HIGH PRESSURE CLAMP CELL FOR DILUTION REFRIGERATOR

GENSHIRO FUJII, YASUKAGE ODA, AND HIROSHI NAGANO

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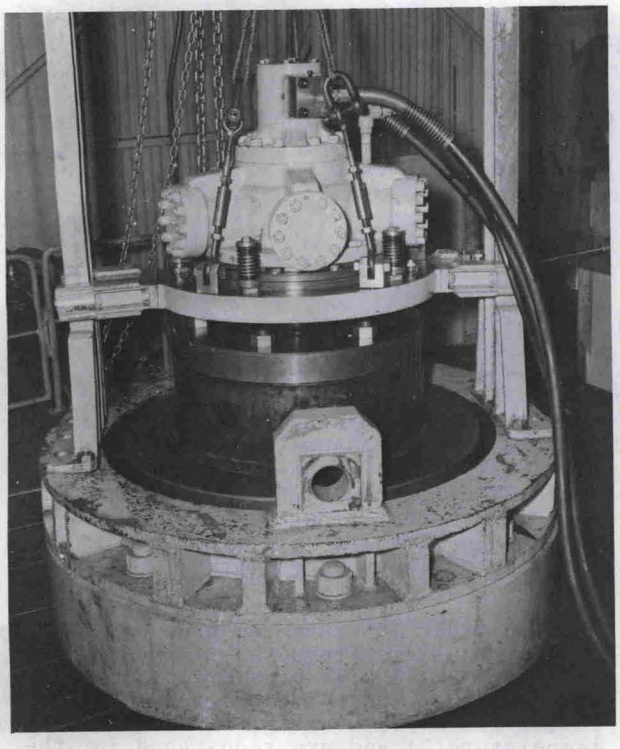

Photo. 2 High Pressure Oil Vessel

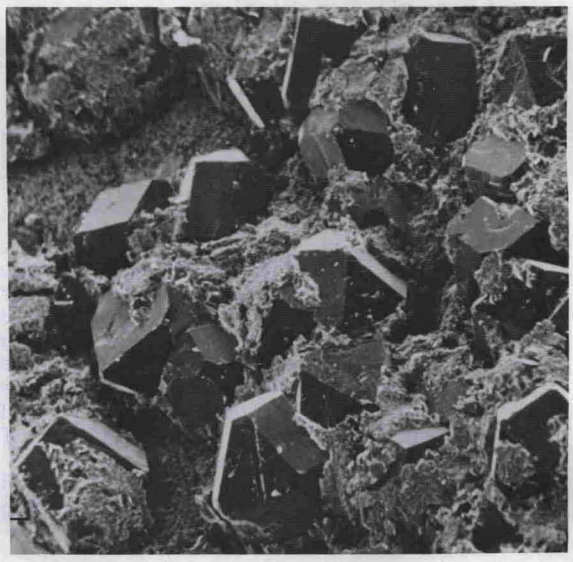

Photo.3 A Scanning Electron Micrograph of the As-grown Diamond Crystals

Table 1 Practically Available Pressure and Volume Relations for These Apparatus

	HO-type				OH-type	SO-type			
edge length a cm	0.5	10	15	25	15	35	45	55	65
anvil enveloped space V_o cm^3	0.059	0.47	1.59	7.366	3.375	20.21	42.96	78.43	129.5
pressure medium volume $(1.3)^3 V_o$ cm^3	0.129	0.53	1.04	16.18	7.425	44.40	94.38	172.31	284.4
achieved practical pressure P_1 kb	250	180	130	100	100	90	75	65	55

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GENSHIRO FUJII, YASUKAGE ODA, AND HIROSHI NAGANO

The Institute for Solid State Physics, University of Tokyo
Roppongi 7, Minato-ku, Tokyo 106, Japan

Two small high pressure clamp cells using at about thirty millikelvin region are devised. These consist of one couple of Bridgman-anvils and two flanges. One is used for a d.c. electric resistance measurement and another is used for an a.c. magnetic susceptibility measurement under high pressure. The sample is placed between two small anvils and a hydraulic press pressurizes it at room temperature. To maintain the pressure applied to the sample, the anvils are clamped by three bolts. The highest pressure attained by these clamp cells is fifty kbar. The clamped high pressure cell is attached to the underside of the mixing chamber of a helium 3/helium 4 dilution refrigerator and is cooled down to thirty millikelvin. Several design points of the clamp cell and its cooling process are described.

