ENLARGED COMPRESSION CHAMBER IN HIGH PRESSURE – HIGH TEMPERATURE MULTI-ANVIL DEVICE

Guerold S. Bobrovnitchii
North Fluminense State University, Av. Alberto Lamego 2000, UENF – Parque Califórnia - Campos dos Goytacazes – RJ – CEP 28015-620, Brazil
guerold@uenf.br

Abstract. *The treatment of materials under high pressures exceeding 4.0 GPa using samples with increased length is a highly relevant problem in high-pressure physics. The work reported on here consisted of the construction of a high-pressure multi-anvil device with a parallelepiped-shaped compression chamber and four active horizontal anvils. This design allows the compression chamber to be used with lengths that are limited only by the dimensions of the working space of the hydraulic press utilized. However, the height of the working space of the model DO044 2500-ton load press is equal to 600 mm, while the maximum height of the compression chamber under a pressure of 5.5 GPa and temperature of 1500^0 C did not exceed 70 mm. The transversal dimensions of the compression chamber are 25 x 25 mm and its anvils are made of rapid steel. The use of WC/Co tungsten carbide for anvils allows for a pressure of 6.5 GPa to be generated. To generate high pressure, the horizontal anvils are driven by applying a hydrostatic pressure of 500 MPa. The experiments carried out so far have demonstrated the efficiency of this design, although the width of the working space of the press is not optimal for this type of device.*

Keywords: High pressure device, High Pressure and High Temperature.

1. Introduction

The 1960-1970s are characterized by an outburst of information regarding the development of high pressure device constructions. From the first diamond syntheses in different countries, equipment construction for treatment by high pressure and high temperature were divided into two groups: uniaxis and multiaxis (Bundy,1962; Tsiclis,1976; Spain,1977). It is interesting to note that the first synthesis was done using the multiaxis device (Platen,1962). The initial ten years of intense development of multiaxis devices or multi-pistons revealed that these constructions are more effective than the multiaxis (Bundy,1988), from the point of view of the generation of almost hydrostatic high pressure from 4,0 up to 10,0 GPa. However, they lost their leadership due to construction complexity, maintenance and also because the products manufactured with such device were too costly.

Amidst the multiaxis constructions, the devices with anvil (pistons) hydrostatic transmission stand out (Zeitlin,1965a; Kawai, 1970; Wittermn and Werkman,1963; Bobrovnitchii and Maksimov, 1974). In the last 15 years, after a prolonged pause of progress, the employment of triaxis high pressure device has once again begun, meaning, devices whose compression chamber has the form of a cube (Bobrovnitchii et al,1992;Chepurnov et al,1997; Bobrovnitchii and Persikov,2001; Bobrovnitchii, 1997). The reactive cell of such devices does not take up the entire height of the cube, which complicates the treatment of prolonged samples. The way out of this situation was seen in the early 1960s, when “Barogenics” (USA) (Zeitlin,1965b) launched the projects of industrialized high pressure devices with a triangular prism-shaped compression chamber 500mm high and 150mm wide. The principal element of this construction consists of the effort of only the elongated anvils in perpendicular direction to the longitudinal axis of the device on account of exertion of the cylinders or hydrostatic pressure. This idea did not loose recognition and divided itself in two courses: generation of high pressure in solid medium on account of plasto-elasctic deformation of barbs (gaskets) amidst lateral surfaces of the anvils or on account of lateral surface sliding, one related to the other (Tyrner,1966). It was the first course that received attention and progressed. However, not all the mentioned constructions were continued or progressed for many reasons, some of these due to complexity, high-costs, and rarefactive application.

The construction presented in this work (Witterman and Werkman,1963) is more responsive to the needs of industry and was approved for synthetic diamond syntheses. Concerning length size of the treated sample, this construction holds advantages, but the diameter of the sample was small.

In the present work the principal element analysis of existing high pressure device constructions, and considering demands that introduce scientific and industrial practices to develop the project, the construction design of the device with the compression chamber in parallelepiped shape and hydrostatic transmission of the active lateral anvils was emphasized.

