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Reprinted from The Review of Scientific Instruments, Vol. 30, No. 10, 874–880, October, 1959
Printed in U. S. A.

Nonmagnetic High-Pressure Vessels*

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(Received May 19, 1959; and in final form, June 25, 1959)

The use of beryllium copper and stainless steel in the construction of pressure vessels to contain 20 000 kg/cm² is described. Machine drawings are given for the construction of different types of pressure vessels, different types of seal, and methods of introducing electrical leads into the apparatus. Experience in the use of high-pressure vessels and plugs beyond the elastic limit of the construction material is reviewed.

1. PURPOSE AND HISTORY

IN this article we propose to review our progress in producing nonmagnetic high-pressure vessels sufficiently strong to hold pressures up to 20 000 kg/cm², but small enough to be placed in a temperature regulating bath between the pole pieces of an electromagnet. Two types of material seem to be suitable for such pressure vessels. The first is beryllium-copper, which, although used previously elsewhere,[1] was first applied at Harvard University by G. B. Benedek in a vessel for nuclear resonance experiments. The details of Benedek's apparatus are found in a paper by Benedek and Purcell[2]; smaller beryllium-copper vessels have since been used for nuclear resonance work by Kushida, Benedek, and Bloembergen[3] and Benedek and Kushida[4]. The high-pressure vessel used by Benedek and Purcell was also used by Benedek, Paul, and Brooks[5] in investigations of the transport properties of carriers in semiconductors. Partly as a result of this work it became evident that some worthwhile experiments would demand higher pressures than had hitherto been contained by beryllium-copper vessels. The major part of this paper is devoted therefore to a discussion of the pressure vessels built and tested to extend the range of bombs to the highest possible pressures.

The second type of material that can be used is one of the nonmagnetic stainless steels. Nonmagnetic, Type 316, stainless steel tubing of quite considerable strength has been in use in this laboratory since 1956,[6] which led us to believe that this stainless steel, properly work-hardened, would be suitable material for pressure vessels. We were fortunate in securing two samples of work-hardened stainless steel through the courtesy of Dr. Lloyd Nesbitt of General Electric; our tests of these will also be described in the following.

2. BERYLLIUM COPPER PRESSURE VESSELS

The first tests used the sample cylinder shown in Fig. 1. The piston hole was carefully reamed and a radius left in the bottom of the hole. The piston was made of tool steel, heat treated to maximum hardness, and drawn in boiling water to anneal out hardening strains. It was ground to be a slide fit to the piston hole. The two washers shown were made of different hardnesses. One was fashioned with a leading coned edge that was deformable under pressure. The first tests were carried out using a lead-indium pressure transmitting medium. This medium transmits the pressure, yet does not leak past the piston when the piston hole expands because of plastic flow. Such a solid was often used in initial tests of cylinders; since relatively little volume change is required to raise the internal pressure to a high value, the test vessel can be subjected to high pressures over practically its entire length. The advantages of easy sealing and small volume change are sacrificed when true fluid transmitters are used; on the other hand, there is no

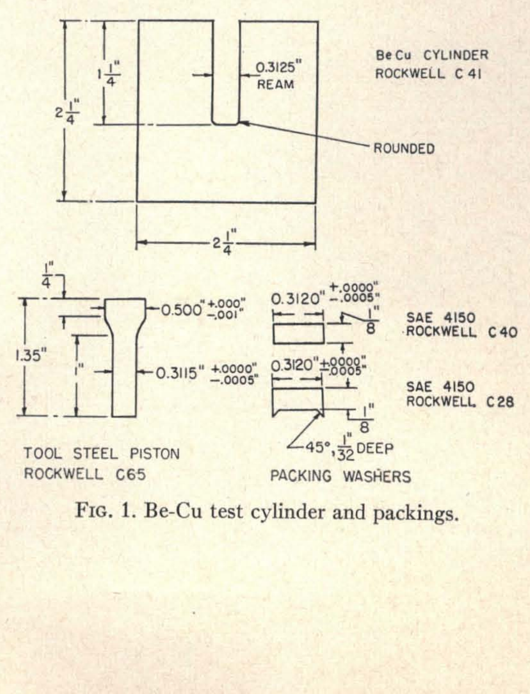

FIG. 1. Be-Cu test cylinder and packings.

