popular materials are pyrophyllite (common lava) which is basically an aluminum silicate, pipestone, talc, sodium chloride, silver chloride, and many more.

Actually, even metals can be used as gasket material for pressures well above their yield point.

Principles Applied to Piston-Cylinder Systems

The two principles - massive support and the plasticized gasket - are applicable to piston-cylinder systems as exemplified in Fig. 11. The tip of the low-pressure piston provides the area of massive support while the support collar

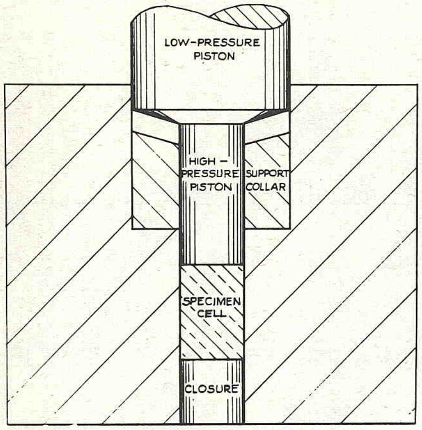
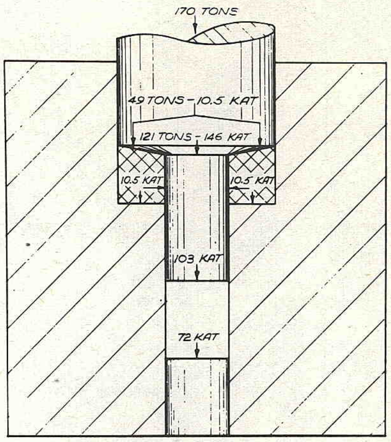


Fig. 11 Supported piston system



NUMERICAL DATA BY MANUFACTURING LAB. INC.

Fig. 13 Stepped piston concept

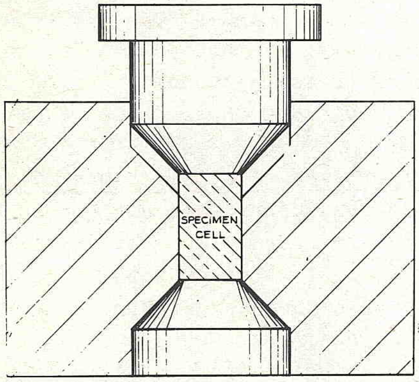


Fig. 12 Massive support applied to a single piston-cylinder system

acting as the plasticized gasket provides radially arranged restraining force around the upper portion of the high-pressure piston. If the high-pressure piston is properly lapped in within the cylindrical bore of the main block, a cylindrical support for the tip is provided by the main block proper. This arrangement improves the working conditions of the piston so that either the piston life can be extended when operating at pressures around 35 kilobars (500,000 psi) or the pressure range can be extended upwards (at the price of short piston life, to be sure).

Fig. 12 indicates the application of the massive-support principle to the lower closure. The space between the upper piston and the main cylinder block can be filled out with an appropriate gasket material.

Fig. 13 shows the pressure distribution in a stepped-piston arrangement similar to the two previous figures. The numerical data indicating the pressure distributions were published by Manufacturing Laboratories, Inc. The pressure data on the slide are in kilo atmospheres which, for the purposes of this presentation, can be assumed to be roughly equal to kilobars. Converted into pounds per square inch we obtain the pressure on the large cylinder to be approximately 160,000 psi. This pressure transmitted to the plasticized gasket generates a radial restraining force on the surface of the small cylinder in the same amount of 160,000 psi. The pressure of approximately 2,000,000 psi generated at the juncture between the large and the small piston drops rapidly due to friction so that at the upper surface of the specimen the pressure is only 1,500,000 psi while at the lower surface of the specimen the pressure drops even farther to approximately 1,100,000 psi.

As stated before, the simple piston-cylinder systems do not have a particularly satisfactory life. The life is still further reduced when the test conditions specify high temperatures within the specimen.