1. Introduction

High pressure experiments at the liqid helium temperature region have been conducted and several pressure-induced superconductors have been discovered(1) in recent years. As there are the interests that whether there is still any other pressure-induced superconductor or not, whether the superconducting properties are destroyed by the application of very high pressure or not(2) and what is the pressure dependence of the superconducting transition temperature of low Tc superconductors, high pressure experiments at further low temperature region are required.

In order to cool the high pressure apparatus down to the several tens millikelvin region, the following technical problems have to be solved.

First: The heat leakage from the outside have to be reduced.

Second: The small size apparatus must be designed.

Third: Whether hydrostaticity of the pressure transmitting medium exists at low temperature in the case of the small apparatus.

In recent years, several papers of high pressure experiments at temperatures lower than 1 kelvin are reported. Those are as follows. Levy and Olsen(3) cooled a clamped apparatus of 21 kbars using a helium 3 cryostat. Brandt and Ginzburg(4) tried to cool a small ice bomb cell down to 80 millikelvin by an adiabatic demagnetization method and measured the superconducting transition temperature of Cd under 27 kbars. Benoit et al.(5) tried to cool a small clamp cell of 10 kbars down to 16 millikelvin also by the adiabatic demagnetization method and studied a nuclear orientation experiment. Stritzker et al.(6) reported to cool a high pressure cell using a dilution refrigerator down to 70 millikelvin for studying the pressure effects on the superconducting materials.

In order to study the pressure effects on low transition temperature superconductors, two small high pressure clamp cells are devised. These are originally designed for the experiments at the liquid helium temperature(7) and are redesigned for the experiments at temperatures of the several tens millikelvin region.

In this paper, the details of two clamp cells, the cooling method, the pressure calibrations and the temperature distribution of the cell during the cooling process are described.

2. Small high pressure clamp cell

Among several types of high pressure apparatus, it is best to use the clamp cell for attaining temperatures lower than 100 millikelvin, because of its small heat capacity and its small heat leakage during the experiment. In this method, the sample is placed between two small anvils and a hydraulic press pressurizes it at the room temperature. To maintain the pressure applied to the sample, the anvils are clamped by the screw mechanism. In Figs. 1 and 2, two clamp cells are shown which are originally designed at liquid helium temperatures(7), and are redesigned for this purpose. The sample is placed between the anvils and these anvils are held between two flanges and these flanges are tightened by three bolts. As two flanges are used in this type of clamp apparatus, it is very easy to replace the sample, to avoid the twisting of the sample and the breaking of lead wires, and it is convenient to fix the measuring coils for an a.c. magnetic susceptibility which is shown in Fig. 2. For attaining the accurate and reproducible pressure, several torque wrenches are used.

The sample assembly for a D-I type cell is shown in Fig. 3. The pyrophyllite ring is heated to 650°C for 30 minutes in order to increase its hardness, and then is fixed with an insulating cement to the face of the anvil. A sample is placed in the talc disc which is a pressure

transmitting medium. The talc disc is made from pressed talc powder. The talc is much more plastic than the pyrophyllite and thus produces sufficient uniformity in the generated pressure.

The materials used in this apparatus are a copper-beryllium alloy and an austenitic stainless steel, as these materials have enough strength and ductility even at very low temperatures. The D-I cell shown in Fig. 1 is used for a d.c. electrical resistance measurement under high pressure. The dimensions of the flange are 30 mm in diameter and 10 mm in thickness. As for the guide of the anvil, a copper cylinder is used. The total weight of this cell is 420 gr.

