April 24, 1962 H. M. STRONG 3,030,661

HIGH PRESSURE REACTION VESSEL

Filed March 31, 1960

Fig. 1.

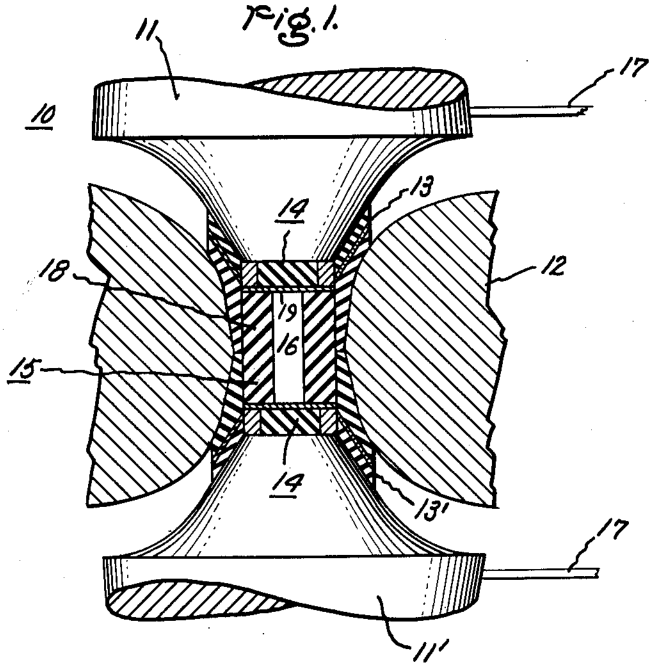


Fig. 2.

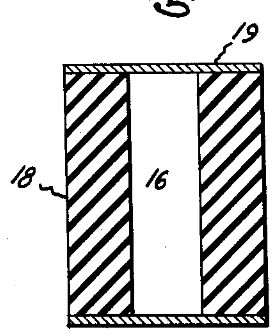
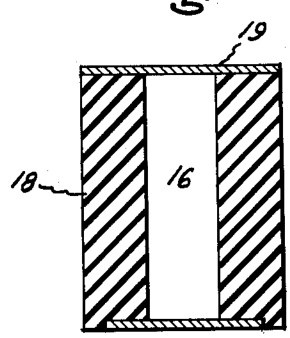


Fig. 3.



Inventor:
Herbert M. Strong,
by James J. Lichiello
His Attorney.

3,030,661

HIGH PRESSURE REACTION VESSEL

Herbert M. Strong, Schenectady, N.Y., assignor to General Electric Company, a corporation of New York

Filed Mar. 31, 1960, Ser. No. 19,023
11 Claims. (Cl. 18—34)

This invention relates to reaction vessels generally, and more particularly to reaction vessels which contain an object or material to be subjected to very high pressures and high temperatures for extended periods of time. This application is a continuation-in-part of copending application Serial No. 537,369, Strong, filed September 29, 1955, and assigned to the same assignee as the present invention and now abandoned.

A reaction vessel generally, is an enclosure, chamber member or receptacle, adapted to contain an object or a specimen material which is to be subjected to very high pressures and temperatures. The receptacle is positioned in a high pressure apparatus such that it is compressed and deformed to a considerable degree together with the material which it contains. In copending application Serial No. 707,432, filed January 6, 1958, and assigned to the same assignee as the present invention, and now Patent 2,949,248 granted June 21, 1960, there is disclosed and claimed one exemplary form of a high pressure high temperature apparatus employed to compress a reaction vessel and the object which it contains. The aforementioned copending application is accordingly incorporated by reference herewith.

Heretofore, various materials have been satisfactorily employed for reaction vessels including pyrophyllite, catlinite, various ceramics, etc. These materials are employed because of various reasons including, that where the vessel is to be subjected to high pressures and high temperatures, the material must have good thermal insulating properties, not only at atmospheric pressure but at high pressures and high temperatures that may be employed; for example, pressures in the range of 40–90,000 atmospheres and greater, and temperatures on the order of, for example, 3000° C. In addition to thermal insulating qualities at these pressure-temperature conditions, where electrical resistance heating is employed to heat the specimen within the reaction vessel, the material must be of an electrically insulating nature. The electrically insulating nature should also be available not only at room temperature and pressure conditions but also at the extreme of the range mentioned. In combination with electrical nonconducting and thermal insulating properties, the material when undergoing such extreme pressures, and in compressing, should have certain characteristics which prevent the material itself from spalling and otherwise failing. The material should be smoothly deformable with minimum friction consistent with being retainable in the pressure chamber without danger of blowout.