Modification of Piston-Cylinder System

In order to obtain better performance, Bridgman and other research workers have suggested two-stage arrangements. The main disadvantages of two-stage arrangements are their intricacy and a further reduction in size of the already small specimen. Therefore, substantial efforts have been exerted by a number of workers to improve the performance of piston-cylinder systems by modifying the configuration.

Fig. 13(a) shows two such modifications: The modification applied to the upper piston uses a tapered pyrophyllite gasket surrounding the plunger tip. As the plunger moves down, more pressure is exerted on the gasket; in this way, an increased lateral support is obtained. The

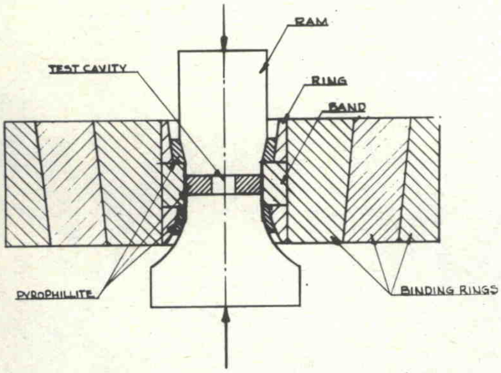


Fig. 13(a) Lateral support principle

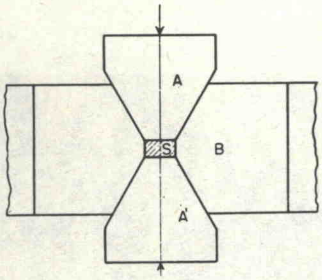


Fig. 13(b) The "Girdle" - H. T. Hall

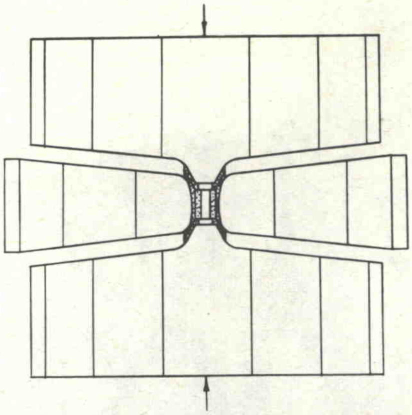


Fig. 14 Belt concept - General Electric Company

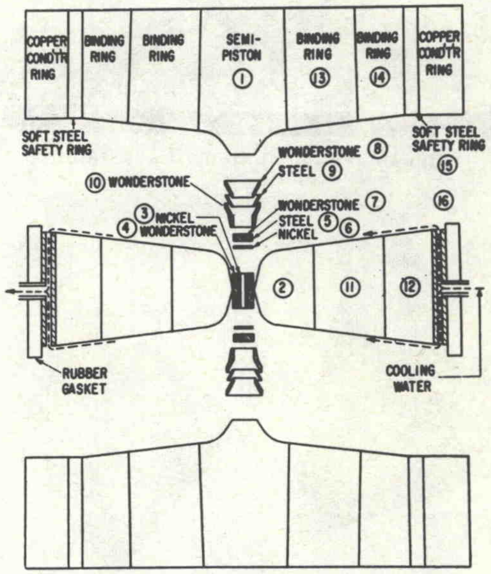


Fig. 15 The "belt" "exploded" assembly - General Electric Company

design of the lower piston goes a step further by flaring out the piston surface and thus increasing the massive-support effect. The General Electric Research Laboratories were particularly successful in this respect. The objective of the designs of the General Electric Research Laboratories is to apply the principle of massive support not only to the ram but also to the cylinder of the piston-cylinder system. The first approach to the problem is shown in Fig. 13(b). The nickname given to this apparatus is, for obvious reasons, "the girdle." The ultimate in this development is the "belt" apparatus, the concept of which is shown in Fig. 14.

The difference between the conceptual presentation and the actual execution is rather substantial as demonstrated by comparing Fig. 14 with Fig. 15.

Even the sophisticated "girdle" and "belt" designs do not overcome the scaling-up problems so that the specimens processed in configurations of these types are confined to fractions of an inch.