This scheme enables to use the compression chamber with a length that only depends on distance from top to bottom of the hydraulic press that is available. Furthermore, it was regarded that the elaborated construction can be employed for other technological processes, particularly, powder sinterization.

2. High pressure device project

Throughout project development, the following were tried to guarantee:
-- Maximum dimensions of compression chamber, considering the limited hydrostatic recipient dimension by resistance of 600mm among the columns of the available press (2500ton force, model D0044, Russia);
-- Height of compression chamber that accepts treatment of samples of 65mm in length (mean dimension for mechanical tests)
-- Maximum convenience and facility for mounting and dismounting, maintenance during loading, removal of samples and transposition of anvils.

2.1 Theoretical part

The construction design of the proposed device is illustrated in Fig. 1. The device functions together with a hydraulic press that applies its force along the vertical axis.

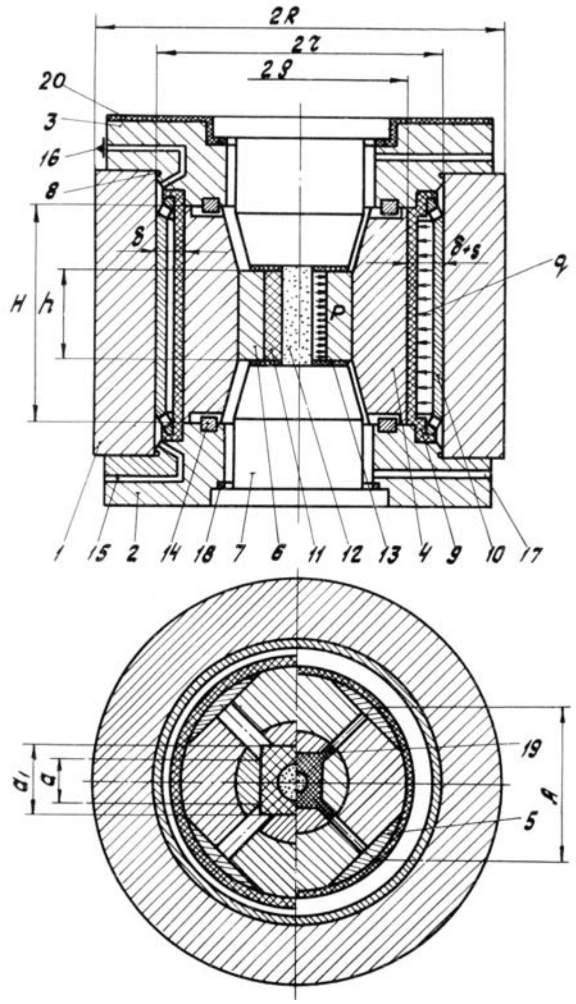


Figure 1. Construction design of high pressure and high temperature device with elongated compression chamber.

The device is composed of a recipient (1), superior lid (3) and lower lid (2) where the four sectors are placed (4). The slits in between the sectors are closed by scutes (5) around the segments. The anvil (6) is fixed on the inner surface of each sector. Connected to the surface at the top of the vertical pistons (7) the anvils (6) shape the compression chamber in parallelepiped shape. The deformable capsule (11) made of pyrophyllite with the sample to be treated is between the anvils (6) and the pistons (7), creating the cracks between the lateral surfaces of the anvils. The gaskets (13) are fixed on top of the anvils. Sealing the space between the internal surface of the recipient (1) and external surfaces of the sections (4) and lids, is accomplished by rubber rings (8) and by the encasement of the thin wall made of polyurethane (9). Extremities of the encasement are compressed between the lids and the column. Movement synchronization of the sectors (4) is not only done by the scuds, but by guides cotters (14). The oil under high pressure goes into the recipient through the channel (aperture) (15). The valve (16) serves to install the manometer. Through the channel (17) refrigeration of the sectors and other elements is achieved. Sealing of refrigeration space is obtained by rubber rings (18).

The device works in the following manner. The device permanently mounted with the deformable capsule (11) and sample (12) is placed between the beams of the hydraulic press. Trough the hydrocylinder of the press, compression of the device in direction of the vertical axis is performed. The crack between the sectors (4) and lids (2,3) disappears or is minimized. The external source (special pump) spins pressure "q" in space "δ"of the recipient. The sectors (4) shift in perpendicular direction to the vertical axis, thus compressing the deformable capsule in parallelepiped shape and forming the gaskets (19) or simultaneously deforming the gaskets (19), initially placed among the sectors.

