

CHAPTER 2

Non-magnetic high-pressure chambers for physical investigations

Fundamental researches by P. Bridgeman [12] initiated studying of the behavior of materials under pressure. Effects of such powerful thermodynamic parameters as temperature and pressure on changes of resistive, resonance, optical and magnetic properties of solids remain still of interest.

Unfortunately, the literature data are deficient, concerning the development of experimental procedures for investigations under pressure. In a number of monographs [12-17], various physical methods are described that are based on joint application of hydrostatic pressure, temperature and magnetic fields used for studying properties of materials.

Current level of research work requires new approaches to stimulate the development of methods and procedures. Utilization of new composite non-magnetic materials demands modernization and unification of the existing methods of pressure generation together with design of pressure vessels.

Here we generalize methods used to study properties of materials under hydrostatic compression up to 3 GPa (30 kbar). Several described HPC modifications are intended for different physical methods of investigation. High attention is paid to the technology of packing, optimization of geometric dimensions. We demonstrate methodic and practical developments of multifunctional nonmagnetic HPC for pressure up to 3.0 GPa in a wide temperature range (0.4-450 K).

The proposed designs and methods can be of interest for experimentalists working in the field of high-pressure physics and technology.

2.1. Non-magnetic HPC (up to 15 GPa) for resonance investigations

One-layer straight resonator-chamber (Fig. 2.1) is designed for experiments in a wide range of frequencies, magnetic fields and pressures. Dimensions of the chamber are optimal: $D/d=3.65$ (D is the external diameter, $D=31$ mm, d is the diameter of working channel, $d=8.5$ mm).

The design and procedure show that the force is transmitted from hydraulic press via plunger 16 made of beryllium bronze (БрБ2) after special thermomechanical processing. At plunger side, the chamber is sealed with rings 13, 15 made of БрБ2 (HRC-33-36). Teflon ring 14 tightened with screw 12 is placed for the case of pentane use. Nut 17 serves for plunger location, nut 18 is for plunger removal for inspection or refilling the chamber. Radio-frequency shutter 3 is anchored by nut 1 and sealed by lead or PTFE ring 5

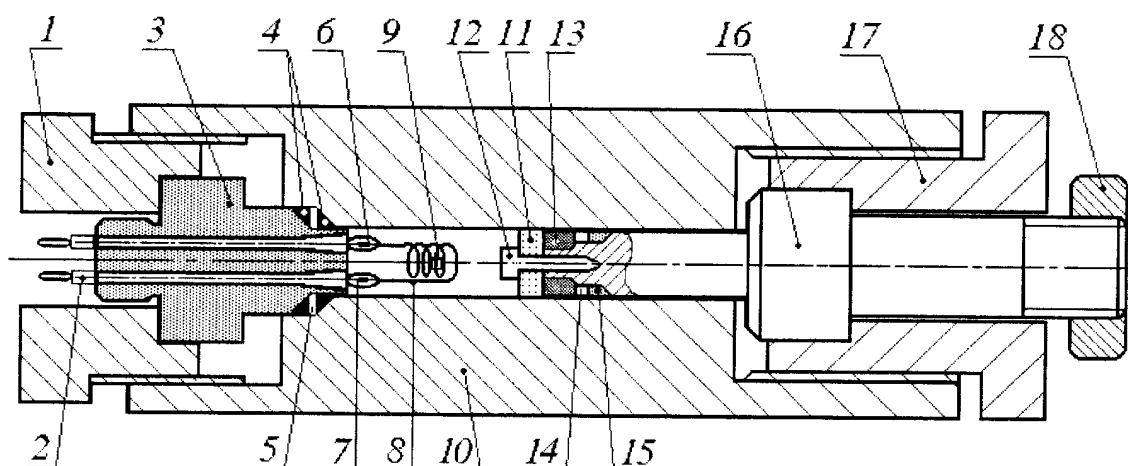


Figure 2.1. Non-magnetic HPC (up to 1.5 GPa) for resonance investigations.

together with anti-extrusion rings 4 made of БрБ2. The shutter has screw for remover. Coaxial bushings 6 and 7 are insulated with araldite rein AA-004.

Chamber casing 10, plunger and shutter were subjected to thermomechanical treatment for increase the pressure range where deformations remain elastic. During the treatment, the channel was autofretted by sequential pushing of high-strength cores-mandrels (steel XБГ, HRC-60-62).