RAM SYSTEM OF H. T. HALL

Having come to the conclusion that it would be hopeless to attempt to scale up piston-cylinder systems, H. T. Hall has suggested a complete elimination of the confining cylinder and the use of a tetrahedrally arranged ram system.

Fig. 16 shows the original machine of Tracy Hall. The four massive support anvils are located at the ends of the four rams actuated by the cylinders seen on the slide.

Fig. 17 shows the cavity assembly in which the specimen is arranged within a solid piece of the gasket material.

The original machine of H. T. Hall was rated at 200 tons per cylinder. In other words, the pressure exerted by each ram on each surface of the tetrahedral cavity assembly was 400,000 psi so that in order to generate 1,500,000 psi (100

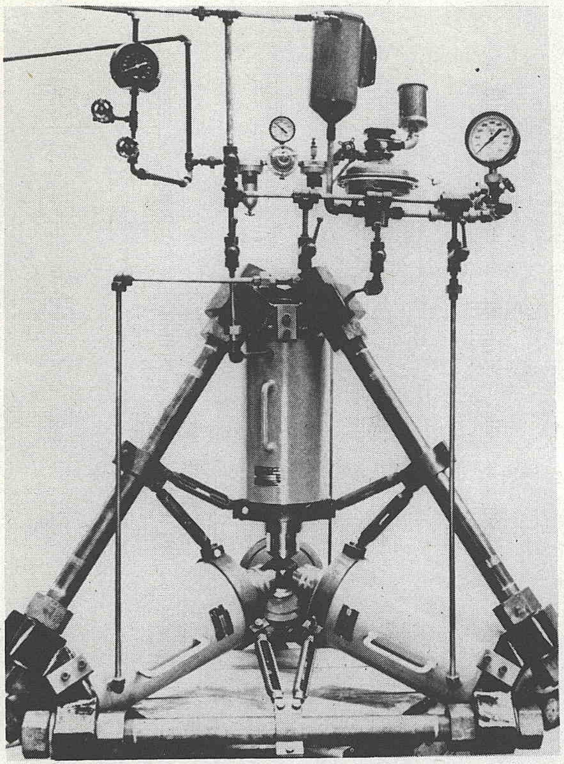


Fig. 16 Tetrahedral anvil apparatus

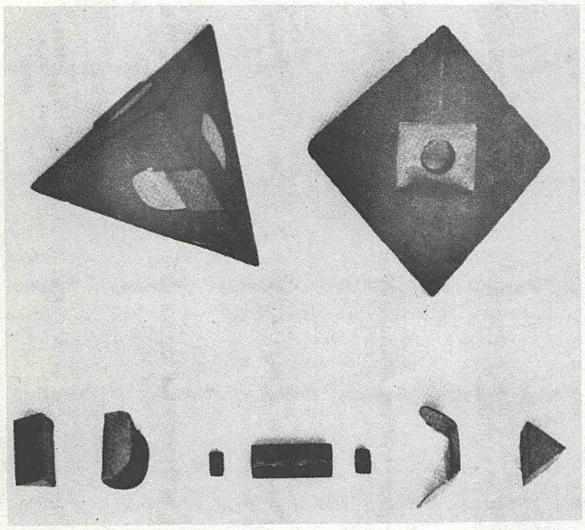


Fig. 17 Cavity assembly

kilobars) within the test cavity, the area of each face of the tetrahedron was limited to 0.33 in.

Preliminary design studies directed towards scaling up of the unit showed the necessity of changing the structural concept, although the principle of the multi-ram arrangement was, of course, maintained.