The device enables heating the sample directly (through the electrical current passing by the pistons and sample). For this purpose the superior piston and lid are isolated by means of isolating joint (20).

2.2 Optimization endeavor of compression chamber dimensions

In order to assure maximum dimensions of compression chamber of the proposed device, it is interesting to show dependency between q pressure in the recipient and the volume of the compression chamber, considering the already known external diameter $2R$ and the value of p pressure in the compression chamber.

For a recipient composed of multi-elements and loaded by internal pressure q, the optimized correlation is(Bobrovnitchii et al,1998).

$$r = R\sqrt{\left(1 - \frac{2q}{n[\sigma]}\right)^n} \qquad (1)$$

Where: r and R– the corresponding internal and external radius; n – number of elements (rings) of composed element; σ - acceptable maximum tension for material of each ring.

On the other hand (see Fig. 1):

$$r = \rho + \delta + S \qquad (2)$$

Where: ρ – is the external radius of the sectors shut by pressure q ;

δ – initial distance between recipient and external surface of sections at initial position; $\delta = 0{,}1\rho$.

$S = (a_1 - a)/2$ – approximate value of compression course of the anvil that corresponds to the pressure value determined p;

a – face dimension of the anvil;

a_i – initial dimension of the deformable capsule.

For expressions close to 6,0 GPa correlation is $a_i/a = 1{,}4$ that is, $S = 0{,}2a$ (Zeitlin and Brayman,1963).

External radius of closed sections under pressure can be presented by equation

$$\rho = \frac{\sqrt{2}}{2} \cdot \frac{p.a.h}{g.H.\eta} \qquad (3)$$

Where: p – pressure in compression chamber;

h – height of compression chamber;

H – section height;

η – coefficient that shows which external force applied to the section transforms into pressure in the compression chamber. The remaining part is applied to forming the gasket. In conformity to pressures from 5,0 up to 6,0 GPa η = 0,6 (Zeitlin and Brayman,1963);

The obtained dependencies lead to the following correlation:

$$a = \frac{R\sqrt{\left(1 - \frac{2q}{n[\sigma]}\right)^n}}{0{,}778 \cdot \frac{p.h}{q.H.\eta} + 0{,}2} \qquad (4)$$

To determine maximum value of a it is necessary to derive the equation (4) in relation to q and equalize the derivation to zero (a' = 0), determine the value of q that cannot surpass a limit that depends of axial force of the press.

Correlation h/H is a totally determined value. In Fig. 2 the dependencies obtained base don resistance calculation for the sector (Timoshenko and Goodien,1970) are presented. Graph analysis shows that the value h/H of must be smaller than 0,4 for the observed case.

For the elaborated high pressure device the minimum value of anvil face is a = 25mm, when R = 270mm; h = 70mm, H = 200mm; $[\sigma]$ = 1300 MPa; p = 6,0 GPa ; q = 515 MPa and n = 2;

2.3. Project development

To assure future operation of the elaborated device, a model in ½ scale made of organic glass was manufactured. This enabled to show the defects of the basic project and future manufacturing, to verify sealing system and develop manufacturing technology of the natural device.

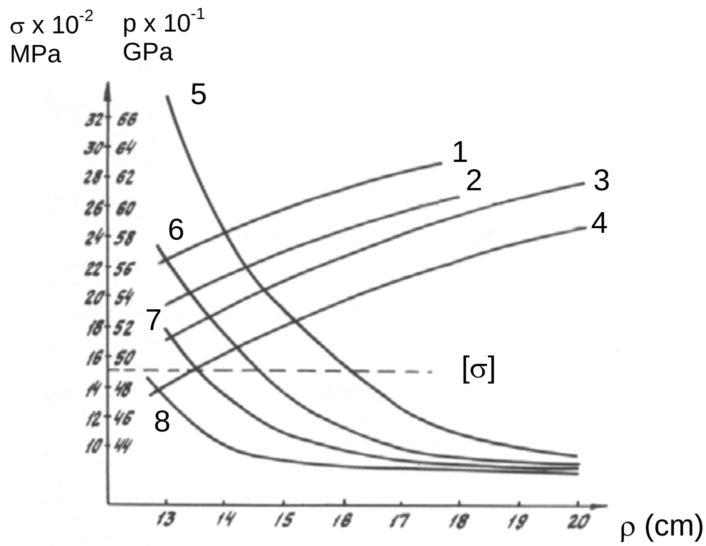


Figure 2 . Dependency among dimensions of the sector ρ, H and its resistance σ and generated pressure p:

