United States Patent [19]

Kuratomi

[11] **3,727,028**

[45] **Apr. 10, 1973**

[54] **ULTRA HIGH PRESSURE-TEMPERATURE APPARATUS**

[76] Inventor: **Tatsuo Kuratomi,** 2-18, 4-chome, Hamatake, Chigasaki-shi, Japan

[22] Filed: **Dec. 3, 1971**

[21] Appl. No.: **204,414**

Related U.S. Application Data

[63] Continuation-in-part of Ser. No. 18,143, March 10, 1970, Pat. No. 3,647,331.

[30] **Foreign Application Priority Data**

Mar. 10, 1969 Japan 44/17548

[52] **U.S. Cl.**.....**219/149,** 425/77
[51] **Int. Cl.**.....**B21j 1/06,** B30b 11/32
[58] **Field of Search**.....219/149, 152, 50; 425/77

[56] **References Cited**

UNITED STATES PATENTS

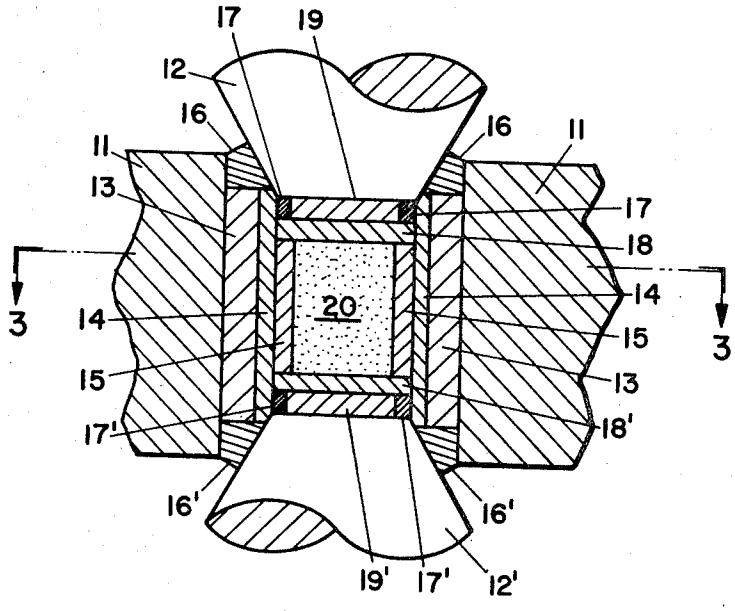
3,546,413 12/1970 Ishizuka.....219/149 X

Primary Examiner—R. F. Staubly
Attorney—David E. Dougherty et al.

[57] **ABSTRACT**

An improvement in ultra high pressure-temperature apparatus in which two concentric, adiabatic, electrically nonconductive, thermostable cylinders, the inner cylinder comprising beryllium oxide, are utilized to protect the outer portions of the apparatus from the pressures and temperatures generated within the innermost parts of the apparatus.