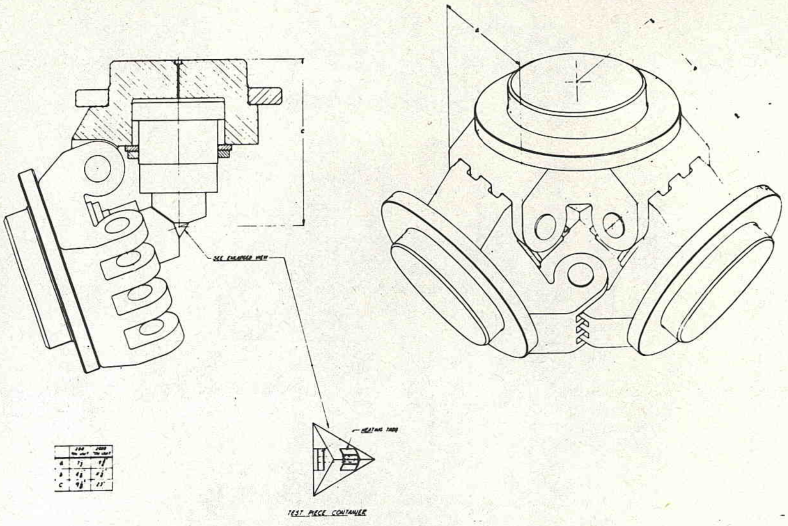


Fig. 18 Hinged tetrahedral anvil apparatus

Fig. 19 Assembly of a 2000/8000 ton tetrahedral hinge unit

Ram of 2000 Ton Capacity

The conceptual sketch of the redesigned unit is shown in Fig. 18. This type of a structure can be scaled up. Units of 2000 ton ram capacity have been built and are operated successfully.

Fig. 19 shows the assembly of the four heads of a 2000-ton unit while Fig. 20 shows the pressure control panel. Fig. 20(a) shows the rather simple hydraulic system arranged in the back of the panel. The magnification obtainable in the hinge apparatus is usually kept between 1 to 50 and 1 to 100 so that, for instance, with a magnification factor of 1 to 100 a primary pressure of

Fig. 20 Hinge unit control panel

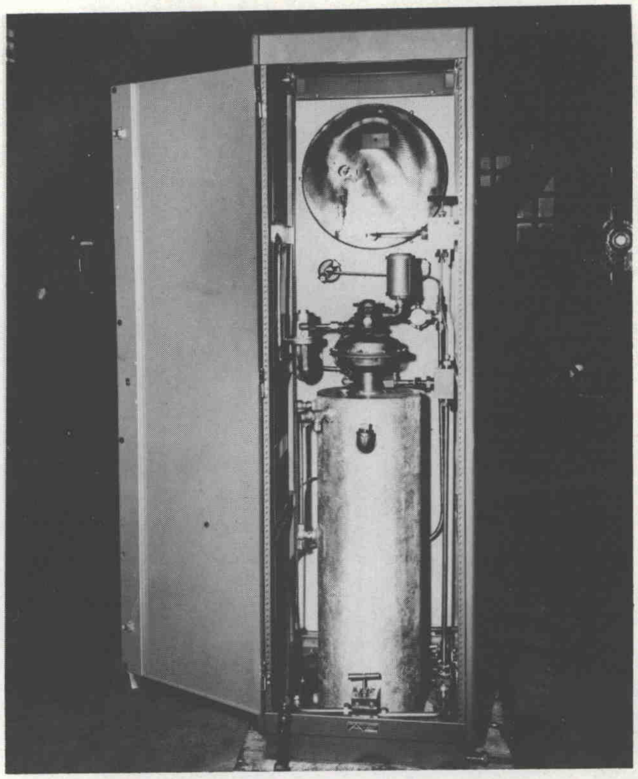


Fig. 20(a) Hinge unit hydraulic system

15,000 psi can be used to generate 1,500,000 psi (100 kilobars) in the test assembly. The extrusion of the gasket material occurs in the areas between the anvil tips so that each anvil tip is surrounded by the extruded gasket and a restraining force is applied to the anvil tip in a very efficient manner.

For a long time the tetrahedral arrangement was considered unique. However, as Fig. 20(b) shows, substantial gain in volume can be obtained by substituting a cubic arrangement for Hall's tetrahedral concept. Therefore, a considerable amount of effort was applied to the development of a cubic unit.