The chamber was used for observation of antiferromagnetic resonance (AFMR) under high hydrostatic pressure for $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$ crystal in decimeter wave band and inclined magnetic field at helium temperatures [18].

The resonator-chamber together with a sample was placed in a Dewar vessel filled with liquid helium at temperature varied from 1.68 to 4.2 K by pumping helium vapor out. The pressure was measured and the elasticity of transmitting medium (oil-kerosene mixture) was tested by a contactless method. A washer of pure tin 11, by 6 mm in diameter and 1mm in height was pasted to screw 12. The temperature of superconducting transition was estimated by vapor of the helium bath.

SHF-power was transmitted from generator to coaxial inlet 7. Sample 2 was placed in the center of spiral 8 on polystyrene holder. Power was transmitted from coaxial terminal 6 via joint 2 to the receiving section. Frequencies were checked by the signal of free radical of biphenyltecrylhydrasile placed in the resonator.

2.2. Two-layer non-magnetic HPC (up to 2.56 GPa)

Peculiar features of the HPC are two-layer embodiment (БрБ2 is used in the both layers), D/d ratio is 5-7 and pressure application is extended up to 2.56 GPa [19-21]. To provide the advantages of a two-layer chamber [19], the autofretting was done (prior to installation) to extend the range of elastic operation of the external layer.

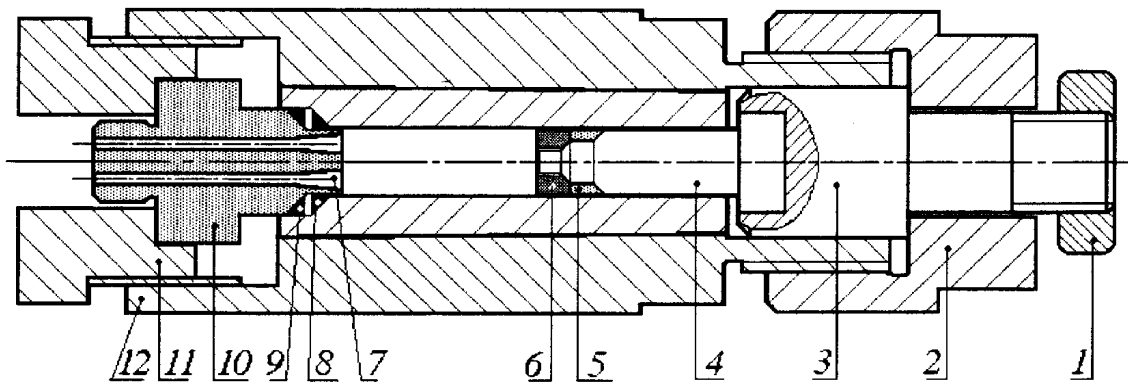


Figure 2.2. Two-layer non-magnetic chamber (up to 2.56 GPa).

The optimal HPC dimensions and the technology of treatment have given such parameters: $d=8.5$ mm, $D=45$ mm ($D/d=5.29$), the length of working cavity under pressure was 20 mm. The external layer of the chamber was autofretted by a steel hardened (HRC-60) mandrel (core angle was 2° , the tension was 0.37 mm). The inner layer was pressed into the external one with tension of 0.15 mm.

Figure 2.2 shows the general view of the chamber. Shutter unit 10,11 and packing of БрБ2 (HRC-30-32) 5,6,8,9 were like those described in [18] and operated safely in the whole pressure range (up to 2.56 GPa).

The working part 4 of the plunger was made of ceramics and aligned with guide part 3 by clearance fit. Insert made of microlite provides normal loading upto 2.3-2.56 GPa at room temperature, that is why the HPC casing was tested by using inserts made of metal-ceramic alloy BK10. Cavity pressure was measured by a manganin manometer.

Inspection and control of HPC operation channel showed no residual deformations in the casing, shutter and loading plunger after multiple pressure tests.

2.3. Solenoid-chamber

To increase the strength of high-pressure container, different methods are used, in particular banding of container bushing by high-strength wire or tape (Fig. 2.3) [22].

