Apparatus for Optical Studies at High Pressure

By THOS. C. POULTER
Department of Physics, Iowa Wesleyan College
(Received March 18, 1932)

The selection of materials for the construction of high pressure equipment is discussed. Simplified piston packing, electrode insulation, temperature control and a compact and safe type construction of pressure equipment is described with particular reference to systems for optical studies at high pressures. Equipment for optical studies at pressures as high as 30,000 atm. and some of their limitations and peculiarities are described.

INTRODUCTION

WHILE not intending to minimize the experimental difficulties encountered in research at extremely high pressure, it is the feeling of the author that much more work would be done in this field if the relative ease and safety with which such work can be carried on were more generally known. P. W. Bridgman[1] in his article, "The Technique of High Pressure Experimenting" and in his book "The Physics of High Pressures" gives a large number of very valuable methods of construction and techniques.

It is the intention in this paper to describe a number of pieces of high pressure equipment not already described in the literature, with particular attention to equipment involving windows for optical methods of attacking high pressure problems. In the treatment of the more important pieces an attempt will be made to give a sufficiently detailed description so that anyone wishing to construct and use them should not encounter great difficulty. These methods are presented as solutions of particular problems that have been encountered by the author during the past few years of designing and constructing high pressure equipment. It is not claimed that there are not other equally good solutions to these problems but only that the methods herein described have served very well.

SELECTION OF MATERIALS AND STEEL

The selection of the proper steel for the construction of high pressure equipment is of major importance from the standpoint of safety as well as that of obtaining experimental data.

Those parts of the equipment in contact with the material under high pressures are required to withstand either a great compressional force or a great tension. Steels that are specially developed for withstanding great compressional forces are of necessity too brittle to be used in places where a great tension is required. Likewise those having a high tensile strength must be sufficiently ductile so that they will adjust themselves to as nearly a uniform stress throughout their cross-sectional area as possible.

[1] P. W. Bridgman, Proc. Am. Acad. Arts and Sci. **49**, 627 (1913–14); The Physics of High Pressures, Macmillan Co., 1931.

After experiencing what might have been some very serious explosions from the use of carbon steels and some alloy steels of unknown physical properties, an attempt was made to find a steel better adapted to the purpose. Upon request the author was supplied specifications as to physical properties of a large number of different grades of steels from several manufacturers. The three physical properties upon which the steel for the construction of cylinders was based were tensile strength, yielding point, and elongation.

High tensile strength and yielding point are necessary for obtaining the pressure, while the elongation largely determines the safety of such equipment. The steel selected as most nearly meeting these requirements was the S. A. E. 3250, steel manufactured by the Republic Steel Corporation. A typical analysis of which is: C 0.50, Ms 0.56, P 0.017, S 0.022, Si 0.224, Ni 1.76, Cr 1.08. This steel has a tensile strength as high as 300,000 pounds, depending upon the seasoning treatment. It also has a remarkably high elongation of about 10 percent. After machining it is heat treated as follows: Neutralized at 1600° to 1700°F, quenched in oil at 1450°F and salt drawn to 750°F. This treatment produces a Brinell hardness of about 445.[2]

Bridgman mentions some difficulty in obtaining pieces of steel free from flaws, a difficulty which has never been encountered with this steel.

There is a certain cylinder-wall thickness, determined by the inside diameter of the cylinder and the steel from which it is constructed, beyond which additional thickness does not add to the pressure the cylinder will withstand. This thickness is reached when the elongation of the steel on the inside surface of the cylinder wall reaches a point where cracks develop before much stress is produced in the steel near the outer surface. For the above steel and an inside cylinder diameter of about 1.6 cm the maximum strength is obtained with a wall thickness of perhaps less than 6 cm.

This difficulty can be partially overcome by a seasoning process described by Bridgman which consists of stretching the cylinders by means of pressure and then regrinding them before they are put into actual service. If this procedure is carried to the point where optimum conditions are obtained for the outer surface of the cylinder, the steel near the center has been elongated to a point where it has been weakened. In order more nearly to obtain optimum conditions throughout the cylinder wall, it is necessary to construct the cylinder of coaxial shells. These shells are made of such a size that when they are pressed together the optimum elongation occurs. Cylinders of this kind are being constructed and studied with the hope of obtaining higher pressure than has yet been produced in the laboratory.