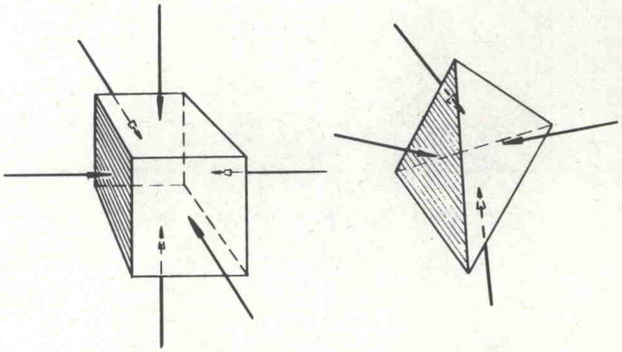


Fig. 20(b) Ram action of polyhedrals

Hinged and Articulated Cubic Frame With Six Rams

Fig. 21 shows the concept of a hinged and articulated cubic frame with six rams converging on the test specimen. As stated before, the advantage of the cubic arrangement consists of an increased volume of the cavity assembly. Of course, the cubic arrangement requires 50 per cent more rams (six instead of four). However, the volume of a cubic assembly is about 2½ times the volume of the equivalent tetrahedral piece so that the utilization of the unit is substantially improved.

Prismatic Specimens

The desire to obtain prismatic (elongated) specimens is taken into account in the arrangement shown in Fig. 22. As can be seen in the figure, the basic arrangement consists of a triangular arrangement (rectangular arrangement is also possible). Utilizing an articulated hinge frame each side of the articulated frame contains one or a multiplicity of cylinders which operated simultaneously transmit the pressing effort through appropriately shaped anvils onto the test assembly. Although this unit looked quite different from all previous ones, it still incorporates the four basic concepts; namely, the massive support in the anvils, the plasticized gasket, the spatial arrangement of rams, and the articulated hinge frame.