Superconducting winding 1 of wire HT-50 was laid with tension of $25-30\text{kg/mm}^2$ in circular cut of bushing 5 made of bronze БрБ2 with hardness HRC-40. Frame and superconductor have no extra isolation. Sample 2 is fixed in working space of bushing channel bounded by plungers with ferromagnetic poles 3 fixed by restraining nuts 6. Current leads are fixed to shutter 4.

At room temperature, the container is loaded up to prescribed pressure by a hydraulic press; plunger position is fixed by nuts. After cooling to the liquid

helium temperature, the material of bushing is hardened by a factor of 1.3 resulting in 10% compensation for the decrease of supporting force due to radial forces acting on the winding in the maximum magnetic field [23, 24].

Dimensions of the unit are as follows: $d=8.5\text{mm}$, the diameter of the external bushing is more than 22 mm, the total length is 46.2 mm, the lengths of upper and lower circular cuts are 35 mm and 25 mm, respectively. Winding with wire HT-50 (diameter of 0.33 mm) provided the magnetic field of 5.54 T for the current of 34 A, sectional winding with wire of diameter of 0.1 mm and 0.5 mm yields 7 T.

For a large solenoid, banding can be in the form of a separate sectional winding (Fig. 2.3, 6). Being banded with a superconductor (diameter of 0.7 mm) and having dimensions $d=8.5\text{ mm}$, diameter of inner bushing is 22 mm, external dimension is 41.3 mm, the structure ($d=8.5\text{mm}$) provides the total magnetic field of 9.2T [24]. After mounting the shutter and the piston made of steel with pole tips made of dysprosium, the container was tested at room temperature and pressure of 1.6 GPa. At 1.8 K and the working pressure of 10 GPa, the total magnetic field in the gap (2mm) between the tips was equal to

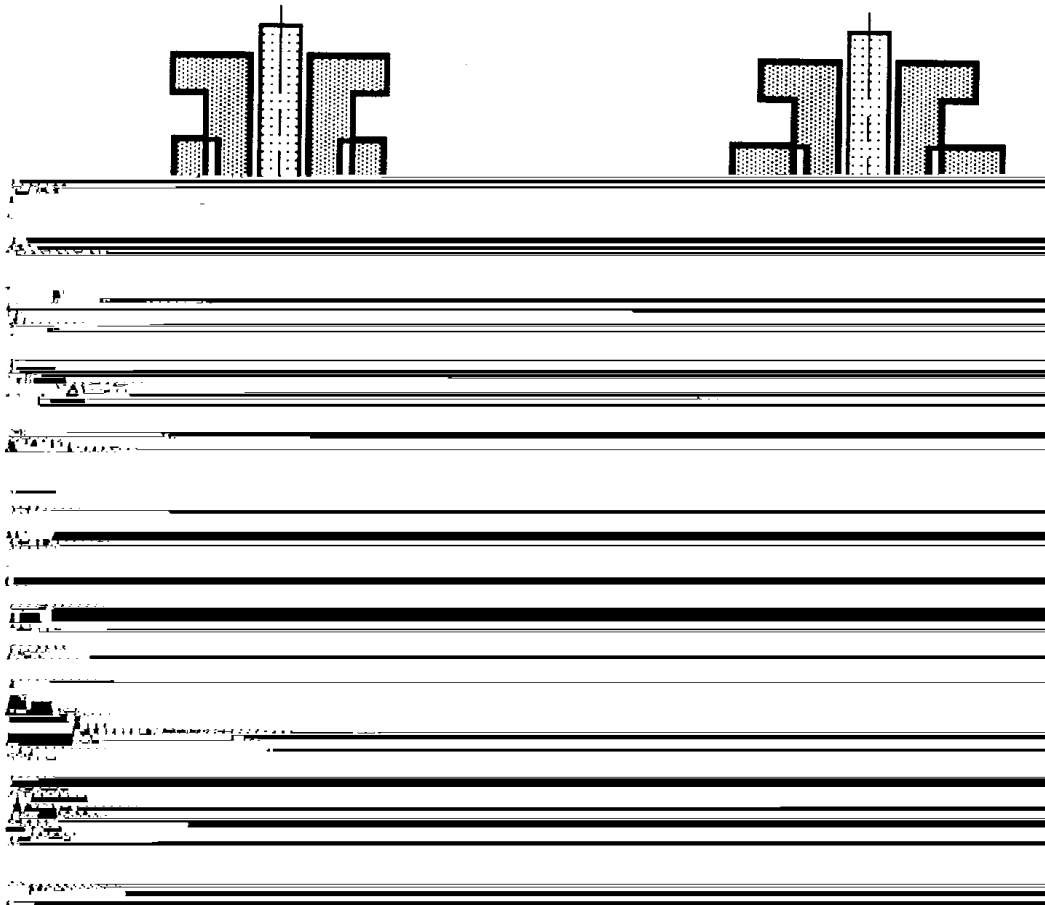


Figure 2.3. Solenoid-chamber with integral (a) and sectional (b) winding.