4 Claims, 3 Drawing Figures



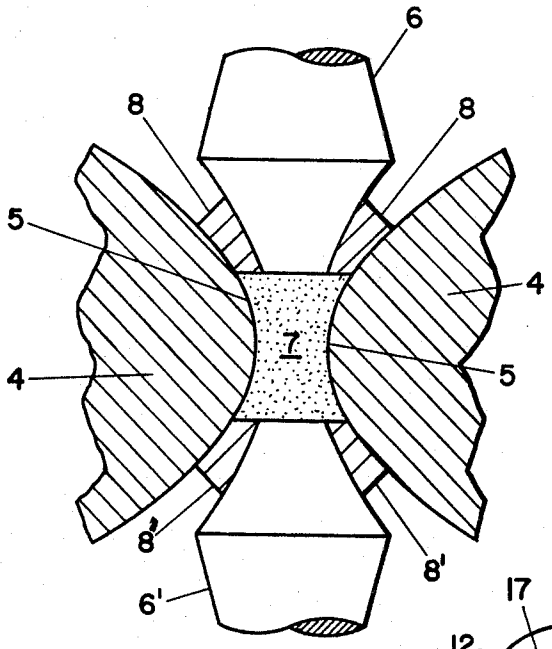


FIG. 1
PRIOR ART

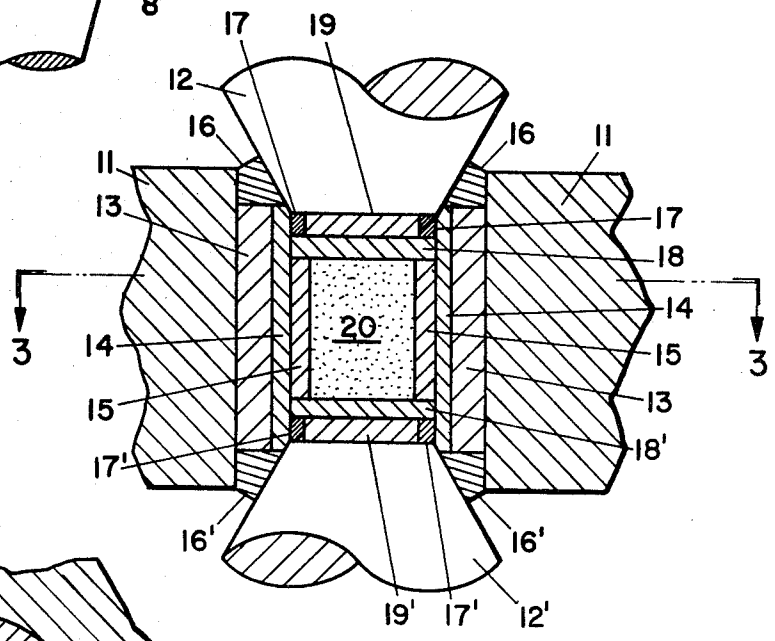


FIG. 2

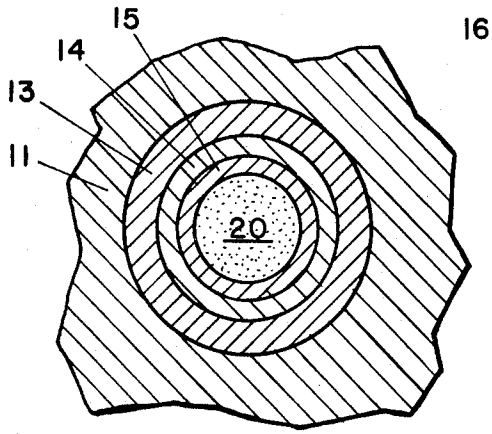


FIG. 3

INVENTOR.
TATSUO KURATOMI
BY Raymond W. Green
David E. Dougherty

ULTRA HIGH PRESSURE-TEMPERATURE APPARATUS

CROSS-REFERENCE TO RELATED APPLICATION

This is a continuation-in-part of application, Ser. No. 18,143 filed Mar. 10, 1970, now U.S. Pat. No. 3,647,331.

BACKGROUND OF THE INVENTION

This invention relates to ultra high pressure-temperature apparatus.

Ultra high pressure-temperature apparatus capable of producing and maintaining pressures of the order of 40,000 to 100,000 atmospheres and temperatures of the order of 1000 to 2500°C is desirable to effect, control and study reactions occurring under these conditions. The reactions of various materials subjected to such pressures and temperatures can be employed for research study purposes or to obtain physical and chemical changes which desirably alter the characteristics of the materials. For example, new compounds are known to be formed by subjecting old materials to very high pressures. Ultra high pressure-temperature apparatus is also useful for studying the changes in phase of various materials which occur at very high pressures, or for studying the compressibility or electrical, optical or magnetic properties of various materials. An example of a change occurring under these conditions which is of considerable practical utility is the catalytic conversion of non-diamond carbon to the diamond form.