The two ends of the basically prismatic ar-

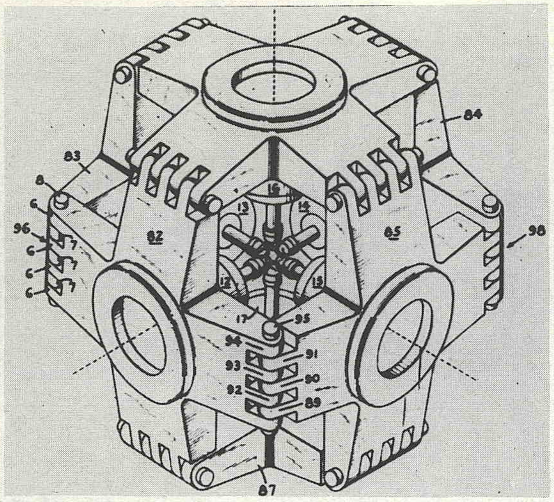


Fig. 21 Hinged cubic apparatus

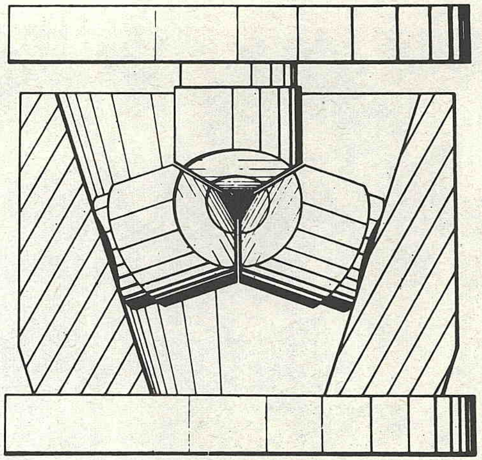


Fig. 23 "NBS" unit, anvil arrangement

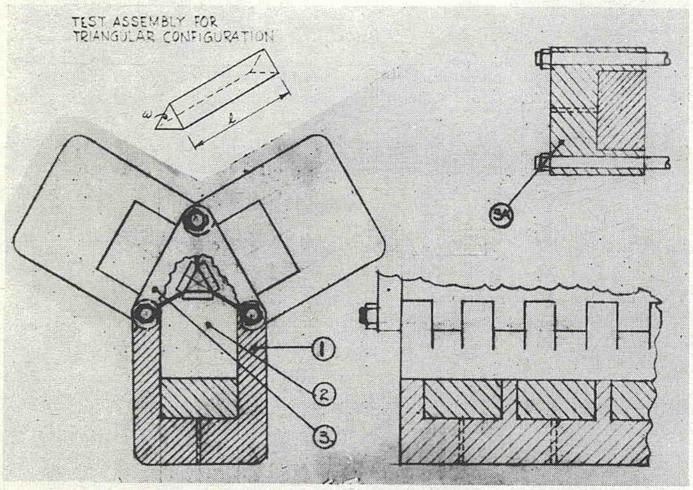


Fig. 22 Delta high pressure apparatus

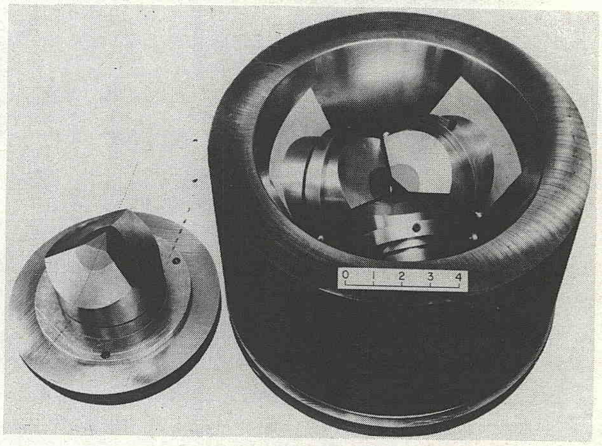


Fig. 24 "NBS" unit - assembled with top anvil removed

rangement are closed by auxiliary cylinders or by simple support plates.

National Bureau of Standards Unit

The desire to have available an inexpensive unit approximating the performance of more intricate spatial ram arrangements led the National Bureau of Standards to suggest a unit as shown in Fig. 23.

In this basically tetrahedral unit the three lower rams are arranged slidingly within a tapered retainer sleeve. The fourth anvil at the top actuated by an outside force energizes the system. This unit performs quite satisfactorily as long as the ratings are low (100 to 200 ton total pressing force on each ram). In larger sizes certain operational problems arise: (1) The friction of the lower three rams against the