* The research in this document was supported jointly by the Office of Naval Research under contract with Harvard Univeristy, and by the Army, Navy, and Air Force under contract with the Massachusetts Institute of Technology.

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[1] H. Ebert and A. Kussmann, Physik Z. **38**, 437 (1937).

[2] G. B. Benedek and E. M. Purcell, J. Chem. Phys. **22**, 2003 (1954).

[3] Kushida, Benedek, and Bloembergen, Phys. Rev. **104**, 1364 (1956).

[4] G. B. Benedek and T. Kushida, J. Phys. Chem. Solids **5**, 241 (1958).

[5] Benedek, Paul, and Brooks, Phys. Rev. **100**, 1129 (1955).

[6] W. Paul and D. M. Warschauer, Rev. Sci. Instr. **27**, 418 (1956).

doubt about possible pressure gradients in the pressure transmitter, and unevenness of pressure seasoning. The pressure was developed using a press of the type described by Bridgman.[7] The beryllium-copper flows plastically and work hardens at pressures above 10 000 kg/cm². The piston and sealing washers expand when the bore of the vessel expands, and it may be difficult to remove them if the expansion is too great. For this reason the maximum pressure was gradually increased in several runs; at the end of each test the hole and piston dimensions were measured, and appropriate changes in the diameter of the latter made. A typical set of four runs showing piston displacement plotted against the pressure behind the piston is shown in Fig. 2. Half of the width of the hysteresis loop for any one run is a rough measure of the friction between the piston head and the cylinder wall. The change in slope of the piston displacement *vs* pressure curve immediately the pressure is increased beyond its previous maximum is evident; such a change usually indicates stretching of the cylinder but is also found if the pressure in the cylinder is leaking at some packing. The cylinder shown in Fig. 1 successfully held 20 000 kg/cm² in the tests of Fig. 2; the piston hole stretched 0.022 in. near the bottom in the process.

After the cylinder had been stretched by a solid pressure transmitter, it was reamed straight, and the new piston and piston head of Fig. 3 made, so that a test with liquid could be carried out. In the particular cylinder of Figs. 1

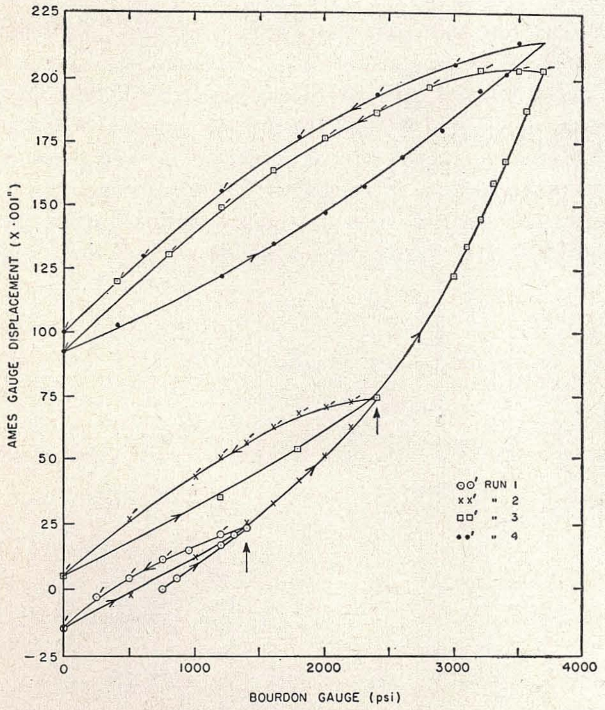


FIG. 2. Pressure tests on cylinder of Fig. 1.

[7] P. W. Bridgman, *The Physics of High Pressure* (G. Bell and Sons, London, England, 1931).