The selection of a steel for the construction of such pieces as pistons or window supports where they must withstand very great compressional forces was based upon hardness and crushing resistance. The steel selected for this purpose is the Rex AA or Rex AAA high speed steel, depending on the hard-

[2] The steel for the construction of our high pressure equipment was supplied through the courtesy of the Republic Steel Corporation. The heat treatment was done by them after the piece had been machined.

ness desired.[3] The hardness and crushing resistence of these steels are largely determined by the particular heat treatment to which they are subjected. The crushing resistence may run to more than a million pounds per square inch.

The pressure equipment used by the author is always constructed in such a way that parts made from high speed steel are completely inclosed by a less brittle steel. In this way it is possible to work close to the crushing limit of the steel without danger from flying pieces.

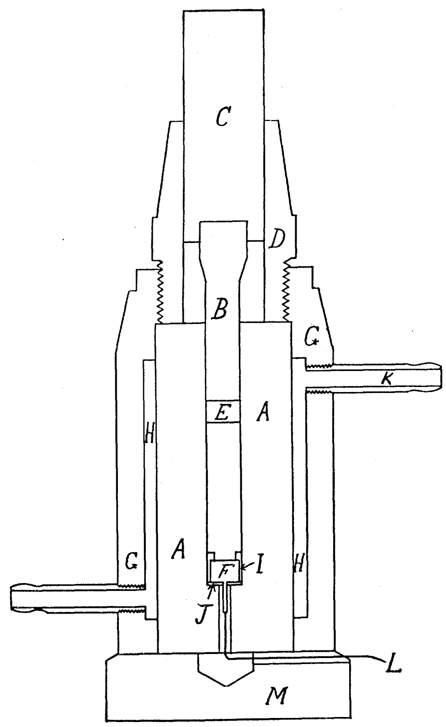

Fig. 1. High pressure cylinder with water jacket for temperature control and insulated electrode through the bottom of cylinder.

Construction of Pressure Chamber

The general method of developing the pressure is the same as that used by Bridgman and others; namely, the forcing of a small piston in a heavy cylinder by means of a hydraulic press.

The main body of the cylinder *A* Fig. 1 is constructed of S. A. E. 3250 steel, heat-treated to maximum tensile strength. The piston *B* is constructed of Rex AA or Rex AAA high speed steel. The piston guide *C* travels in the housing *D* thus preventing sidestrains on the piston. This is very important since even with the large four-rod two-hundred-ton hydraulic press used in this work sufficient sidestrains are produced to cause fracture of the pistons

[3] This steel is manufactured by and supplied through the courtesy of the Crucible Steel Company of America.

if such a guide is not employed. This housing also prevents flying pieces from getting out into the laboratory in case the piston shatters or a leak develops.

Fig. 2 shows the pieces of a piston which shattered while under 38,000 atmospheres pressure. Many pieces were imbedded in the soft steel housing surrounding the piston.

In one case an assistant received a severe bruise from flying oil and in another instance flying sulfuric acid penetrated two thicknesses of glass.

The rubber plug *E* makes a tight joint around the end of the piston. The housing *G* serves as a water jacket and as protection against flying pieces in case a cylinder splits. The cylinder is kept at constant temperature by circulating water from a thermostat around the pressure cylinder in the space *H* and the platins of the hydraulic press.

Fig. 2. Piston shattered at 38,000 atmosphere.