The D-II cell shown in Fig. 2 is used for the a.c. magnetic susceptibility measurement. For the a.c. magnetic susceptibility measurement, the primary and the secondary coils are both divided in three parts. The coils are wound with an insulated copper wire (0.08 mm-diam). The inner coil is the primary which is totally 900 turns in the same direction. The outer coil is the secondary which is totally 2000 turns. The turn of the second (middle) part of the coil is reverse in its direction from the first (upper) and the third (lower) part and the induced voltages from each parts of the coil are compensated each others. Detection of the superconducting transition by the magnetic method does not need to use the lead wires which directly contact to the sample. The total weight of the clamp cell including the coils is 260 gr.

3. Cooling process of clamp cell

A helium 3/helium 4 dilution refrigerator has been made to cool the high pressure clamp cells. Fig. 4 shows the main part of the helium 3/helium 4 dilution refrigerator. The refrigeration process of the helium 3/helium 4 dilution refrigerator is as follows. The concentrated helium 3 stream is cooled gradually by liquid He (4.2 K) and is further cooled by liquid He which vaporizes under reduced pressure (1.2 K). Then the helium 3 is liquefied and further cooled by the still and the heat exchangers. This cooled concentrated helium 3 mixes with liquid helium 4 in the mixing chamber and absorbs the heat of mixing (heat of dilution). This cooled helium 3/helium 4 mixture (dilute phase) returns to the heat exchangers and cools the incoming concentrated helium 3. As the vapor pressure of helium 3 is different from that of helium 4, helium 3 is selectively vaporized in the still. This vaporized helium 3 is pumped and recirculated. Our dilution refrigerator is able to attain about ten millikelvin. Its refrigeration power at 100 millikelvin is about 50 μW and its circulation rate is 5×10^{-5} mol helium 3/sec. The clamped high pressure cell is attached to the underside of the mixing chamber by a screw and fine copper wires are used between the guide of the anvil and the mixing chamber for good thermal conduction.

4. Pressure calibration

The generated pressure at room temperatures is calibrated using several fixed points which are the phase transition of Bi I-II, III-V, Tl I-II and Sn I-II, by means of the d.c. electrical resistance method. Fig. 5 shows the pressure-load calibration curve for the small Bridgman anvil (4.0 mm diameter of face). The pressure is based on the N.B.S. Symposium Scale of 1968(8). When the high pressure clamp cell is cooled down to low temperatures, the pressure in the cell is reduced by the effect of the differential thermal contraction of the materials used. Therefore, the clamped pressure has to be calibrated at low temperatures.

A pressure manometer at low temperatures is readily available in the form of a number of superconductors whose transition temperatures (Tc) are sufficiently sensitive to the change of pressure(9). The pressure calibration of the D-I cell and the D-II cell are done using the tin manometer(10). Fig. 6 shows the pressure-load calibration curve for the anvil with a 4 mm and a 5 mm diameter face. The tin sample of 99.999 % purity is rolled to a thickness of 0.03 mm and is annealed at 150°C, for 2 hours. The superconducting transition temperature is taken from the midpoint of the transition. As the transitions become sharply, the homogeneity of generated pressure is considered to be fairly good. From Figs. 5 and 6, a pressure loss of about 40 % is estimated.

5. Temperature distribution of cell

At temperatures lower than 100 millikelvin, the Kapitza's thermal boundary resistance increases and it is very difficult to cool the high pressure cell by heat conduction and thermal contacts.

The temperature distribution of the D-I apparatus is measured by the Speer carbon resistors. These are 220 Ω, Grade 1002, 1/2W which are calibrated in Cerium Magnesium Nitrate (CMN) magnetic temperature T*.

The resistance thermometers are attached to several parts of the apparatus, the underside of the mixing chamber, the upper flange and the lower flange. These temperatures are compared with the carbon resistance thermometers which are placed inside the mixing chamber. Fig. 7 shows the results.