Prior art high pressure-temperature apparatus of this type comprises generally (1) a pair of opposed punch assemblies, each of the punch assemblies terminating in a tapered electrically conductive piston; (2) means for exerting pressure on the punch assemblies, whereby an electrically conductive object positioned between the opposed pistons can be subjected to high pressure; (3) a lateral pressure resisting annulus, positioned between the opposed pistons and provided with a substantially central aperture circumferentially surrounding the object to be subjected to high pressure, the annulus having a pressure resisting inner wall surface: (4) means for passing electrical current through the pistons and the object to be subjected to high pressure, whereby to produce a high temperature within the object simultaneously with the high pressure; and (5) thermal and electrical insulating gaskets positioned in the aperture of the lateral pressure resisting annulus and circumferentially surrounding the tapered pistons.

A typical prior art apparatus is illustrated in FIG. 1, wherein a lateral pressure resisting annulus 4 having a pressure resisting inner wall surface 5 is shown surrounding object 7. Opposed tapered electrically conductive pistons 6 and 6′ are urged toward object 7 by conventional means (not shown) to produce high pressure in object 7. Thermally and electrically insulating gaskets 8 and 8′ are provided to separate pistons 6 and 6′ from annulus 4, but not from object 7. Electricity is then passed through piston 6, object 7 and piston 6′, thus heating object 7 by internal resistance heating. Annulus 4 is strengthened and reinforced with several layers of steel rings.

Apparatus of this type is frequently capable of pressure in excess of 80,000 atmospheres and temperatures in excess of 2000°C, and further details of its construction and operation are described in the prior art, for example U.S. Pat. No. 2,941,248. Such apparatus is subject to the disadvantage, however, that the pressure-resisting annulus 4 and pistons 6 and 6′ bear much of the pressure and heat of the reaction within object 7, with the result that these parts are subject to fracture and frequently need replacement. Furthermore, it is difficult to enlarge any given annulus 4 to accommodate a larger object 7, inasmuch as the inner walls 5 of cylinder 4 are tapered toward its center.

SUMMARY OF THE INVENTION

It is, therefore, an object of this invention to provide a new and improved high pressure-temperature apparatus of the type described which overcomes the above-noted problems of the prior art. These and other objects are achieved by providing the pressure resisting inner wall surface of the pressure resisting annulus with a vertical cylindrical shape, and in combination therewith, two concentric, heat-insulating, electrically nonconductive, thermostable hollow cylinders positioned within the aperture of the lateral pressure resisting annulus and circumferentially surrounding the object to be subjected to high pressure (e.g., a reaction chamber for the conversion of non-diamond carbonaceous material to diamond). The outer portions of the apparatus are thus protected from the pressures and temperatures within the innermost parts of the apparatus.

In application Ser. No. 18,143, the first (inner) heat-insulating electrically nonconductive, thermostable hollow cylinder is specified as consisting essentially of thorium oxide, zirconium oxide, hafnium oxide, or mixtures thereof. In the present invention this cylinder comprises beryllium oxide, and in particular, consists essentially of beryllium oxide or mixtures thereof with thorium oxide, zirconium oxide, hafnium oxide, or mixtures thereof. Further details and preferred embodiments are indicated below.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a sectional schematic diagram of the central portion of conventional high pressure-temperature apparatus as described above.

FIG. 2 is a sectional schematic diagram of the central portion of apparatus according to the present invention.

FIG. 3 is a sectional view of the apparatus of FIG. 2, taken along line 3—3 of FIG. 2.

DETAILED DESCRIPTION

Referring now to FIGS. 2 and 3 of the drawings, the apparatus of the present invention will now be described in detail.

Lateral pressure resisting annulus 11 having a vertical cylindrical shape for its pressure resisting inner wall surface surrounds circumferentially the object 20 to be subjected to high pressure. Materials which can be used for annulus 11 include ultrahard alloys, high speed steel an die steel; these materials themselves are known in the art. Above and below object 20 are opposed punch assemblies (not shown), terminating in tapered electrically conductive pistons 12 and 12′. It should be noted that references herein to "above," "below," "lateral"

and the like are merely indicative of orientation when the apparatus is arranged with the pistons **12** and **12'** in a vertical configuration, and the apparatus need not be so arranged. This designation is convenient, however, in indicating the relative orientation of the various parts of the apparatus herein described. Materials which can be used for pistons **12** and **12'** include die steels and ultrahard alloys such as cemented tungsten carbide. One such alloy which is commercially available contains 94 percent tungsten carbide and 6 percent cobalt.