The body of the cylinder is machined from annealed steel allowing five or ten thousandths of an inch for grinding after being heat treated. They are ground on the inside with a high speed internal grinder.[4] This leaves the cylinder wall very smooth and reduces the friction of the piston to a minimum. The pistons are machined to a few thousandths of an inch oversize from annealed high speed steel. After machining they are heat treated and then ground to fit the cylinder tight enough so that from five to fifteen pounds are required to slip the piston in the cylinder. Before using, the center in the end of the piston used in supporting the piston in the lathe while machining should be ground off leaving the end of the piston flat. If this is not done the life of the piston is greatly reduced and the piston will usually split at the end before it has been used more than a few times. Leakage past the piston is prevented by placing a close fitting good grade rubber stopper from 5 to 10 mm in length, just ahead of the piston. This type of piston packing has four advantages over that used by Bridgman: (a) It can be used to a higher pressure because the entire cross-sectional area of the piston supports the pressure. There is no unsupported area in the center to make the pressure on the steel

[4] This grinder was purchased with a grant from the Iowa Academy of Science in 1928 and has been very valuable in the construction of pressure equipment.

of the end of the piston greater than the actual pressure in the cylinder. (b) Another advantage is that it offers less resistance to movements of the piston. (c) A rubber plug such as this is much more easily removed from the cylinder than the assembly used by Bridgman. (d) An important factor particularly in work of this type is its greater simplicity of construction and convenience of operation.

In this type of cylinder the material being studied under pressure is placed directly in the cylinder instead of in another chamber. This does away with the necessity of piping the liquid under pressure to the other chamber and thereby prevents difficulty due to leaks and transmission of pressure. When an electrical connection is necessary, a terminal is inserted through the bottom of the cylinder as shown in the figure.

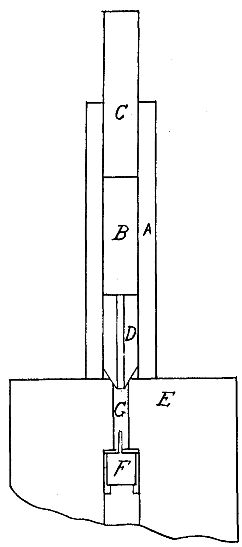

Fig. 3. Oil gun used to remove electrodes or pistons that have become fast in the cylinder.

The hardened high speed steel plug F is insulated from the sides of the cylinder by an enveloping shell of fiber I and from the end of the cylinder by a sheet of mica J about 0.5 mm or less in thickness. The insulated wire L is soldered to the plug F and is carried out through the hole in the bottom of the cylinder and through the groove in the base M. When the liquid in the cylinder is oil such electrodes serve very well at pressures as high as 30,000 atmospheres or higher and maintain a resistance of more than 300,000 ohms.

In case it is desired to remove such an electrode for any reason, a convenient method is to force it out with oil in the following manner. The cylinder is turned upside down and the wire pulled loose from F; a heavy walled tube A Fig. 3 having an inside diameter of 5/8 of an inch is placed over the center of the cylinder; the plug D having a 1/8 inch hole through the center is placed with its tapered end in the end of opening G through the bottom of the cylinder E; the tube is then partially filled with oil and the piston C put in place; this piston is then driven in with a heavy hammer and the oil will force the electrode out. This method can be used to remove a piston in case one sticks in the cylinder.

When assembled, the cylinders are mounted in a vertical, four-rod hydraulic press. The pressure is developed by forcing the piston in with the press. The pressure in the high pressure cylinder can be calculated by the following equation in which P is the pressure in the high pressure cylinder, A is

$$P = \frac{(Ap) - (F + f)}{a}$$

the area of the press cylinder, p is the pressure in the press cylinder, F is the frictional force of the piston in the press cylinder, f is the frictional force of the high pressure piston, and a the cross-sectional area of the high pressure piston. If the pressure is being decreased the quantity $(F+f)$ has a plus sign in front of it instead of a minus. The diameter of the hydraulic press cylinder used in this work is about seven and one half inches.

The leather used in the hydraulic press cylinder is a self-acting leather packing which offers a frictional resistance less than 0.5 percent of the total force.[5] The friction of the small cylinder was determined by means of a cylinder having a piston in each end. This system is put under pressure and the force necessary to move the cylinder over the two pistons is measured. With carefully controlled conditions of metal surfaces the friction of the high pressure piston is reproducible to within 1 percent. In some particular designs this frictional force may run as high as 8 percent of the total force.