It has been discovered that the prior materials mentioned are not the optimum materials in high pressure high temperature reaction vessels, simply because it is evident that the force load on the punches must be very high to develop a given pressure within the specimen in the vessel, and again that the pressure transmitting characteristics are considerably less than desired or substantially less than hydrostatic. In combination, these characteristics lead to uneven pressure distribution in the sample or object, a requirement of more force load and load application over a longer period of time. In commercial production of diamonds, for example, the deterrent features furthermore become economical disadvantages.

Accordingly, it is an object of this invention to provide

an improved reaction vessel to be subjected to high pressures and temperatures.

It is a further object of this invention to provide an improved reaction vessel with greater hydrostatic transmission properties.

It is another object of this invention to provide an improved reaction vessel with reduced frictional opposition under large compressive forces.

It is another object of this invention to provide a reaction vessel utilizing less compressive force to develop a given pressure within the object which it contained.

Briefly described this invention in one form includes a reaction vessel of a talc material or composition which, under high pressure and high temperature conditions, acts as an improved hydrostatic transmission material or provides a reduction in press load necessary to provide a given pressure within the vessel.

These and various other objects, features and advantages of this invention will be better understood from the following description taken in connection with the accompanying drawing in which;

FIG. 1 is a partial and cross sectional elevational view of a reaction vessel positioned within an exemplary high pressure high temperature apparatus;

FIG. 2 is a sectional view of the reaction vessel of FIG. 1; and

FIG. 3 is a sectional view of a modified reaction vessel.

Referring now to FIG. 1, there is illustrated an exemplary form of a high pressure high temperature apparatus in accordance with the teachings of the aforementioned copending application Serial No. 707,432. The high pressure high temperature apparatus **10** generally includes a pair of punches **11** and **11'** and a die member **12** having an aperture therethrough and positioned concentrically with punches **11** and **11'**. Gasket assemblies **13** and **13'** are positioned upon each punch member to engage the surfaces of the punch and the aperture of the die. An end cap assembly **14** is thereafter positioned upon each punch face, and the combination of punches and dies together with the gasket assemblies and end caps define a volume or chamber in which the reaction vessel **15** is positioned. Reaction vessel **15** contains a specimen material **16** to be subjected to high pressure high temperature conditions, and where resistance heating is employed, a pair of conductors **17** and **17'** are connected to a source of power, not shown, and to punch **11** and **11'** to provide a flow of current from the punches through the specimen to be heated. Reaction vessel **15**, for purposes of this invention, includes in one form, a thermally insulating and electrically non-conductive cylinder **18** and, where desirable, a pair of metallic disk members **19** which are electrically conductive to provide a current path through the specimen.

Considerable effort has been expended in attempting to discover an improved material for the cylinder **18** which would have, and maintain, thermally insulating and electrically nonconductive properties at high pressures and high temperatures, and also have reduced spalling characteristics. More importantly a material is needed which will transmit pressures from the punches to the specimen material substantially hydrostatically in an even manner, and with reduced punch load. High punch loading is subjecting the apparatus to more deteriorating forces than is necessary to develop a given pressure in the specimen. Ordinarily, such material may not be merely chosen from a listing of a given material simply because knowledge of the reaction which some materials undergo at high pressures and high temperatures is relatively unknown, and that what may be, for example, an ordinarily soft material, may become extremely hard at high pressures and minimize any transmission of pressure.

Furthermore, materials may become thermally non-insulating at high temperatures and also may become electrically conductive at high temperatures of vice versa. In this respect, innumerable materials when subjected to high pressures and high temperatures have actually prevented a given high pressure from being reached within the specimen in the reaction vessel and yet maximum force or load has been applied to the punches. Furthermore, many given materials will ordinarily decompose at about 1200–1300° C. and it is obvious that such condition could not be permitted in high pressure high temperature apparatus. However, it is known that, with some materials which will ordinarily decompose at these temperatures, when subjected to very high pressures and high temperatures, they do not decompose.