Within the aperture of lateral pressure resisting annulus **11** and circumferentially surrounding object **20** is an heat-insulating, electrically nonconductive, thermostable hollow cylinder **15**. Cylinder **15** consists essentially of beryllium oxide, or mixtures thereof with thorium oxide, zirconium oxide, hafnium oxide, or mixtures thereof. In order that temperatures of 2500°C can be generated within object **20**, the material for hollow cylinder **15** must have a melting point of at least 2600°C and be heat-insulating and electrically nonconductive. Materials with sufficiently high melting points and electrical resistivities include the following:

Material	Melting Point
Thorium Oxide	3300°C
Magnesium Oxide	2825°C
Zirconium Oxide	2700°C
Calcium Oxide	2630°C
Hafnium Oxide	2800°C
Beryllium Oxide	2530°C

Of these materials, however, calcium oxide and magnesium oxide are not preferred because of their comparatively high thermal conductivities. Calcium oxide has the further disadvantage of reacting with carbon at temperatures above 2000°C to form calcium carbide. The carbonization reaction advances rapidly and continuously, whereas the carbides of thorium and zirconium, for example, form thin filmy protective coatings on the inner face of hollow cylinder **15**, so that further carbonization does not occur.

In a preferred embodiment of this invention, there is provided a pair of heat-insulating thermostable discs **19** and **19'**, disc **19** being positioned above object **20**, between object **20** and piston **12**; and disc **19'** being positioned below object **20**, between object **20** and piston **12** . Discs **19** and **19'** are of such diameter, however, as to allow the passage of electrical current from one piston to the other piston through object **20**. This is preferably accomplished by providing (1) a pair of electrically conductive rings **17** and **17'** surrounding discs **19** and **19'**, respectively, and in electrical contact with pistons **12** and **12'**, respectively; and (2) a pair of electrically conductive discs **18** and **18'**, disc **18** being positioned above object **20**, between object **20** and disc **19**; and disc **18'** being positioned below object **20**, between object **20** and disc **19'**. Discs **18** and **18'** are simultaneously in electrical contact with rings **17** and **17'**, respectively, on the one hand, and object **20** on the other. Thus electrical current can pass from piston **12** to ring **17** to disc **18** to object **20** to disc **18'** to ring **17'** to piston **12'**, while pistons **12** and **12'** are shielded by heat-insulating thermostable discs **19** and **19'** from the heat and pressure generated in object **20**. Preferred materials for heat-insulating thermostable discs **19** and **19'** include beryllium oxide, thorium oxide, zirconium oxide, hafnium oxide and mixtures thereof.

A still more preferred embodiment of this invention includes a second heat-insulating, electrically nonconductive, thermostable hollow cylinder **14** positioned within the aperture of the lateral pressure resisting annulus **11**, and circumferentially surrounding (1) the first heat-insulating, electrically nonconductive, thermostable, hollow cylinder **15**; (2) the heat-insulating, thermostable discs **19** and **19'**; (3) the electrically conductive rings **17** and **17'**; (4) the electrically conductive discs **18** and **18'**; and (5) the object **20** to be subjected to high pressure. Preferred materials for heat-insulating electrically nonconductive, thermostable, hollow cylinder **14** include zirconium oxide, pyrophyllite, and mixtures thereof.

A still more preferred embodiment of this invention includes a metallic hollow cylinder **13**, positioned within the aperture of the lateral pressure resisting annulus **11** and circumferentially surrounding the second heat-insulating, electrically nonconductive, thermostable, hollow cylinder **14**. Cylinder **13** should be of a metallic material of great hardness and tenacity so as to reduce the high pressures generated within the apparatus (by urging pistons **12** and **12'** towards object **20**), and thereby subject annulus **11** to less stress than would otherwise be the case. Thus annulus **11** is able to maintain its hardness and tenacity without being melted or worsened in quality by the full heat and pressures generated within object **20**. Preferred materials for cylinder **13** include ultrahard alloys, high speed steel, die steel and ceramic-metallic (cermet) alloys.