The corrected values check closely with those obtained by the manganin wire resistance method up to 13,000 atmospheres. The pressure effect of the resistance of manganin wire has never been measured directly at pressures above 13,000 atmospheres. Since extrapolated values are never safe, particularly to the extent that would be necessary in working with pressure as high as 30,000 atmospheres, the author has chosen what seems to be a more direct and reliable method.

Construction and Behavior of High Pressure Optical Systems

In this part will be given an outline of the work leading to the development of a satisfactory window mounting for either glass or quartz windows for withstanding pressures of occasionally as high as 30,000 atmospheres. The field of optical studies has been one of considerable investigation since the early work of E. H. Amagat in 1887–96.[6]

Amagat constructed a window mounting consisting of a piece of glass in the form of a truncated cone with an enveloping cone of ivory or other material as a gasket between the glass and the steel. This window was mounted in a tapered hole with the small end of the window in the direction of the ob-

[5] Hydraulic Engineering, Houghton Research Staff, Page 48–49 (1926).

[6] E. H. Amagat, Journ. Chem. Phys. [6] **29**, 68, 96, 505 (1893); Comptes Rendus **105**, 165 (1887); **117**, 507 (1893). Notice sur les Travoux Scientifiques de M. E. H. Amagat Paris, 1896.

server. Numerous other investigators have used modifications of Amagat's window-mounting with varying degrees of success.[7]

In the course of an investigation[8] that was being carried out in this laboratory, it was shown that glass capsules made from Pyrex or soft glass tubing (6 mm inside diameter) by sealing the ends off round would sometimes withstand an external hydrostatic pressure as great as 12,000 atmospheres. These capsules were about 20 to 30 mm in length and had a wall thickness of about 1.5 mm. This led us to believe that if the proper window support could be obtained it would be possible to use windows at pressures much higher than those reported in the literature. A review of the literature showed that in all window mountings previously employed a gasket material of some kind had been used between the glass and the steel support. Some experiments were then recalled in which samples of very hard steel were being tested with the view of using it for the construction of pistons. In these experiments, samples of the steel one half inch in diameter and one inch long were placed endwise between two blocks of hardened steel and pressure was then applied until the

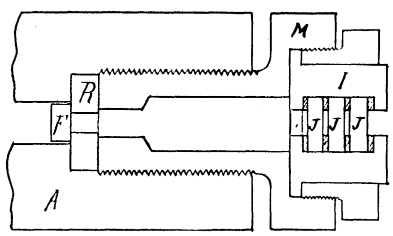

Fig. 4. Window mounting drawn to scale.

test pieces were crushed. Thinking that they would probably withstand a greater load if a thin sheet of soft steel were placed between the ends of the test pieces and the hardened steel blocks, sheets of soft steel 0.25 mm thick were so placed. Contrary to expectation, under these conditions, the test pieces were split at about half the load they had previously withstood.

Apparently in this case, the thin sheets of steel exerted lateral forces due to their flowing under pressure great enough to split the steel blocks. It seemed quite likely that a similar thing was happening in the case of the windows and the gasket material on which they were mounted. A window support must fulfill another condition, namely that its supporting surface must be perpendicular to the resultant force produced on it by the material in the cylinder. If such is not the case, the maximum strength of the glass cannot be utilized. With these facts as a guide the window mounting shown in Fig. 4 was constructed, in which the window F was mounted without a gasket and in contact with the polished surface of a very hard steel disk R about 10 mm in thickness with a 6 mm hole in the center.

The window and support were then mounted as shown in the figure and the cylinder and observation chamber filled with a light paraffin oil. Pressures

[7] Walter Wahl, Phil. Trans. Soc. **A212,** 117 (1913); Frances G. Wick, Proc. Am. Acad. Arts and Science **56,** 557–573 (1923).

[8] Thos. C. Poulter and Glen E. Frazer, paper in preparation.

were then applied in the usual way by forcing the piston in with the hydraulic press. The windows thus mounted would, in some cases, withstand a pressure of 30,000 atmospheres. The pressure in the observation chamber was determined as previously described. Thinking that this pressure was possibly in error as a result of the solidification of the oil, the following tests were made. These tests showed conclusively that there was but little pressure gradient in the oil and that therefore the pressures were substantially those computed.