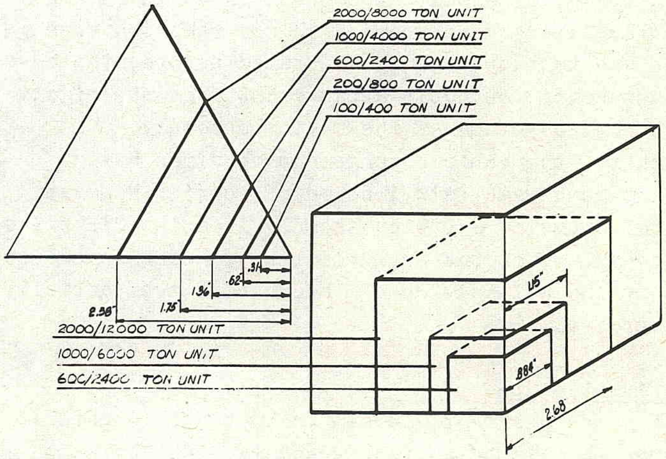


Fig. 24(a) Relative size of tetrahedral and cubic units

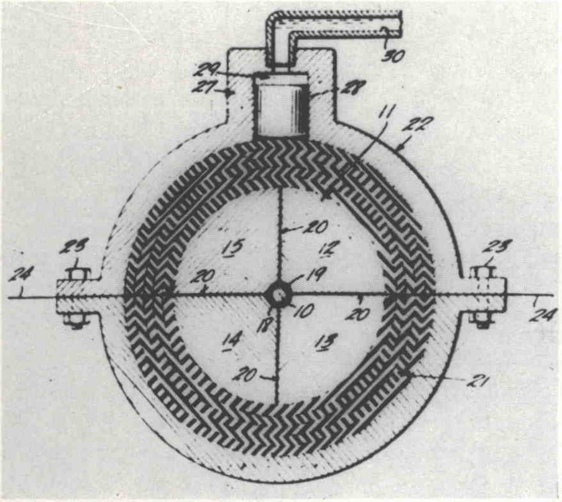


Fig. 25 Mt. Vernon super high pressure - unit I

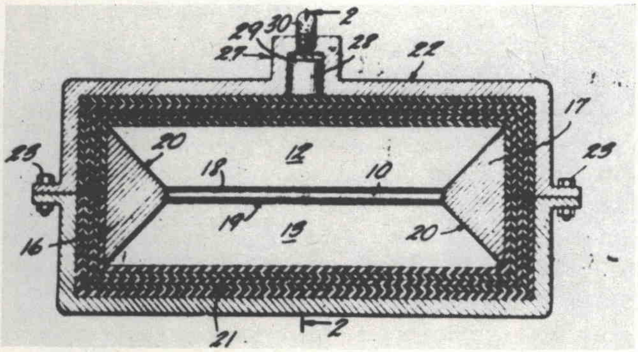


Fig. 26 Mt. Vernon super high pressure - unit II

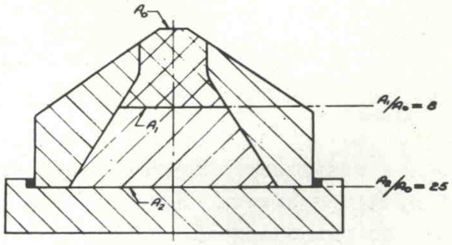


Fig. 27 Tooling arrangement

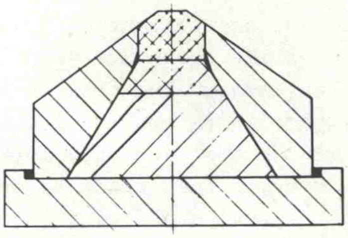


Fig. 28 Tooling arrangement

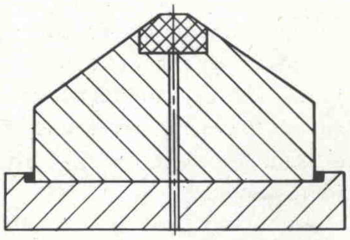


Fig. 29 Tooling arrangement

retainer sleeve is transmitted to the contact area between anvils and the test assembly thus distorting the pressure distribution and influencing unfavorably the life of the anvils. (2) The lower three anvils have at the best a line contact with the retainer sleeve except in the lowest position which they never reach in actual operation. This line contact causes excessive wear. In addition, the concentrated reactions of the lower anvils against the retainer sleeve distort the sleeve.

For all that, units of 100 to 200 ton rating perform satisfactorily. Fig. 24 shows a photograph of such unit.

Tetrahedral and Cubic Units

I would like to come back to the discussion of the tetrahedral and cubic units with the articulated hinge frame. Units of this design can be scaled up to substantial dimensions. Ram raings of 2000 tons have been built and 8000-ton ram ratings have been designed. A tetrahedral unit with four 2000-ton rams permits the generation of pressures of 100 kilobars (1,500,000 psi) in a pyrophyllite tetrahedron with 2⅞ in. edge. The volume of such tetrahedral is 2.8 cu in. A corresponding 2000-ton cubic configuration allows the use of a test assembly cube with an edge 1⅞ in. long and with a volume of 6.55 cu in. At 50 kilobars (750,000 psi) the same 2000-ton cubic assembly allows the use of a cube with a 2⅝-in. edge and a volume of 18 cu in. Fig. 24(a) illustrates the dependence of the size of the test assembly on the rating of the unit.