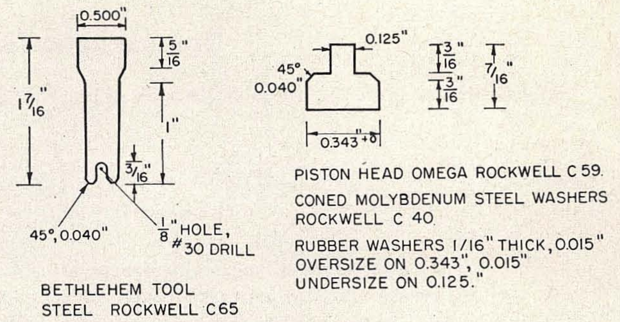


FIG. 3. Piston and packings for liquid tests.

and 2, these tests were unsuccessful; the pressure leaked past blowholes in the walls of the piston hole. This experience is an example of the special troubles that can be encountered with Be-Cu, unless care is taken to secure stock free of blowholes. Another cylinder without blowholes of almost identical dimensions, and subjected to the identical initial stretching procedure with a lead-indium pressure transmitter, held fluid pressures to 28 500 kg/cm² in repeated use without perceptible leak.

If the Be-Cu pressure vessel can be used in the form shown in Fig. 1 without a second inlet for electrical leads, its usable limit is at least 28 000 kg/cm². Very often it is necessary to take several electrical leads out of the vessel by a second port. In our experience we have found that the usable pressure limit is set, not by the bursting pressure of the cylinder, but by the leaking at one of the seals. In order to keep the number of seals into the vessel as low as possible, we built the cylinder shown in Fig. 4. This vessel was machined from a single piece of beryllium-copper. The pressure was produced by a piston and piston head operating in the top ½-in. hole, and the bottom hole was sealed with a plug very similar to that which will be described in connection with a cylinder of later design. Unfortunately, this vessel burst at an internal pressure of 110 000 psi with a crack some two inches long, located near the center of its length, despite the fact that the tensile strength of the material of the bomb in its hardened state was about 200 000 psi. Bridgman[7] noted that the bursting pressure in his tests of cylinders was very often about twice the nominal tensile strength.

When the burst pressure vessel was sectioned, a large blowhole was found exactly in the region of the break. It was concluded that the most likely cause of failure was that the blowhole had acted as a stress-riser, causing local failure which propagated through the rest of the material. The initial cause was bad casting by the suppliers; forging subsequent to the casting was unable to close the blowhole. It seems clear that beryllium-copper ingots should be cooled from the bottom at a slow enough rate that gas bubbles trapped in the melt in the pouring process will have sufficient time to reach the upper surface. As a result of this failure, we have advised the suppliers of subsequent

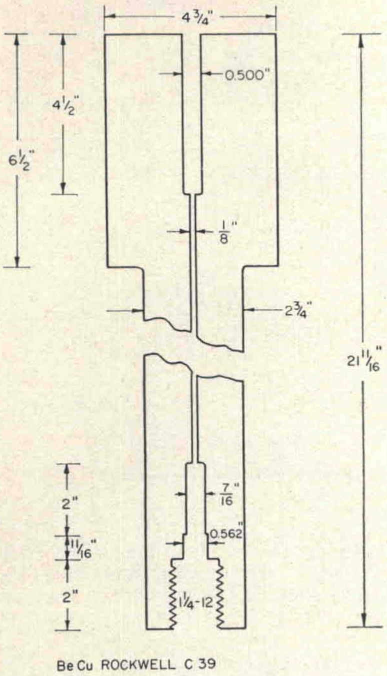


FIG. 4. Be-Cu pressure vessel.

bars of beryllium-copper to cut the pieces from the bottom of the ingot and to test them immediately for pipe or porosity using deep etchants. In addition, we have sent all pieces of beryllium-copper used in our pressure work for tests by ultrasonic methods for flaws or blowholes. The ultrasonic frequency was 30 Mc, so that flaws of, say, $\frac{1}{10}$ mm diam can be expected to be detectable. No beryllium-copper piece successfully tested in this way failed as a pressure vessel. This is not to say that flaws smaller than this cannot act as stress risers and cause eventual failure; nor is it to say that beryllium-copper with detectable flaws of this magnitude must always fail as pressure containers.