For this investigation a number of other liquids, alcohol, ether, water, and glycerine were used to transmit the pressure. With alcohol, water, and ether the windows would usually break at pressures below 8000 atmospheres, whereas with glycerine the results were similar to those obtained with oil. Tests were then made to see if the piston was moving freely with oil or glycerine in the high pressure cylinder by means of the two-piston cylinder previously described. It was found that the pistons moved practically as freely with glycerine as with oil. Oil was then placed in the cylinder and observation chamber and its compressibility determined by means of the travel of the piston. There was detected no change in volume without a corresponding change in pressure such as would indicate a change of phase. However, such a change probably would not be expected for a material that is a mixture such as this paraffin oil. Some water was then placed in a thin walled rubber capsule in the observation chamber near the window, then with the remaining space filled with oil the piston travel was again measured as the pressure was built up. If the travel of the piston is measured as the pressure is built up, a decrease in volume is obtained with no increase in pressure between 10,500 and 11,000 atmospheres for a temperature of 30°C. This corresponds very closely to the pressure obtained by Bridgman for the formation of ice VI at that temperature. This shows that there is little or no pressure gradient in the paraffin oil below 11,000 atmospheres.

The freedom from a pressure gradient is probably to a large extent due to the compactness of the pressure system, which makes it necessary to transmit the pressure only a short distance. While it is not claimed that the viscosity of the oil has not reached a rather high value, the above experiments show that the plastic flow of the medium transmitting the pressure soon restores equilibrium.

In order further to test the ability of a window to withstand pressure a heavy-walled high-speed-steel tube was mounted in the pressure chamber of the cylinder shown in Fig. 1. One end of this tube is over the opening in the bottom of the cylinder and a window is mounted on the other end. This brought the window to within two centimeters of the end of the piston. In this case the pressure would be transmitted to the window even though the oil did solidify. Two windows were tested in this position, one breaking at 25,000 atmospheres and the other at 35,000 atmospheres. A window was then mounted with a little water in contact with it and the pressure was transmitted by means of oil. The oil and water were separated by a thin sheet of rubber. Under these conditions windows were again broken at pressures below 8000 atmospheres.

A window was then mounted in the middle of a piece of rubber tubing with oil on one side of the glass and water on the other. The two ends of the tube were then stoppered and this tube was placed in the observation chamber and surrounded with oil. A pressure of 21,000 atmospheres was built up and allowed to remain for about five minutes. It was then released and the side of the window that was in contact with the water was badly cracked and chipped while that in contact with the oil was intact. This shows that the breaking of the windows is in some way related to the material in contact with the glass as well as to the actual pressure employed. Now if a large pressure gradient does not exist, as has been shown, and the friction of the piston is corrected for, as described above, it is the belief of the author that the pressures used in this investigation are substantially those recorded.

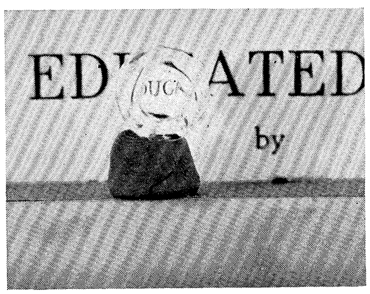

Fig. 5. Diverging lens effect of fractured window.

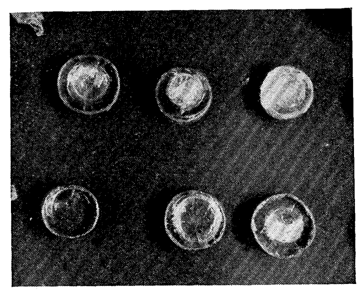

Fig. 6. Fractured windows before concentric shells have been removed.

Some valuable information relative to the difference in the behavior of windows when different liquids are used in contact with them can be obtained from the different types of fractures produced. A pressure window will frequently hold the pressure with but little leakage for some time after it has developed many cracks. Where such windows have been removed usually one of two general types of fracture has occurred.