12 T. The pressure was estimated by initial loading of the container in the hydraulic press.

Thus, the solenoid-container is a promising structure for a complex use of high pressure, low temperature and strong magnetic field.

2.4. HPC (up to 3 GPa) for wide application

Figure 2.4 illustrates a two-layer non-magnetic HPC. It has been constructed by auto fastening of inner and external layers [19, 25] that has given container of smaller dimensions ($D/d=4.3-4.8$) and pressure increase by a factor of two.

Container with load – carrying parts made of БрБ2 has the overall dimensions of 30x130 mm, those of the active part are 7x30 mm. The piston is sectional. The packing is an antiextrusion ring of triangle section made of БрБ2 (HRC-35), teflon cone and a plug of benzo-oil resistant rubber. Pusher and rod are made of nickel alloy 38XHIO.

The shutter has two modifications depending on the measurement procedure. In the former case, it has two cone-like coaxial inlets for resonance investigations. In the latter case, current-carrying and measurement leads are taken off the zone of pressure by using cone insert as a sealing. These characteristics provide high technological quality of preparation and simple application.

The depicted container [26, 27] was used to investigate the influence of high hydrostatic pressure on the character of phase transitions in three-dimensional Heisenberg ferromagnets $\text{CuM}_2\text{Cl}_4 \cdot 2\text{H}_2\text{O}$ (M can be K, NH_4 , Rb). For these crystals, the measurements of magnetic susceptibility were made under pressure up to 1.76 GPa in the temperature range of 0.4-4.2 K. The pressure dependence of the Curie temperature has been determined.

A modified container structure (with all load-carrying elements made of alloy of 38XHIO –type and rod of hard alloy BK8, $d=6.5$ mm, $D=31$ mm, $D/d=4.8$) has made it possible to raise pressure in container working volume up to 2.7 GPa (2.1 GPa under helium temperatures). The container was used in

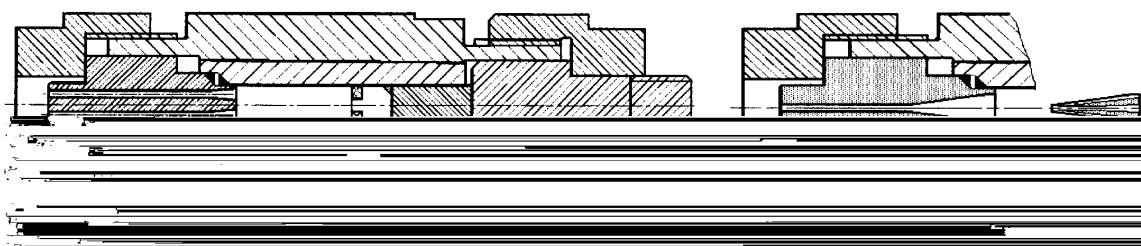


Figure 2.4. HPC (up to 3 GPa) for wide application (a) (b is the modification of shutter).

studies of high hydrostatic pressure effect on the Neel temperature and H-T phase diagram of $\text{GdBa}_2\text{Cu}_3\text{O}_{7-\delta}$ ($\delta=1$) [27].

Non-magnetic material with improved strength characteristics (40XHIO) permitted to extend the range of intra-chamber pressure up to 3.0 GPa with a 500-cycles loading-unloading process. Papers [28-32] contain the results of investigations on magnetic-field (to 8 kOe) effect on resistive properties of manganites for pressure to 1.8 GPa in 77-350 K temperature range.

2.5. Small-sized non-magnetic HPC (to 1.2 GPa)

Important features of HPC structure are the minimal length of 90 mm, that of operation space 23 mm, $D=18$ mm, $d=8$ mm and 5.4 mm ($D/d=2.2$ and 3.3, respectively), see Figure 2.5 [33].