Because of the Kapitza's thermal resistance and the heat leakage from the lead wires, the temperature of the lower flange is the highest, and the temperature difference between the lower flange and the outside surface of the mixing chamber is about 10 millikelvin. The temperature of this D-I cell reaches 30 millikelvin, however, the sample is heated up by the platinum lead wires if there is excessive currency running. For example, at a temperature of 50 millikelvin, the critical current which maintains thermal equilibrium is 15 μA.

In the case of the D-II cell, the mutual inductance coils are put outside the anvils instead of the copper guide, so that it is difficult to cool by heat conduction, and the eddy current, which is induced by the a.c. magnetic susceptibility

measurement, heats up the whole cell and the lowest temperature attained by this cell is 80 millikelvin. An a.c. current of 70 Hz is applied to the primary coil. The critical current is 100 μA at the temperature of 80 millikelvin which maintains the thermal equilibrium.

6. Pressure dependence of superconducting transition temperature in Cd and Ag_2F

The transition temperature of most superconductors are reduced by increasing the pressure and a linear extrapolation of the low pressure data on dTc/dP might lead one to suppose that the superconducting transition temperature would become zero under high pressure. Brandt and Ginzburg(11), Smith and Chu(2), Seiden(12) and Boughton et al.(13) estimated the disappearance of superconductivity on cadmium at 120 kbar, 38 kbar, 700 kbar and 325 kbar, respectively. The experimental check of these disagreement is very interesting.

On the other hand, the physics of one dimensional and two dimensional materials become the topic at present. A silver subfluoride, Ag_2F, which is a two dimensional material, is a superconductor with a transition temperature of 0.066 K(14). The pressure effect of the superconducting transition temperature on Ag_2F might be a check of new superconducting mechanism.

The preliminary experiment of the superconducting transition temperature of Cd and Ag_2F are done. The superconducting transition temperature of Cd is 0.515 K at normal pressure. The superconducting transition temperature under 8 kbar and 24 kbar are measured using the D-I type cell (4 mm Bridgman-anvil). The Tc is 180 mK and 140 mK, respectively. Further experiment is still doing.

The pressure effect on the superconducting transition temperature of Ag_2F is measured but is not completed yet. The main reason is the low resistivity of the sample ($10^{-3}\Omega$ at 1 K). As the cooling power of the refrigerator decreases rapidly at lower temperatures, the maximum current in the case of the d.c. resistance measurement is limited. At a temperature of 66 mK, the critical current which maintains thermal equilibrium is 25 μA.

7. Conclusion

Two small high pressure clamp cell are devised. The highest pressure attained is 50 kbars. They are attached to the helium 3/helium 4 dilution refrigerator and are cooled down to the several tens millikelvin. One is used for the measurement of the d.c. electrical resistance and another is used for the measurement of the a.c. magnetic susceptibility. The former cell is cooled down to 30 mK and its temperature homogeneity is $\pm$ 5 mK, but the latter cell is cooled down to 80 mK owing to its structural reason, and the temperature distribution is not homogeneous. As the cooling power of the dilution refrigerator decreases rapidly at lower temperatures, the usual method for detecting the superconducting transition temperature is not applicable because of the small resistance change in the case of the d.c. method and the a.c. eddy current heating in the case of the a.c. method. Therefore, it is necessary to develop further methods of detection.