The first is a spherical concentric shell fracture with the outside of the curvature towards the pressure. Sometimes four or five such shells break out leaving surfaces smooth enough to produce a diverging lens which will produce a fairly uniform image. Fig. 5 shows such a window mounted two inches in front of a printed page.

In all cases where such fractures have occurred the window has either been mounted on a gasket and the pressure transmitted by any of the five liquids used or with no gasket and the pressure transmitted by water, alcohol, or ether.

The second type of fracture occurs when no gasket is used and the pressure is transmitted with oil or glycerine. With this type of fracture only a thin layer of the glass which is over the opening in the window support is chipped off leaving a rough surface. Fig. 7 shows a group of such windows, placed fractured face down for photographing. The photograph also shows freedom from other cracks. In either case the main portion of the window may be free from cracks. The above facts lead the author to suggest the following explanation for the difference in the behavior of pressure windows as different liquids are used to transmit the pressure. The concentric shell fracture, in cases where a gasket material is used, is probably due to the movement of the gasket as it flows under pressure. This would give the effect of a window supported on a moving surface. Since the fractures are of the same type with no gasket and the pressure being transmitted with water, alcohol, or ether, as those produced with a gasket and any of the liquids employed, the strains in the two cases must be similar. It may, therefore, be that these mobile liquids penetrate between the window and its support, thus producing the effect of a moving support. The relatively high viscosity of the oil and glycerine, particularly at the higher pressure, probably prevents such penetration thus making it possible to utilize the maximum strength of the glass or quartz.

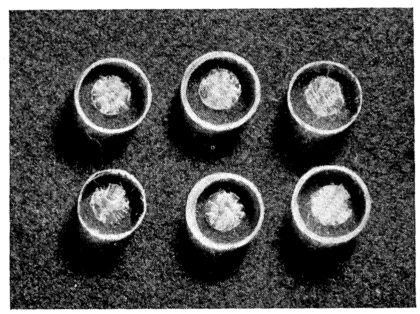

Fig. 7. Windows showing second type of fracture. Top row is quartz and bottom row is glass.

It is, therefore, the belief of the author that if glass or quartz windows are properly mounted they will withstand a pressure of 30,000 atmospheres provided that a light paraffin oil is used to transmit the pressures.

For work in which it is necessary to pass light into the material under pressure or make observations where windows are necessary, it has been found most convenient to construct the pressure cylinder and the observation chamber of a single piece of steel. In assemblies of this kind the end of the cylinder in which the pressure is developed opens into the observation chamber. This does away with the necessity of making connections to transmit the pressure from the cylinder to the observation chamber, and thus decreases the chances of leaks. This cylinder and observation chamber is enclosed in a water jacket for temperature control.

That portion of the cylinder in which the pressure is developed is identical with the corresponding part of the cylinder shown in Fig. 1. The observation chamber L contains the material being studied. The glass or quartz windows F are mounted against the polished surface of the high-speed-steel window supports R which are in turn held in place by means of the plugs M. The polished surface of the window support makes a tight joint with the shoulder at the end of the observation chamber. The safety windows J are made of the same material as that used in the pressure windows and are mounted between fiber washers and held in place by the housing I. In earlier work only one safety window was used on each end but experience showed that one was insufficient. The purpose of the safety window is to catch flying pieces in case a pressure window shatters or a leak develops. The windows K are mounted in a metal tube which fits the inner end of the plug M. This puts them on the opposite side of the window support from the pressure windows. The intervening space is filled with a liquid having the same index of refraction as that of the pressure window. This corrects for the lens effects, due to the bulging of the pressure windows.[9]

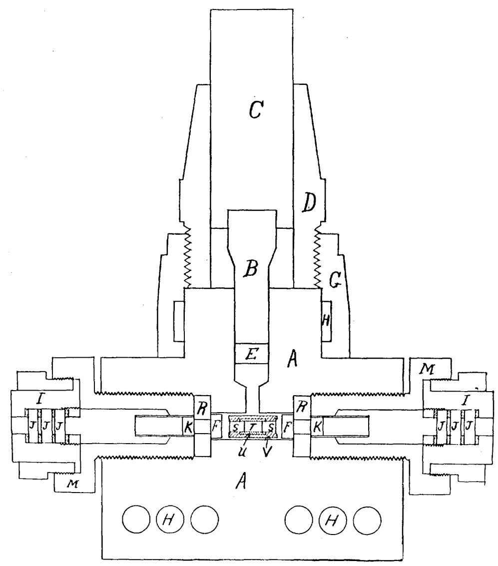

Fig. 8. Pressure equipment with windows in place, drawn to scale.