In this structure, the hydraulic-press force is transmitted via pusher made of alloy 40XHIO (HRC-45-47), rod is made of structural ceramics (HRC-68), proton-free fluorite fluid C_8F_{12} is used to transmit pressure. Casing and sleeve nut of the rod are made of 40XHIO (HRC-45-47), shutter support is made of БрБ2 (HRC-37).

The reduction of the dimensions (see figure 2.6) required the optimization of supporting area of check nut, to have larger supporting area and shorter length of the shutter.

The chamber [33] was used for NMR studies in proton-containing samples. Figure 2.7 illustrates the general view of all HPC described above.

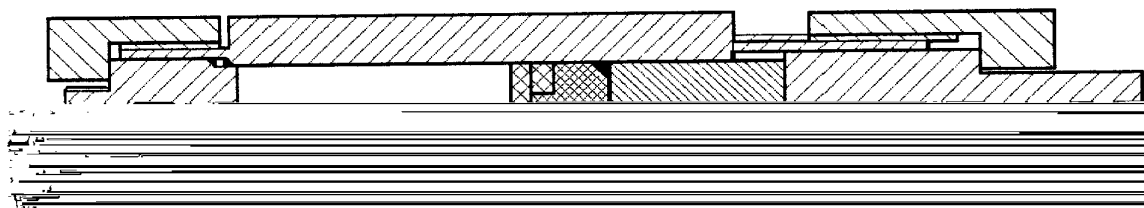


Figure 2.5. Small-sized nonmagnetic HPC (to 1.2 GPa).

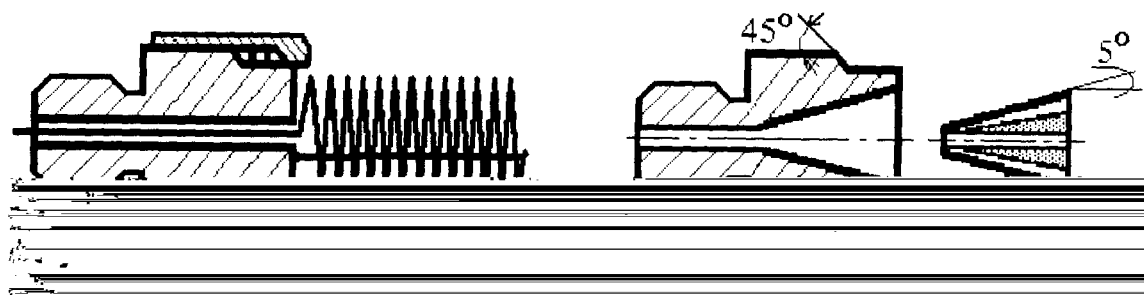


Figure 2.6. Modification of shutter for resonance (a) and resistivity (b) investigations.



Figure 2.7. High-pressure chambers.

2.6. X-ray diffraction studies at high pressures

During X-ray diffraction studies, the compressibility factors are determined by the relative changes of the dimensions of the crystal lattice under the uniform compression. The advantages of the method of measurement of the compressibility are shown in [34].

It is especially suitable when the crystals of large enough linear dimensions are not available. This is the only method enabling the investigation of compressibility anisotropy in polycrystalline samples. Therefore, the application of X-ray diffraction method makes the problem of obtaining the test samples much simpler because the requirements as to the linear dimensions and state of the samples are reduced. The measurements can be made on single crystals, polycrystals and powders of any degree of dispersion.

The peculiarity of the X-ray diffraction method used to measure the compressibility is that there is no contribution to the diffraction pattern from those parts of the samples where the crystal structure is distorted on the account of the defects and cracks. Thus, the information is taken by the method from crystal parts of the perfect structure.

High accuracy is provided by the modern high-resolution X-ray equipment and design of the high-pressure chamber with which both the measured parameters and the experimental conditions (pressure, temperature) can be

controlled. The accuracy is also improved by the statistical methods of data processing. The measurements done on the single-crystalline samples are much more accurate than those on polycrystals.

The detailed description of the high-pressure chamber is given in [35]. In our case, the chamber is of cylinder-piston type. It is an attachment to the home-made DRON-1,5 type horizontal diffractometer.