Acknowledgement

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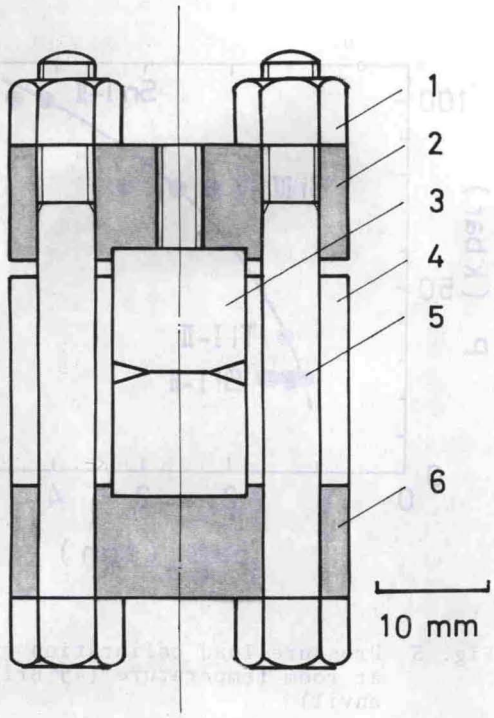

Fig. 1 D-I type high pressure clamp cell
This cell is used for a.d.c. electrical resistance measurement

1. Fixing nut
2. Upper flange
3. Tungsten carbide anvil
4. Anvil guide
5. Fixing bolt
6. Lower flange

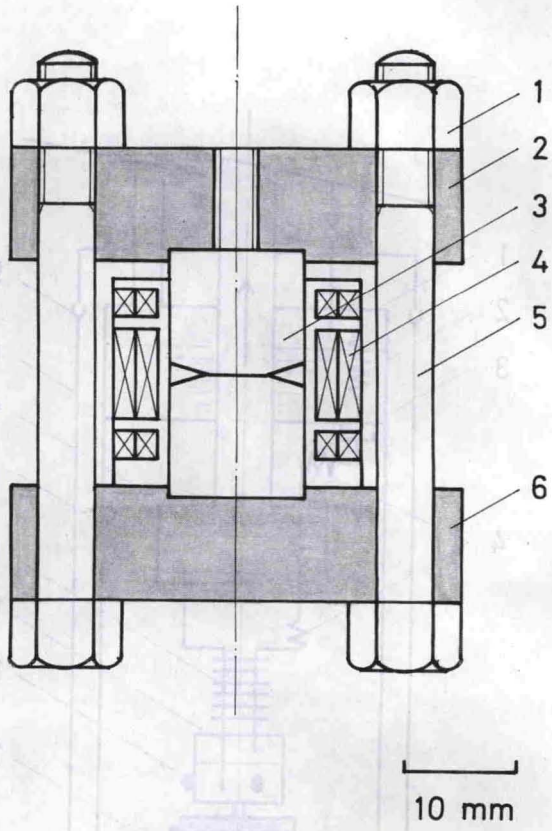

Fig. 2 D-II type high pressure clamp cell
This cell is used for an a.c. magnetic susceptibility measurement