It has been discovered that a talc material or composition substantially overcomes the inherent disadvantages of pyrophyllite, catlinite and other ceramics by meeting the desired requirements as described. A talc composition or material as described in this invention is composed of an unfired hydrated magnesium silicate which has the general chemical composition of $3MgO \cdot 4SiO_2 \cdot H_2O$. This material is a very slippery type of stone which is incapable of supporting high shear stresses. When an insulating cylinder or reaction vessel of pyrophyllite or catlinite is employed, a part of the force on the punch which is required to produce a certain pressure in the reaction vessel must overcome the frictional opposition to flow or compression in the reaction vessel material. This frictional resistance occurs not only between the reaction vessel and its associated pressure resisting walls, but also within the vessel material itself. Talc has been discovered to reduce both the frictional opposition and the normal press load. For example, the barium electrical resistance transition, which occurs at about 77,400 atmospheres is now obtained in prophyllite when a 182-ton load is applied to the punches. However, when employing a talc reaction vessel in the same apparatus only a 144-ton load is required to secure the same transition. This indicates a reduced load of 38 tons on the high pressure apparatus. As a result of using talc material for a reaction vessel, it has also been discovered that the material is a better hydrostatic transmitting material than the pyrophyllite, catlinite and other ceramics. Evidence of this has been shown because of the more even pressure distribution evidenced in the samples after subjecting to high pressures and high temperatures, and increase in diamond yield compared to identical pyrophyllite vessels. The talc material retains its thermally insulating and electrical nonconducting properties at the extremes of high pressures and temperatures and without undergoing any detrimental phase changes or decomposition. Decomposition is very important because pyrophyllite or catlinite vessels have been discovered to enter into, for example, diamond growth reaction, in a detrimental manner to affect yield, quality, and growth conditions generally.

As one example, the talc material employed for the reaction vessel is that of natural talc in solid form which is machined or otherwisee formed into the desired configuration. The talc cylinder may be in some instances heat treated in a somewhat minor form to improve its general characteristics, but must not be of the hard fired variety because a hard fired talc will not provide the optimum advantages of non-hard fired talc.

The talc material for the reaction vessel may also be provided in alternate ways. The vagaries of nature may sometimes produce talc which is of a non-uniform composition and which may contain rather large interstices or voids not apparent in the reaction vessel in finished form. Such voids and interstices must be explicitly avoided in high pressure high temperature apparatus in order to maintain a reaction vessel in integral form and to provide uniform pressure in the specimen to be subjected to high pressure high temperature conditions. Natural talc also may contain impurities such as minerals other than talc which mask the free deforming characteristics of talc.

Accordingly, when natural talc is under suspicion of having voids and interstices or where the talc may be of a non-uniform composition, a reaction vessel in accordance with the teachings of this invention may be composed of a talc composition or material.

In one form talc composition includes reducing or obtaining talc in small particle or powder form and pressing to desired shape. Good results have been obtained in compacting talc at about 3–4 tons per square inch to form a reaction vessel as illustrated in FIGS. 2 and 3. Such a talc composition or material may be acquired through reducing or obtaining talc in particle or powder form and thereafter mixing with a suitable binder, plasticizing agents, clays, etc., and molding the finished object. In this respect many of the lower grades of talc, such as steatite, soap stone, etc., may be employed where they are either reduced or obtained in small particle size, mixed with various plasticizers, bonding agents, etc., and thereafter molded or compacted to the desired configuration. In addition to the economic advantages of employing different quality or lower grade talc and the increased availability of such materials for the reaction vessel, the bonding and molding of a given volume provide an opportunity depending on the characteristics of the bonding and plasticizing mediums to predetermine and improve certain conditions, as compression characteristics, contributing to uniform hydrostatic pressure transmitting characteristics. In other words, the binder itself may char or powder or undergo certain deformation characteritsics which increase the overall characteristics of the reaction vessel. The following are exemplary of various talc composition reaction vessels in accordance with the teachings of this invention. Catalysts employed were taken from the metals of group VIII of the periodic table of elements, chromium, manganese, and tantalum. Carbonaceous material was graphite of spectroscopic purity.