All of our subsequent work on beryllium-copper has been carried out on much smaller cylinders with the pressure producing apparatus separate, but connected to the vessel by tubing. This has the advantage of allowing us to work with smaller pieces of beryllium-copper, which are presumably easier to produce and also to heat treat, but unfortunately has the disadvantage that more seals into the pressure vessel are required. It has turned out over a period of years that the difficulty of sealing into the pressure vessel has been the worst one to solve. It should be added, however, that the separation of the pressure-producing vessel from that in which the experiment is done allows the latter to be placed in a temperature controlled enclosure in a magnetic field with much greater facility. Since 1954 our laboratory has used, as mentioned,[6] hard drawn type 316 stainless steel tubing in connecting pressure vessels, and this too is a great advantage when it comes to controlling the temperature of the final bomb or positioning it accurately. While it is still felt that one-piece vessels of well-cast, well-hardened beryllium-copper are best if one wishes to attain really high pressures, it is likely that for many practical purposes the difficulty of sealing into smaller vessels with the attendant advantages of flexibility and temperature control is one that can be borne easily.

In Fig. 5 is shown a diagram of the beryllium-copper vessel which we have taken to the highest pressures under actual conditions of solid-state research. The pressure is generated in a separate steel cylinder and led through stainless steel tubing into the bomb via sealing plug A, Fig. 6. The tubing is $\frac{3}{16}$ in. in outer diam, but in most of our tests the $\frac{1}{8}$ in. diam tubing was used.[6] Although this tubing has been used to pressures in excess of 20 000 kg/cm², it occasionally burst at around 18 000 kg/cm². Therefore, we had especially made up for us some $\frac{3}{16}$-in.

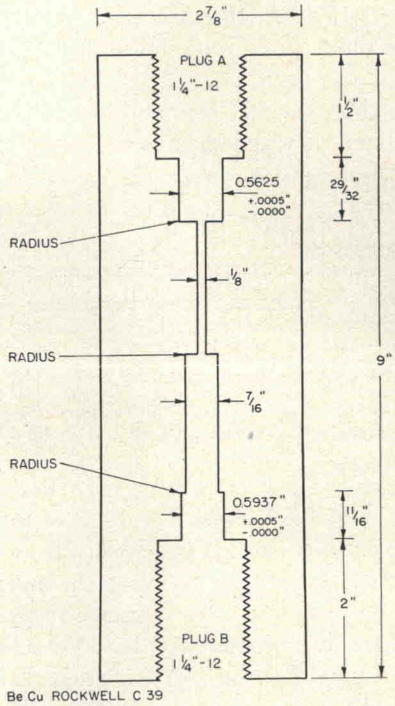


FIG. 5. Be-Cu pressure vessel for work to 20 000 kg/cm.² Diameters given are before pressure stretching.

diam, hard drawn, type 316 stainless steel tubing which has been tested successfully to pressures up to 23 000 kg/cm², and at the time of writing has only once burst at lower pressures.[8] This has also been reported briefly.[9] Plug A and the extractor in front of the washers can be made of hardened steel, if this part of the apparatus is out of the magnetic field; otherwise Be-Cu or Everdur is used. The operation of this type of plug in sealing the pressure is described in Bridgman's book,[7] and is illustrated in Fig. 8.

The electrical terminals lead into the apparatus through a 7-terminal plug B, as shown in Figs. 7 and 8. This plug

[8] This burst occurred where the tubing had been filed and heated.
[9] D. M. Warschauer and W. Paul, Rev. Sci. Instr. 28, 62 (1957).

is of a type used for many years by Professor Bridgman[7]; its design is particularly suitable for this pressure vessel. A discussion of some of the criteria we have found necessary for successful sealing in our pressure vessels of plugs such as A and B will be given below.

This pressure vessel has been tested many times to 20 000 kg/cm^2. In its initial tests, the cylinder stretched several thousandths of an inch. When the stretch in the sealing holes became unbearably large, the holes were reamed or bored straight again and new sealing plugs made. After about six applications of pressure, most of the stretching ceased. The difficulties left were then not those of having a pressure vessel that would hold pressure of this magnitude, but of making seals into the pressure vessel that would not leak slowly with time. To solve this difficulty we have tried many arrangements of the type described by Warschauer and Paul,[6,9] but have found nothing quite as satisfactory as plugs of the type shown in Figs. 6 and 7.