- curves 1,2,3,4 of function ***p = f (q)*** for H 240, H 210, H 180,H 170mm,correspondingly;
- curves 5,6,7,8 of function ***= f (q)*** for H 240, H 210, H 180,H 170mm,correspondingly;

The authentic project construction of the device is done in accordance to results of the model study. Furthermore, complementary questions were solved: the possibility of moving the device with the pressure source ***q***; refrigeration; cleaning internal space of the remaining deformed capsule; fabrication technology of the polyurethane enclosure; fabrication of deformable capsule in pyrophyllite parallelepiped shape with longitudinal orifice; a hydraulic compressor GKM – 6/7000 (High Pressure Physics Institute, Moscow) was chosen as ***q*** pressure source.

Main technical characteristics of elaborated high pressure device: external dimensions – 550x470; maximum pressure in recipient 500MPa; weight – 800Kg; horizontal movement course is 750mm. The pressure to be generated in the compression chamber **–** 3,5 GPa for chamber of 55x55x120mm dimensions and for one of 6,0GPa dimensions are 25x25x70mm. Each of these modifications foresees two anvil and piston variations: superior and lower tops of anvils and pistons paralleled, and superior and lower tops of anvils inclined and pistons are forms of truncated symmetrical pyramid..

2.4. Fabrication Details

Most of the device’s force elements were made of high resistance steel type 4340. The anvils and pistons were manufactured in fast steel type W-2 and hard metal WC 6% Co. The anvils of fast steel obtained HRC 62-64 hardness. Manufacturing technology did not present difficulties and precision was in the range of 0,02 to 0,05 mm.

What presented more difficulty was the polyurethane encasement cylinder shaped of thin wall with extremities folded outward. As the wall was thin t = 2mm and significant height was of 270mm, the centrifuge method was applied to form the encasement in a special mold.

2.5. Previous device tests

Ten previous tests were carried out using aluminum capsules under 100 to 300MPa pressure in the recipient. During maintenance of 350 MPa pressure the level dropped by 1,0 MPa during thirty minutes. The control dismantling showed that all parts, including the encasement, do not have deformation signs. The difference in the irregular formation of gaskets (barbs) was noted. The difference of gasket density in 10 loading was of 0,2mm up to 0,6mm, which can cause bursting of compressed ceramic material in future experiments. This defect was caused by low parallelism between lids and sectors, causing movement synchronization problems of the anvils in the center. Some of the construction defects were eliminated to a certain degree.

3. Experimental Part

The elaborated methodology of the study of the new high pressure device construction includes:

-- Determination of type of pressure distribution in the transversal section of the compression chamber during pressure generation, applying in the initial parallelepiped transversal section the typographic ink net with precise dimensions of 2x2mm and then parallelepiped compression under pressure of $\boldsymbol{q}$ = 300MPa;

-- Preparation of pyrophyllite deformable capsules (parallelepiped) from the mineral parts by milling;

-- Preparation of reactive mix composed of powdered graphite 50% and chipped alloy Ni60%Mn. Compaction pressure was of 300 MPa. Diameter of compacts was of 15mm and height was 10mm;

-- Heater lids were compacted under the same pressure from graphite mix 50% and pyrophyllite 50%.