1. Fixing nut
2. Upper flange
3. Tungsten carbide anvil
4. Measuring coil
5. Fixing bolt
6. Lower flange

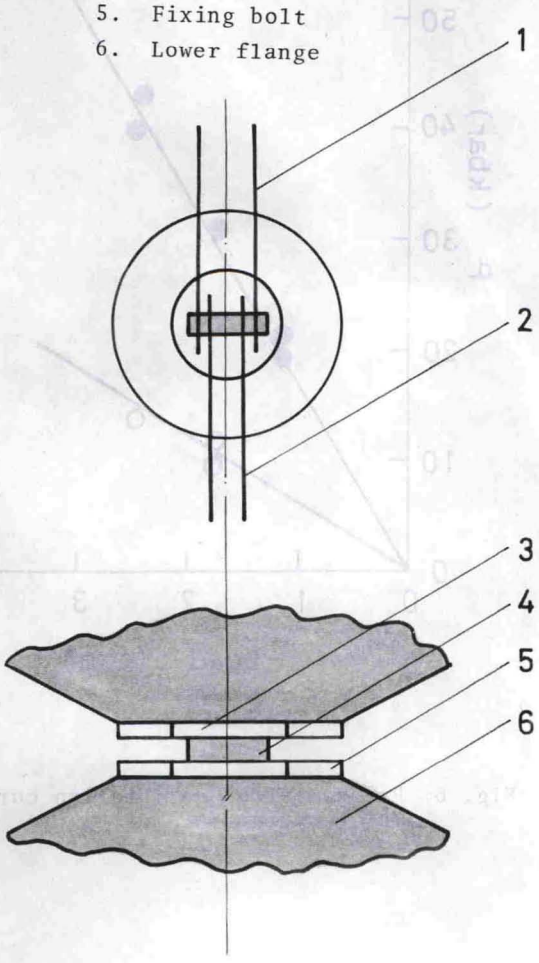

Fig. 3 Sample assembly for d.c. method

1. Current lead
2. Potential lead
3. Talc disk
4. Sample
5. Phyrophyllite ring
6. Tungsten carbide anvil

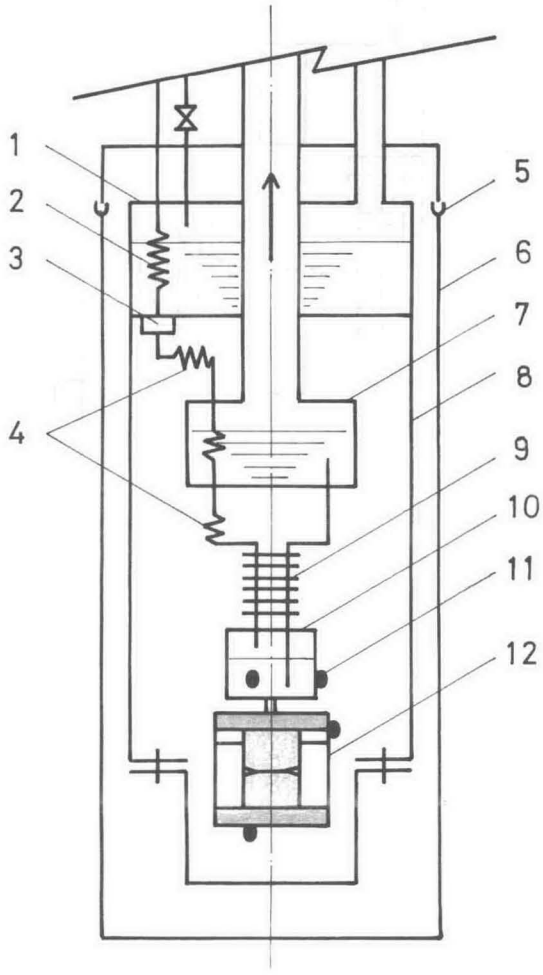

Fig. 4 Dilution cryostat for high pressure experiments

1. He^4 evaporator
2. Heat exchanger
3. Condenser
4. Impedance
5. Wood's metal seal
6. Vacuum jacket
7. Still
8. Shield jacket
9. Heat exchanger
10. Mixing chamber
11. Thermometer
12. High pressure cell

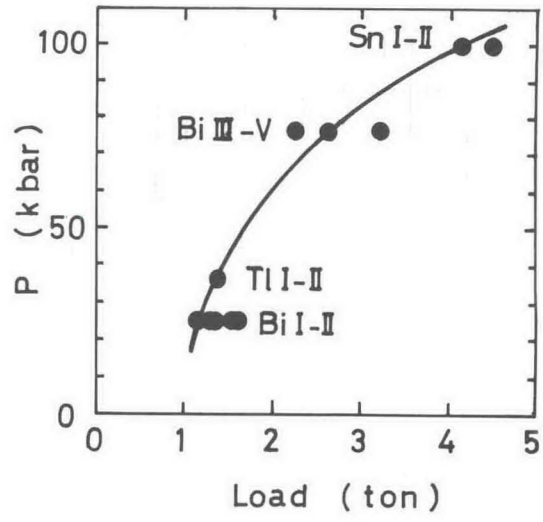

Fig. 5 Pressure-load calibration curve at room temperature (4ϕ Bridgman anvil)