After the initial stretching ceased, the 0.5625-in. sealing hole reached the configuration and diameters shown in Fig. 9. The small stretch at the bottom is a result of the support given this part by the material underneath it.

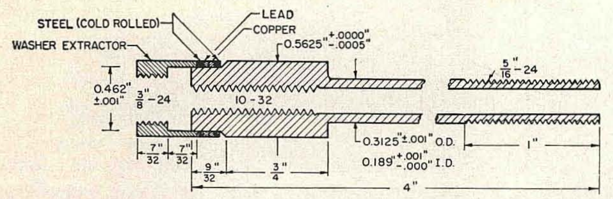


Fig. 6. Sealing plug for Be-Cu vessel of Fig. 5. Plug A. The tubing used in the above plug is $\frac{3}{16}$ in. o.d. A drive plug with a $1\frac{1}{4}$ in.-12 thread backs up plug A.

Since the pressure seal is at least $\frac{1}{8}$ in. inside the hole, the material between this point and the entrance to the hole is not stretched by the fluid pressure, but by expansion of the $\frac{9}{16}$-in. stem of the plug. The final configuration near the hole entrance depends on this expansion: if it is small, the hole will remain small at the mouth, even if it expands by plastic flow inside. Difficulty may then be experienced in removing the sealing plug from the hole. Subsequent sealing without remachining is then difficult. We have found it a good general rule to avoid the removal of material that may have been work hardened under pressure. When a sealing hole is not quite straight, or when it has "belled" only slightly, we have found it best to continue the same sealing arrangement using brute force when necessary to obtain an initial seal. Sometimes the plug expands in compression plastically, so that the diameter of the sealing hole at its mouth increases, and the plug becomes cone-shaped. Then it is advisable not to try to straighten plug and hole, but to continue sealing the coned surfaces; if this becomes too difficult in a case of great expansion, the washers can be made larger, and, in the last resort, remachining of plug and pressure vessel done.

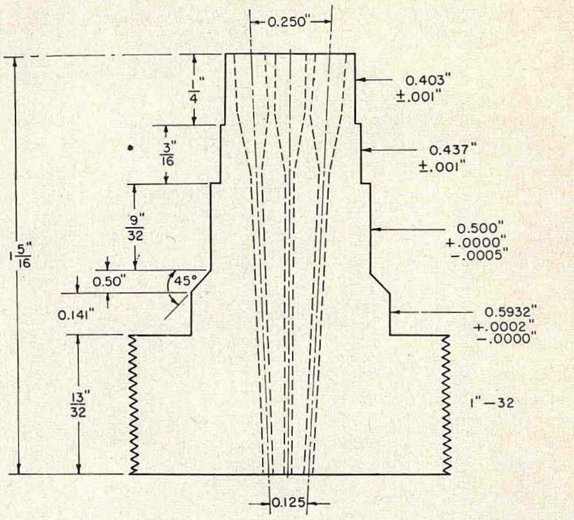


Fig. 7. 7-terminal Be-Cu plug for vessel of Fig. 5. Plug B, Rockwell C 40. The details of the terminals are shown in Fig. 8. A drive plug with a $1\frac{1}{4}$ in.-12 thread backs up Plug B. The extractor and washer assembly are similar to those in Fig. 6.

The sealing plug A is usually used with two $\frac{1}{16}$-in. thick cold rolled steel washers and one $\frac{1}{16}$-in. thick lead washer. On occasion, a $\frac{1}{16}$-in. thick copper washer was inserted between the lead and one of the steel washers as shown in Fig. 6; this is not felt to be absolutely necessary. In use, the drive plug is tightened before the pressure run until the back steel washer has flowed part way down the 45° cone. The pressure flows the washer still further; leakage of fluid occurs when the washer has flowed all of the way down the cone.