Example 1

The reaction vessel as illustrated in FIG. 2 was machined from a block of natural talc and a carbonaceous material in the form of graphite of spectroscopic purity, and a catalyst, nickel, placed therein. After subjection to 76,000 atmospheres and 1450° C., a large yield of diamonds was obtained.

Example 2

Example 1 was repeated employing iron as the catalyst with corresponding pressure of 80,000 atmospheres and a temperature of 1500° C. Similar results were achieved.

Example 3

A reaction vessel (FIG. 2) was machined from a pressed block of powdered talc utilizing a phosphate binder and commercially obtained from the American Lava Corporation, Code 681F. Nickel was used as a catalyst and spectroscopic purity graphite as a carbonaceous material. Pressure was about 75,000 atmospheres and temperature about 1500° C. A large yield of diamonds was obtained.

Example 4

The reaction vessel of Example 3 was employed containing spectroscopic purity graphite and a nickel chrome alloy catalyst, 95% and 5% by weight of catalyst respectively. Pressure was about 75,000 atmospheres and temperature about 1400° C. Similar results were obtained.

Example 5

A number of reaction vessels were made from talc powder with a binder of polyvinyl alcohol in the range of 10–17% by weight of the vessel, and a compacting force of about 4 tons per square inch. These vessels were heat treated at various temperatures up to about 900° C., and for periods of time including 45 minutes. In all instances diamond growth was obtained therein with larger yields than with pyrophyllite.

Reaction vessels were made from talc from various geographical locations and with numerous binders. All vessels employed, produced a greater yield of diamond at the same pressure temperature conditions than has been obtained from catlinite or pyrophyllite vessels. Further examples of binder materials include cellulose starch, oxidized linseed oil, etc., of about 10% by weight of the vessel.

The exemplary reaction vessel illustrated, for example, in FIG. 2 includes a cylindrical element **18** together with a pair of electrically conductive disks **19**. These electrically conductive disks are utilized in conjunction with the illustration as given in FIG. 1 to provide a flow of electrical current either through specimen **16** itself where the specimen is electrically conductive, or through a suitable resistance heater positioned within the specimen. In some instances, closure disks **19** may be suitably attached or bonded to cylinder **18** to provide a unitary structure for a reaction vessel and to effectively seal the specimen within the vessel. The closure disks may not be necessary where no electrical heating is employed or where the specimen material will not affect the punch faces. In a diamond reaction such a disk not only serves to protect the punches and convey electric current, but also may be of a catalyst material for a given reaction.

In FIG. 3 there is illustrated a modification of the end disks where either one or both of the disks **19** are recessed within the ends of cylinder **18**.

A suitable talc reaction vessel may be employed in various other configurations, the particular configuration being relatively unimportant and of no great significance in order to obtain the advantages as described. Therefore, the vessel may be best described as a receptacle member adapted to contain, in a suitable opening or aperture, a specimen material to be subjected to high pressures and high temperatures where the receptacle member is positioned within a high pressure apparatus and undergoes considerable deformation or compression. Accordingly, the reaction vessel may be, externally, of any geometrical or irregular configuration, specifically adapted to fit within a particular volume or chamber of a high pressure apparatus. For example, the reaction vessel may be of the configuration illustrated in copending application Serial No. 855,867, Bundy, filed November 27, 1959, and assigned to the same assignee as the copending application or, for example, the reaction vessel may be of a configuration in the form of a tetrahedral to fit the apparatus described in U.S. Patent 2,918,699, Hall.

It is thus understood by those skilled in the art that the objects, features and advantages of this invention have been obtained by the use, broadly, of a non-hard fired talc, which includes talc materials or compositions, for a reaction vessel where a significant amount of talc improves the compression characteristics of the vessel. The vessel may be machined or otherwise formed from talc in the natural state or a talc material is suitably divided into particle or powder form and compressed or mixed with a bonding agent and then molded or compressed to the desired configuration. The amount and/or kind of binder material may be employed to predetermine deformation characteristics. Talc is the primary and key material in such a reaction vessel, but good results are evidenced when as much as about 50% by weight or parts is an additional material. Where the reaction vessel is to be subjected to high temperatures in addition to high pressures, and where electrical resistance heating is employed, electrical connection means are employed, for example, in the form of disks or other well known electrical devices or connections.