The figure 2.8 consists of the high-pressure chamber and a thermal stabilization system (not shown). Inside the chamber casing 1, there are beryllium container 4, gripped by two punches 2 and 3. The container is a bilateral casing with an axial hole, 2.5 mm in diameter. The outer diameter in the medium section of the casing makes 10 mm, taper angle is 4° . Nut 5 is for the container compression. Gaskets 6 made of annealed copper are between the container and the punches; they prevent the pressure-transmitting liquid from

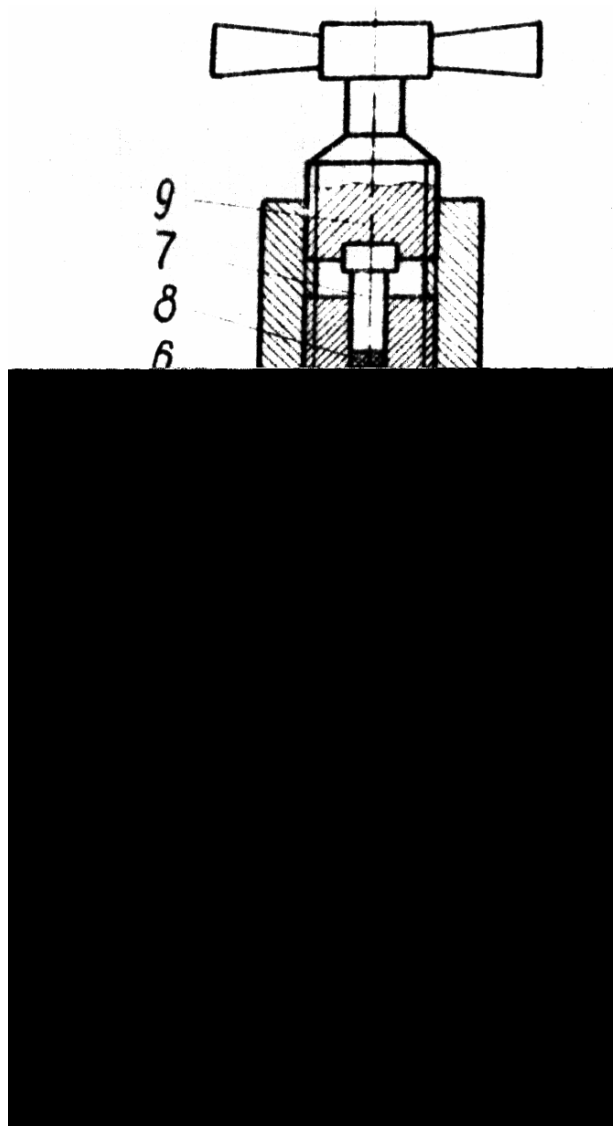


Figure 2.8. The high-pressure chamber for X-ray investigation.

the leakage. In the upper punch, there is a 3 mm diameter orifice where the pressure is generated with the help of piston 7. The piston diameter is 0.02-0.05 mm smaller than that of the channel. The piston is sealed by a two-layer plug 8 made of benzo-resistant rubber and paraffin. The benzene is a pressure transmitting liquid. Nut 9 is used to vary the pressure in the chamber. The pressure value is determined by a change of the resistance of manganite transducer 10. The resistance is measured by a direct-current bridge MO-62. With a microvoltammeter F18 Used as an external galvanometer for the bridge, it was possible to register the pressure changes in the chamber to within ± 10 bars. The temperature was measured by a copper-constantan thermocouple. The transducer and the thermocouple leads go through the channel in the lower punch and are pasted with epoxy resin. The investigated sample 11 is introduced to the container by holder 12 through the channel in the upper punch (the sealing plug 8 is removed). The substitution of the sample needs on serious dismounting of the chamber. In the chamber casing, there are partition windows for the fixation of X-ray pictures; The position of the chamber windows is not symmetric, so the range of the measured angles can be extended with the stable cell strength. The chamber is mounted on the goniometer of X-ray diffractometer DRON-1,5 by means of an adjusting device 13 which ensures the chamber motion in two mutually perpendicular directions with respect to goniometer axis during the adjustment with two micrometer screws 14. The rigid enough adjusting device makes it possible to vary the pressure directly at the goniometer and to avoid sudden errors in the measurement of relative changes in the crystal-lattice parameters induced by the pressure and by the use of removable chambers as a result.