It is generally advantageous to arrange the seal so that

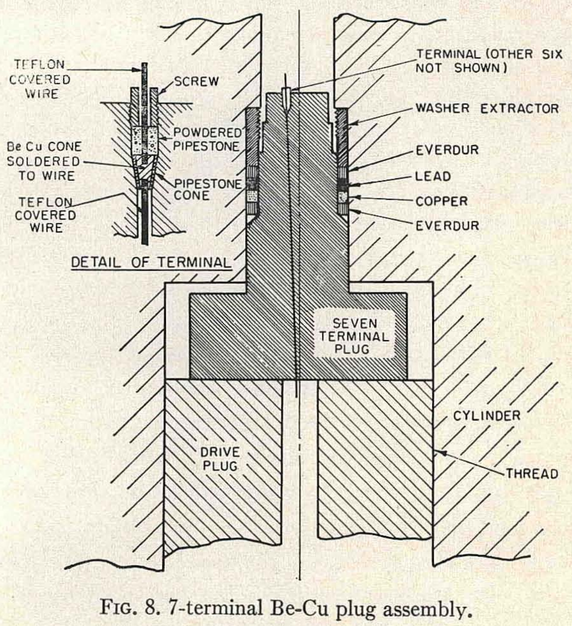
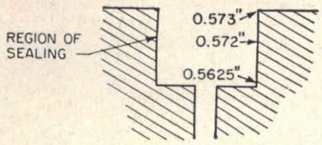
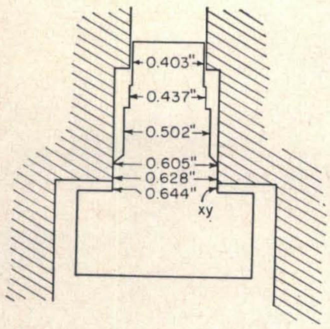


Fig. 8. 7-terminal Be-Cu plug assembly.



Dimensions of sealing hole for plug A (see Figs. 5 and 6 for dimensions before stretching).



Dimensions of plug B and sealing hole for plug B (see Figs. 5 and 7).

FIG. 9. Dimensions of pressure cylinder after stretching.

the flowing steel washer is well inside the sealing hole. Presumably this is because the wall has some support there. It is also advantageous to make the length of the 0.5625-in. diam stem projecting from the sealing hole as short as possible. Neglect of this precaution may result in a stem expanded and shortened by the high compressional stress under pressure.

The present dimensions of the first seven-terminal plug and its sealing hole are also shown in Fig. 9. It is seen that this hole stretched more than the other, presumably because there is less supporting material around it. The stem of the plug is coned, partly to match the hole, but partly because the section xy is outside of the sealing hole, and, because it is unsupported, expands radially in compression. The succession of washers has worked well consistently in sealing, provided they are made a push fit on the plug stem and in the sealing hole. Little trouble is experienced with the electrical terminals inside the plug. We have made five 7-terminal plugs for this pressure vessel. It is essential that the Be-Cu be entirely free from flaws. The placing of the seven terminal holes is a tricky machining operation, and at least one plug failed because two holes were too close together. The most common cause of loss, in our experience, has been the result of unsuccessful attempts to remove a Be-Cu cone terminal that has shorted electrically in its sealing hole; if the pressure has firmly forced the cone into place, it is almost impossible to remove it.

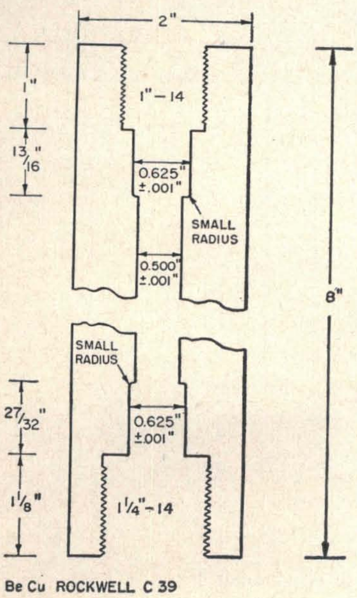


FIG. 10. Be-Cu pressure vessel for Hall effect measurements.