While other modifications of this invention and variations of apparatus have not been described, the invention is intended to include all such as may be embraced in the following claims.

What I claim as new and desire to secure by Letters Patent of the United States is:

1. A high pressure high temperature reaction vessel which is subjected to compression and deformation comprising, a receptacle member, said receptacle member having an aperture therein adapted to contain a specimen material to be subjected to high pressures, said receptacle member characterized by containing a significant amount of non-hard fired talc which improves the compression characteristics of the vessel.

2. A high pressure high temperature reaction vessel which is subjected to compression and deformation comprising, a receptacle member having an aperture therein adapted to contain a specimen material to be subjected to high temperatures and pressures, said receptacle member consisting essentially of a non-hard fired talc composition.

3. In a high pressure high temperature reaction vessel which is subjected to compression and deformation the combination comprising, a receptacle member having an opening therein adapted to contain a specimen material to be subjected to high pressure high temperature conditions, said receptacle member comprising a compressed mixture of a talc material and a bonding medium, said talc material containing a significant quantity of talc to improve the hydrostatic pressure transmission characteristics of the vessel.

4. In a high pressure high temperature reaction vessel which is subjected to compression and deformation, the combination comprising a receptacle member having an aperture therein adapted to contain a specimen material to be subjected to high pressure and high temperature, said receptacle member characterized by being formed from solid natural talc.

5. In a high pressure apparatus reaction vessel which is subjected to compression by a die member, the combination comprising, a talc cylinder, said cylinder having a central aperture adapted to contain a specimen, and an electrically conductive closure for each end of said cylinder to contain the specimen therein.

6. A high pressure high temperature reaction vessel which is subjected to compression and deformation comprising, a receptacle member, said receptacle member having an aperture therein adapted to contain a specimen material to be subjected to high pressures, said receptacle member characterized by containing at least about 50% non-hard fired talc to improve its compression characteristics in a high pressure high temperature apparatus, and electrical connection means operatively connected to said receptacle member to provide electrical resistance heating for said specimen material.

7. In a high pressure high temperature apparatus reaction vessel the combination comprising, a non-hard fired talc cylinder, said cylinder having a central aperture adapted to contain a specimen, and an electrically conductive disk closing each end of said aperture to contain the specimen therein.

8. In a high pressure high temperature apparatus reaction vessel which is subjected to compression by a die member, the combination comprising an unfired talc cylinder, said cylinder having a central aperture adapted to contain an electrically conductive specimen, and an electrically conductive disk coaxially positioned with each end of said cylinder to seal said specimen therebetween, at least one of said disks having a smaller diameter than the outside diameter of the cylinder.

9. In a high pressure high temperature apparatus reaction vessel which is subjected to compression by a die member, the combination comprising an unfired talc cylinder, said cylinder having a central aperture adapted to contain an electrically conductive specimen, an elec-

trically conductive closure for one end of said cylinder, the opposite cylinder end being recessed adjacent to periphery of said aperture, and a second electrically conductive closure positioned in said recess to close the other end of said aperture and seal the said specimen therebetween.

10. The invention as recited in claim 3 wherein said bonding medium is taken from the group consisting of polyvinyl alcohol, clay, cellulose starch, oxidized linseed oil and phosphate.

11. The invention as recited in claim 10 wherein said bonding medium is polyvinyl alcohol in a range of about 10 to 17% by weight of said vessel.

References Cited in the file of this patent

UNITED STATES PATENTS

2,195,297	Engle	Mar. 26, 1940
2,694,922	Vilella	Nov. 23, 1954
2,941,250	Hall	June 21, 1960

OTHER REFERENCES

Von Baur et al.: "Uber das Problem des Diamasten," Zeitschrift für Anorganische Chemie, vol. 92 (1915), pages 313–327.

Bridgman: "Resistance of 72 Elements, Alloys and Compounds to 100,000 kg./cm.^{2}," American Academy of Arts and Sciences, vol. 81 #2 (1952), pages 169–184.