-- Capsule assembly was accomplished the following manner: within orifice six reactive mix compacts are placed and at extremities the heater lids are positioned;

-- Dependency between "***q***" of recipient and "***p***" at the center of the compression chamber was determined by registration of phase transformation under applied pressure of Bi (2,55 GPa), PbSe (4,23GPa) and Ba (5,5 GPa). The base of these measurements was constructed by calibration curve $\boldsymbol{p = f(q)}$ (Zeitlin and Brayman,1963)

-- Dependency between temperature at the three points in the compression chamber axis and the heater's electrical current was gradually determined using the Pt/Pt-10%Rh thermocouples. Measurements enabled to construct calibration curve $\boldsymbol{T^o = f(I)}$ (Houck and Hulton,1973)

-- Diamond synthesis with ***p,T***-parameters of 4,3 to 4,9 GPa and from 1150º to 1350º C was accomplished in the Ni-Mn-C system during 3 to 9 min. From the obtained agglomerates extraction and purification of diamonds was performed.

-- Sinterizaton of long samples from WC + 6%Co powder was accomplished under pressure of up to 3,5GPa and temperatures of up to 1200º C.

4. Results and Discussion

Fig. 3 illustrates the parallelepiped transversal section of the aluminum treated with q = 300MPa pressure. As can be seen in the central part of the section the squares were not deformed. This means the absence of plastic deformation. If 5% is considered shearing deformity of net quadrate as a consequence of hydrostatic pressure, it can be confirmed that almost hydrostatic pressure operates in 22 to 25% of the volume of the compression chamber.

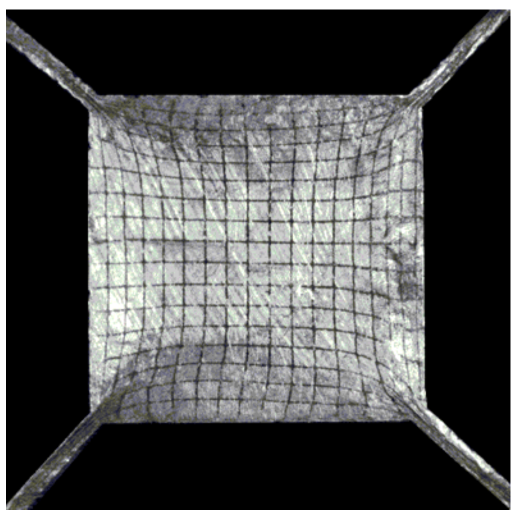


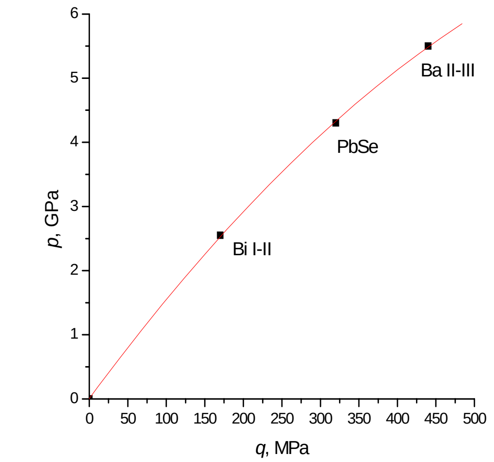
Figure 3. Photograph image of deformation in the transversal section of compressed parallelepiped in the elaborated device.

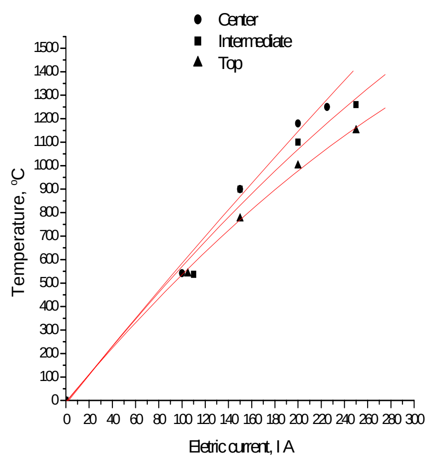
The continuation of gasket irregular formations was revealed. The density difference was measured in 0,32mm. Thus, with the a_i/a increase, it has the tendency to grow. Using previously formed gaskets diminishes the difference to 0,11mm.

Pressure calibration shows non-linear dependency between ***p*** pressures insides the chamber and ***q*** of the recipient (Fig. 4a). With the ***q*** pressure increase, pressure ***p*** in the compression chamber tends to diminish value growth. It was noted that this effect depends on initial dimensions of capsule $\boldsymbol{a_i}$ in correlation a_i/a . This dependency is seen in Fig. 5.