The window is a piece of glass or fused quartz approximately 11 mm in diameter and from 4 to 10 mm in thickness. The surface of the window support is ground flat to an accuracy of 1×10^{-4} cm which is measured by placing the window in contact with its surface and counting the interference fringes produced between them. These windows are held in place while assembling the cylinder by a small quantity of liquid balsam. The pressure is applied slowly so as to permit the balsam to flow out and allow the window to come in contact with the steel support. The liquid in the observation chamber has access to all sides of the window except its supporting surface thus producing as nearly a compressional force on the window as possible.

If it is desired to separate the oil in the cylinder from any other liquid being investigated, it is most convenient to place such a liquid in a small cell

[9] Thos. C. Poulter and Carl Benz. The Line Effect of Pressure Windows, Phys. Rev. 40, 872. (1932).

with windows in each end. This cell is placed in the observation chamber and surrounded with oil. If a constant quantity of material is desired in the light path it is necessary to use a telescoping cell.

If a constant length of a column of liquid is desired, the windows can be rigidly mounted in the ends of a hard steel tube. This tube can have a number of holes in the side which are covered by placing this tube in a thin walled

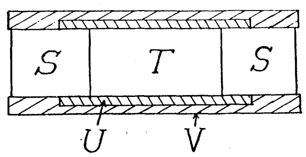

Fig. 9. Telescoping cell. U=high speed steel cell case with movable windows S. V= rubber tube surrounding cell and windows.

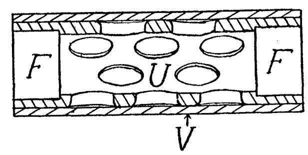

Fig. 10. Constant length cell. V=rubber tube stretched over outside of cell. F=windows cemented in place. U=high speed steel body of cell.

rubber tube. The compressibility of the hardened high speed steel is so small that very little error is introduced thereby.

While the author has had many experiences with explosions of high pressure apparatus that he does not care to have repeated, it is his belief that there is very little danger attached to investigation of this kind if the precautions herein described are carefully observed.

The author wishes to express his appreciation for the assistance in the form of a grant from the Bache Fund of the National Academy of Science.

Fig. 2. Piston shattered at 38,000 atmosphere.

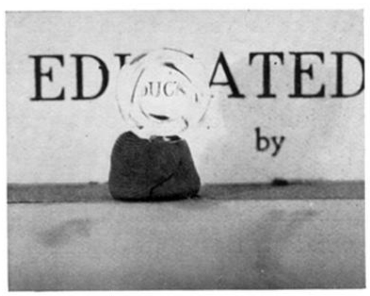

Fig. 5. Diverging lens effect of fractured window.

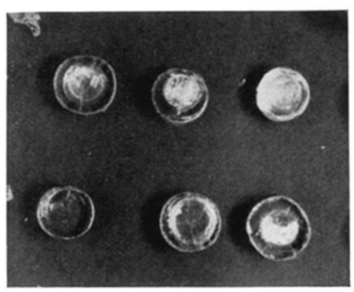

Fig. 6. Fractured windows before concentric shells have been removed.

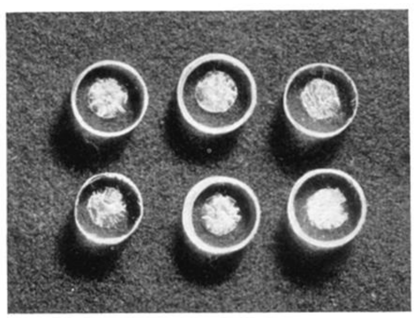

Fig. 7. Windows showing second type of fracture. Top row is quartz and bottom row is glass.