By the use of cylindrical shapes for the pressure resisting inner wall surface of pressure resisting annulus **11** and for cylinders **13**, **14** and !5, these innermost parts which bear the greatest heat and pressure of the apparatus can easily be replaced, if need be. The cylindrical shape also aids in the transmission of pressures from pistons **12** and **12'** to object **20**.

Thermal and electrical insulating gaskets **16** and **16'**, preferably of pyrophyllite, surround pistons **12** and **12'**, respectively, to complete the portion of the apparatus shown.

I claim:

1. In a high pressure-temperature apparatus for subjecting an object to high pressure, comprising:
   a. a pair of opposed, tapered, electrically conductive pistons;
   b. a lateral, pressure-resisting annulus, positioned between the opposed pistons and coaxial therewith, provided with a substantially central aperture circumferentially surrounding the object to be subjected to high pressure, the annulus having a pressure-resisting inner wall surface; and
   c. a thermal and electrical insulating gasket positioned in the aperture of the lateral pressure-resisting annulus and circumferentially surrounding the tapered pistons, the invention which comprises the provision of a vertical cylindrical shape for the pressure-resisting inner wall surface of the pressure-resisting annulus, and in combination therewith
      1. a first heat-insulating, electrically nonconductive, thermostable, hollow cylinder, positioned within the aperture of the lateral, pressure-resisting annulus, and circumferentially surrounding the object to be subjected to high pressure, said first heat-insulating, electrically noncon-

ductive, thermostable, hollow cylinder consisting essentially of beryllium oxide, or mixtures thereof with thorium oxide, zirconium oxide, hafnium oxide, or mixtures thereof;

2. a pair of heat-insulating, thermostable discs, one disc being positioned above and one disc being positioned below the object to be subjected to high pressure, between said object and the opposed pistons; said discs being of such diameter as to allow the passage of electrical current from one piston to the other piston through the object to be subjected to high pressure, said heat-insulating, thermostable discs consisting essentially of beryllium oxide, thorium oxide, zirconium oxide, hafnium oxide, or mixtures thereof;

3. a pair of electrically conductive rings each ring surrounding one of the heat-insulating thermostable discs and being in electrical contact with one of the opposed pistons;

4. a pair of electrically conductive discs, one electrically conductive disc being positioned above and one electrically conductive disc being positioned below the object to be subjected to high pressure, between said object and the heat-insulating thermostable discs, each of said electrically conductive discs being electrically in contact with one of the electrically conductive rings and with the object to be subjected to high pressure;

5. a second heat-insulating, electrically nonconductive, thermostable, hollow cylinder, positioned within the aperture of the lateral pressure resisting annulus and circumferentially surrounding

a. the first heat-insulating, electrically nonconductive, thermostable, hollow cylinder;

b. the heat-insulating, thermostable discs;

c. the electrically conductive rings;

d. the electrically conductive discs; and

e. the object to be subjected to high pressure; said second heat-insulating, electrically nonconductive, thermostable, hollow cylinder consisting essentially of zirconium oxide, pyrophyllite, or mixtures thereof; and

6. a metallic hollow cylinder, positioned within the aperture of the lateral pressure-resisting annulus and circumferentially surrounding the second heat-insulating, electrically nonconductive, thermostable, hollow cylinder.

2. The invention of claim 1 wherein the first heat-insulating, electrically nonconductive, thermostable hollow cylinder consists essentially of beryllium oxide.

3. The invention of claim 1 wherein the heat-insulating, thermostable discs consist essentially of beryllium oxide.

4. The invention of claim 1 wherein the metallic hollow cylinder is provided with flat upper and lower surfaces.

* * * * *