This vessel is in use for Hall effect measurements to

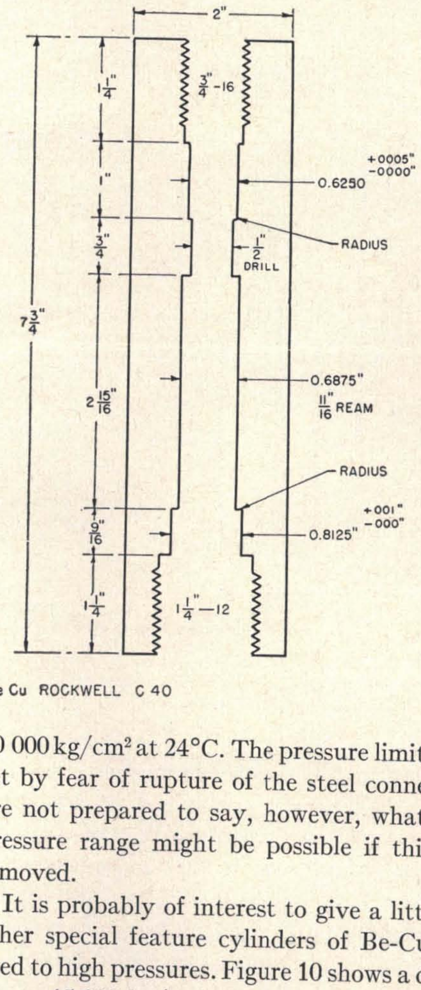


FIG. 11. Be-Cu pressure vessel for microwave measurements.

20 000 kg/cm^2 at 24°C. The pressure limitation is at present set by fear of rupture of the steel connecting tubing. We are not prepared to say, however, what extension of the pressure range might be possible if this limitation were removed.

It is probably of interest to give a little information on other special feature cylinders of Be-Cu that have been used to high pressures. Figure 10 shows a cylinder in regular use to 15 000 kg/cm^2. Its interest lies in the fact that the o.d. to i.d. ratio, 4:1, is considerably smaller than in the one we have described in detail. At this pressure the sealing holes had stretched 0.005 in. from their initial dimension. The usable limit for the vessel is not known. Figure 11 shows another vessel of 3:1 o.d. to i.d. ratio that has been used to 10 000 kg/cm^2, in an application where the highest pressure is not necessary but a large internal diameter (for a sample holder) and a small external diameter (for high-magnetic field) are overriding considerations. Figure 12 shows the typical vessel used for nuclear magnetic resonance work at Harvard.

3. STAINLESS STEEL PRESSURE VESSELS

Finally, we shall report briefly our limited experience in testing two stainless steel vessels constructed from the material supplied by General Electric.

Figure 13 shows a vessel of G.E. type 316 steel that had been hardened by cold working to Rockwell C 32 before machining. In its first tests, the hole marked as 0.500 in.

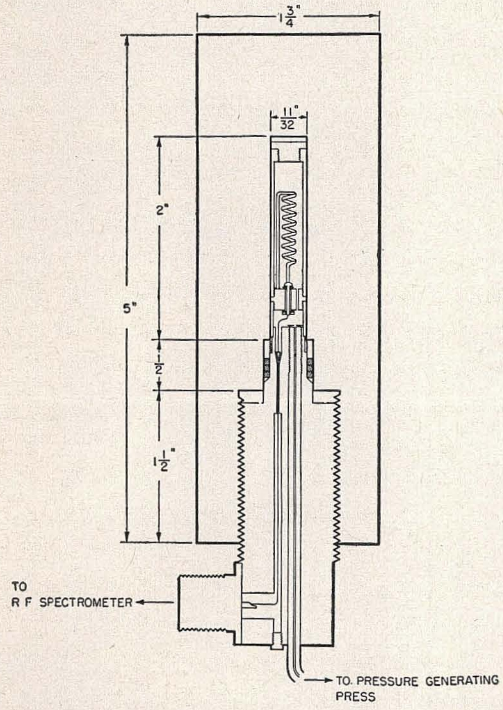


FIG. 12. Be-Cu pressure vessel for nuclear magnetic resonance measurements.

diam was only 0.125 in and the 0.5625-in. hole measured only 0.500 in. The pressure was introduced via a conventional T plug and $\frac{1}{8}$ or $\frac{3}{16}$-in. tubing in the manner explained in a previous paper.[9] Although the material started to flow plastically at 11 000 kg/cm^2, the pressure was increased to a maximum of 14 000 kg/cm^2. A second pressure run showed that permanent deformation had taken place, but that the material had work hardened to include 14 000 kg/cm^2 in its elastic range. When further work hardening was attempted, the cylinder stretched very rapidly; subsequent examination showed that the bore increased from 0.500 to 0.562 in. and that the outside had expanded locally by 0.020 in. The vessel was then drilled and reamed to the dimensions of Fig. 13 with a wall ratio of 3:1. The first