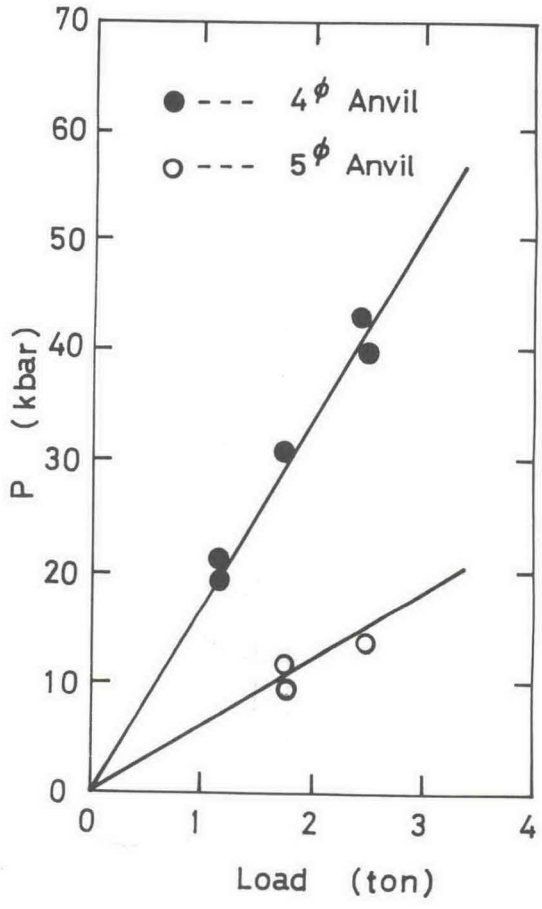

Fig. 6 Pressure-load calibration curve at low temperature

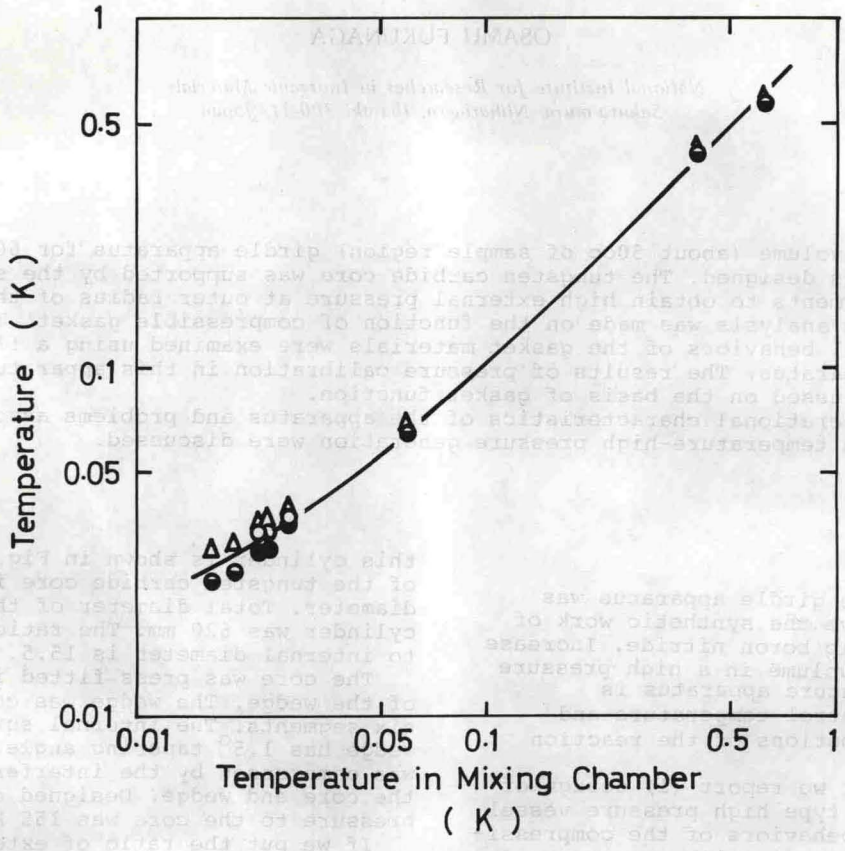

Fig. 7 Temperature distribution in the D-I apparatus

● ..... temperature of mixing chamber body

o ..... temperature of upper flange

Δ ..... temperature of lower flange

SUPPORTED WEDGE TYPE GIRDLE APPARATUS

OSAMU FUKUNAGA

National Institute for Researches in Inorganic Materials
Sakura-mura, Niihari-gun, Ibaraki 300-31, Japan

Large volume (about 50cc of sample region) girdle apparatus for 60 kb region was designed. The tungsten carbide core was supported by the six wedge segments to obtain high external pressure at outer radius of the core. The analysis was made on the function of compressible gasket. The mechanical behaviors of the gasket materials were examined using a flat anvil apparatus. The results of pressure calibration in this apparatus were discussed on the basis of gasket function.

The operational characteristics of the apparatus and problems associated with high temperature-high pressure generation were discussed.

Introduction

A large scale girdle apparatus was designed to serve the synthetic work of diamond and cubic boron nitride. Increase of pressurized volume in a high pressure and high temperature apparatus is important to control temperature and pressure distributions of the reaction chamber.