Of course, one must realize that the total volume enclosed between the anvil tips cannot be occupied by the test specimen. A substantial portion of the volume must be occupied by the pressure-transmitting plasticized medium (pyrophyllite or the like). Still, specimens of 1¼ in. diam and 2½ in. long can be processed in the 2000-ton cubic unit. A prismatic configuration with a 2000-ton rating would allow the generation of 100 kilobars in a specimen more than 5 in. long.

The 8000-ton units would allow, of course, the use of still larger specimens.

Multi-Ram Arrangement

The multi-ram arrangement in an articulated hinge frame has resulted in a very substantial increase of specimen sizes. However, the frames are rigid and do not allow the changeover from a tetrahedral to a cubic or to a prismatic configuration. The desire to obtain greater versatility resulted in the development of a different structure allowing for the interchangeable use of tetrahedral, cubic, and prismatic arrangements.

Fig. 25 shows the structure when using a cubic anvil arrangement. The cross section shows four of the six anvils (items 12, 13, 14, and 15) abutting against the test assembly 10. The anvils are so arranged that space 20 remains between two neighbor anvils allowing their advance when the plasticized gasket extrudes from the test assembly 10 into the space 20. The entire anvil assembly is contained within a resilient shell 21 which, in turn, is located within a pressure vessel 22. A small amount of pressurized liquid introduced into the spherical space between items 21 and 22 exerts pressure upon the resilient shell 21 which, in turn, transmits the pressure to the anvils thus generating the compressive force then applied by the anvils upon the test specimen assembly 10.

It can be seen that the tetrahedral-anvil arrangement can be easily substituted for the cubic arrangement shown and that the same shell can accommodate a prismatic arrangement. Fig. 26 shows a shell specifically designed for a prismatic arrangement.

Apart from the simplification in the structure itself this so-called hydrostatic arrangement promises fewer problems when scaling it up beyond the present limitations of the articulated-hinge design.

The hydrostatic concept is being incorporated in a number of units at present. Upon completion of the development work a further simplification of this concept can be expected.

Progress in Anvil Design

The following illustration indicates progress in the anvil design for multi-ram configurations. Fig. 27 shows an anvil with a very substantial tungsten-carbide tip placed within a confining steel ring and supported by a tapered steel base.

Fig. 28 shows a somewhat less expensive design in which the tungsten-carbide tip has been split into two layers of which only one is subjected to frequent replacement while the lower portion enjoys a very long life.

Finally, Fig. 29 shows a simplified design, with a comparatively small tungsten-carbide tip, which has proven itself as being quite satisfactory for pressures up to 100 kilobars (1,500,000 psi). As a matter of fact, the current experience indicates that anvils made exclusively of steel without the use of tungsten carbide perform satisfactorily for pressures up to 70 kilobars (1,000,000 psi).

High Temperatures

The majority of the modern high-pressure arrangements can be used for simultaneous generation of high temperatures. Two methods of heating are available; either a thermit charge surrounding the specimen or heating by means of electrical resistances. In this latter case, the heat can be generated by passing the current through the specimen. This can be accomplished, of course, only if the specimen is conducting and on the other hand, the resistance of the specimen is not exorbitantly low. Specimens with a resistance of 0.001 ohm would allow the generation of 1 kw by the application of 1 volt across the specimen.

If the specimen is nonconducting or if the resistance of the specimen is much lower than indicated in the foregoing example, then the best method is to generate heat in a separate resistance element placed near the specimen, whereupon the heat is conveyed to the specimen indirectly.

In both cases (direct and indirect resistance heating) plungers or anvils can be used to introduce the current into the test assembly.

It is to be hoped that the opportunities offered to research workers in the field of ultra high pressure by the new types of equipment and greatly increased sizes of test specimens will lead soon to many new industrial applications of the ultra-high-pressure technique in the field of chemistry, metallurgy, and related areas.