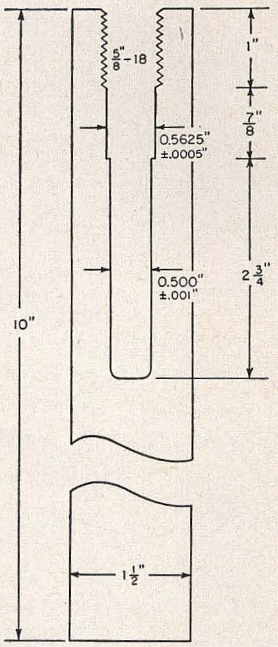


FIG. 13. Stainless steel pressure vessel. Rockwell C 32.

application of 10 000 kg/cm^2 stretched the holes 0.005 in., the second showed that the material had work hardened to include this pressure in its elastic range. 10 000 kg/cm^2 is probably close to the limit of usable pressure for this cylinder with this wall ratio.

Figure 14 shows a second vessel of the same material, similarly hardened to Rockwell C 32 before construction. The wall ratio is much larger, approximately 9:1. In its first tests this vessel had a 0.452 in. diam, $2\frac{1}{8}$-in. deep blind hole. The sealing piston, piston head, and washers were of the same design and of the same material as those in Fig. 3. In its first tests under hydrostatic pressures to 23 000 kg/cm^2, the hole stretched to 0.467 in. uniformly over its length. Subsequent tests to the same pressure produced no further stretching of the hole.

A second series of tests was carried out with the configuration shown, so that the sealing properties (without

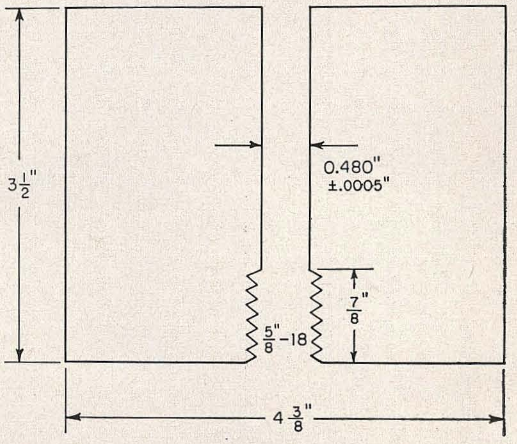


FIG. 14. Stainless steel pressure vessel. Rockwell C 32.

slow leaking) could be tested as well as the gross pressure holding behavior. The seal at the bottom of the cylinder was a conventional T head one, shown in Fig. 1 of reference 7. In these tests the cylinder held 20 000 kg/cm^2 for three days. The *average* leak rate over this period was 25 $kg/cm^2/hr$, but the terminal rate was much less than this. At the end of the test the sealing washers, plugs, and cylinder bore had all flowed slightly. The cylinder bore had increased to 0.486 in. near its bottom; however, the stainless steel there was work-hardened in the process.

Beryllium-copper and stainless steel are probably equally suitable materials for high-pressure vessels. The stainless steel is easily machinable in the hardened state, should be free from flaws in manufacture, and has low heat conductivity which may be important in some applications; on the other hand, the beryllium-copper is easily machinable when soft, can be hardened much more easily in the average laboratory than can the steel, and has been tested under more rigorous conditions over a longer time.

ACKNOWLEDGMENTS

It is a pleasure to record our appreciation of the help given to us by Professor P. W. Bridgman over a period of years. We have also benefited from discussions with Professors Harvey Brooks, Bruce Chalmers, and Richard S. Davis. The development of the techniques was greatly aided by the laboratory work of Dr. M. G. Holland, Dr. M. I. Nathan, and Dr. A. C. Smith. Dr. Holland and Dr. Smith completed the testing of the stainless steel cylinders. The careful machine work on the cylinders and plugs was carried out by Mr. Charles Chase, Mr. Stanley Gardner, Mr. James Inglis, and Mr. Alden Peavey.