As can be seen, for each level of pressure ***p*** there is the a_i / a correlation value. Fig. 4b enables possibility of evaluating influence of electrical current of heating over generated temperature over the five points of the compression chamber. Constant temperature was measured in 3 points. Applied potency to obtain the same temperatures, as in the anvil-type concave device is reasonably low.

Synthesis results are shown in Tab. 1





a b

Figure 4. – a - pressure calibration curve in compression chamber ***p*** in relation to pressure in recipient ***q***.
b - Temperature calibration curves inside compression chamber.

Table 1. Results of synthesis for each series in average.

Sample series	Pressure, GPa	Temperature, ºC	Synthesis time, min	Synthesis result	Efficiency, carat
1	4,3	1150	3	_	_
2	4,3	1200	3	48/20; cube and octahedral cube	2,5
3	4,3	1300	3	160/125	12,8
4	4,6	1200	5	125/100; Mostly octahedral cube	3,8
5	4,6	1225	5	250/200; Half cube octahedral	13,1
6	4,6	1350	5	500/250; Mostly octahedral	21
7	4,9	1250	8	125/80; Octahedral and cube	11,2
8	4,9	1325	8	450/200; Mostly octahedral	19,3
9	4,9	1400	8	Explosion after 1,5-3 min current ligament	-----

As can be seen diamond productivity and form are associated to ***p,T-*** parameters, generated in the compression chamber and time. Due to irregular gasket density formation, it was not possible to finalize the tests. Comparing diamond production in Belt devices with the same press force, it can be stated that the new device loses competition, despite producing larger sized diamonds. It can be explained the following manner: the press adapted for Belt is not suitable for a new device construction. Compared to the productivity of the anvil-type device of concavity diameter of 55mm, the developed device has advantages.

Durability life-span of lateral anvils made of hard steel or fast steel decreases with pressure and temperature increase.

Under pressure in the compression chamber of about 4,3 GPa and T = 1300ºC after 52 operations, a hard metal lateral anvil cracked in the middle. Anvils of fast steel endured these parameters up to 98 operations. Under pressure of 4,6 GPa and temperature 1350ºC both lateral anvils of hard metal lost quality after 49 operations and the anvil of fast steel after 83 loadings. For pressures of 4,9 GPa and temperatures of 1400ºC, durability life-span of anvils diminished to 28 and 46 operations, relatively. Analyzing deformation of anvils and lateral sections after cracks, it was determined that plastic deformation of section flexion caused the cracks, that is, section height was not correctly calculated.

Attempt to sinterize metal-ceramic materials under other pressures and temperatures showed that without previous treatment of the 18mm diameter and 50mm high joggle, it was impossible to obtain powder samples of WC and Co. The four pre-compacted samples in 500 pressure MPa matrixes of 15mm in height and mounted in the central orifice of the capsule after applying 4,5 GPa pressure and 1350ºC temperature, had porosity from 0,6 to 0,9%. During tensile test the sample divided into four samples with the crack region near regions of previous contact. Only hydrostatic pre-compaction resolved the quality problem of the sample, however, the lateral surface was no longer cylindrical or the axis curved. This can be explained by the pressure gradients along the height of the compression chamber and lack of experience in forming prolonged samples.

The results of this work show that the construction design of the high pressure device with elongated compression chamber has chances to be used. Employment usefulness depends on optimization of some construction parameters: accurate correlation between anvil heights and the sections, among the external and internal radius of the section during pressure generation and synchronized movement in the center. These main questions can be solved only in a new device project projecting it with a special force structure type press with a small course of the primary cylinder.

5. Conclusions

The developed construction of the high pressure device with elongated pressure chamber demonstrated its work capacity to generate pressure up to 5,0GPa and temperature up to 1325ºC in 36cm^3 volume.

The elaborated device demonstrated capacity to sinterize diamonds of narrow granulometric thickness of 140 up to 430 μm with less electric potency application.

For practical application, it is necessary to modernize construction, diminishing section heights and enlarging the internal diameter of the recipient.

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