In this paper we report (1) design of supported wedge type high pressure vessel (2) mechanical behaviors of the compressible gasket and (3) miscellaneous operation conditions of the apparatus up to 60 kb and 1500°C region.

Design of girdle cylinder

The girdle cylinder was designed so as to reduce the ratio of external to internal diameter. The dimension of this cylinder is shown in Fig.1. The bor of the tungsten carbide core is 40 mm in diameter. Total diameter of the girdle cylinder was 620 mm. The ratio of externa to internal diameter is 15.5.

The core was press-fitted into the bor of the wedge. The wedge was composed of six segments. The internal surface of the wedge has 1.5° tapering angle. The core was compressed by the interference betwee the core and wedge. Designed external pressure to the core was 155 kg/mm^2.

If we put the ratio of external to internal diameter of the core as k_o, the calculated maximum tangential stress at the inner radius of the core is,

$$\sigma_t = \frac{k_o^2+1}{k_o^2-1} P_i - \frac{2k_o}{k_o^2-1} P_m$$

where Pi is internal pressure and Pm external pressure at the outer radius of t core.

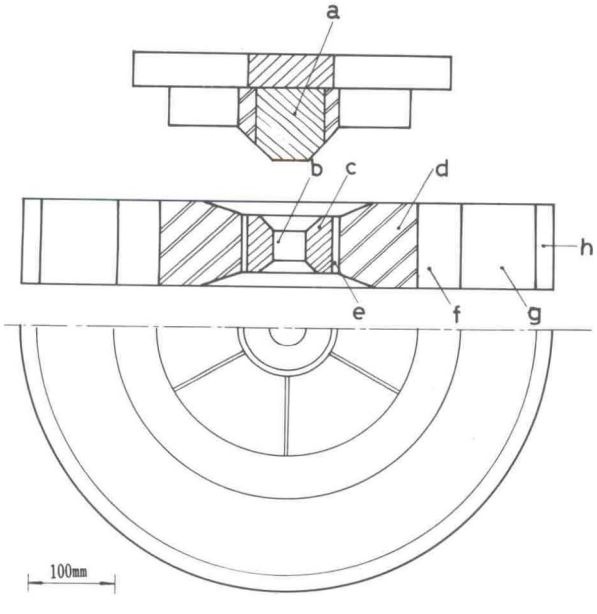

FIG.1. The schematic section of the girdle cylinder and anvil.

a. Tungsten carbide anvil
b. Sample chamber
c. Tungsten carbide core
d. High-speed-steel wedge
e. Shim
f. Binding ring(II) SNCM-8 steel
g. Binding ring (I) SNCM-8 steel
h. Safety ring