

**USE OF HIGH PRESSURES AND HIGH TEMPERATURES IN PRODUCTION OF
YTTERBIA-BASED CERAMIC RADIATION SOURCES FOR BRACHYTHERAPY**

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Abstract

Method and apparatus for sintering ceramics of high density of ytterbium oxide for use in brachytherapy are suggested

Keywords: *high-dose-rate brachytherapy, ytterbia-based ceramic, high pressure*

1. INTRODUCTION

High-dose-rate brachytherapy (HDRB) is a modern method for treatment of oncological diseases, when an ionizing radiation source is inserted directly into a tumor through a catheter and removed after the end of the treatment session, thus substantially reducing irradiation of the normal tissues and organs in the patient. Usually, Iridium-192 (Ir-192) is used for HDRB as a radiation source in the form of metal wire with the diameter 0.6 mm and the length 3.0 mm. A new, more effective radionuclide Ytterbium-169 (Yb-169) is currently researched as a possible replacement of Iridium-192 in the HDRB technology. Yb-169 is generated by irradiation of a target containing Ytterbium-168 (Yb-168) isotope by neutrons in the core of a nuclear reactor. Since metallic samples with Yb-168 are not available yet, it has been proposed to produce radiation sources from a ceramics based on ytterbia (Yb₂O₃) powder enriched with Yb-168 isotope. Taking into account that Yb-168 enrichment in ytterbia powder can be up to 40% now, it is necessary to develop the ceramics with the highest possible density to achieve its high specific activity. At atmospheric pressure, Yb₂O₃ has cubic structure with the density of 9.2 g/cm³. In available works on the production of ceramics from Yb₂O₃ (Powell B.R. et al., 1971; Son K.J. et al. 2006), powders of this oxide were initially formed at relatively high pressures, and then sintered at high temperatures. In the first work, the forming pressure was 0.35 GPa, and the maximum density of the ceramics (8.7 g/cm³) was achieved by 30-minutes sintering of the formed sample at 2200° C. In the second work, the forming pressure was approximately 1.3 GPa, and the sintering was performed at 1150 ° C for 3 hours. Sintered samples were cylinder-shaped with the diameter of 1 mm and the length of 1 mm. Their density was only 6.0 g/cm³, too low to allow their effective usage in the HDRB technology.

The article describes an alternative method for production of small-size high density ceramic samples from Yb₂O₃ powder by its sintering at high pressures.

2. METHOD, EQUIPMENT, AND RESULTS

First, initial ytterbia powder with cubic structure is formed in a metal mold at the pressure of 0.1-0.5 GPa without addition of any plasticity agents. Internal diameter of the molds is selected depending on the objectives of the experiment. A mold with the internal diameter of 3 mm was used for development of the ytterbia sintering technology, a mold with the internal diameter of 1 mm was used for improvement of the sintering technology and determination of the sample shrinkage, and a mold with the internal diameter of 0.8 mm was used to produce deliverable samples. Fig. 1 shows a photo of the mold with the internal diameter of 0.8 mm consisting of a matrix and two dies. The upper surface of the matrix is spherical to facilitate pouring fine powder into its hole. The matrix is made of a hard alloy, and the dies are made of a special hardened steel.



Fig. 1

Next, the sample formed undergoes thermobaric treatment in a toroid-type high pressure chamber (Khvostantsev L.G., Vereshchagin L.F., Novikov A.P., 1977) with the 15-mm diameter of its central impression. Lithographic stone container with the reaction cell is placed between two profiled hard-face anvils of the high-pressure chamber. One of the two anvils of the high-pressure chamber and lithographic stone containers used in the experiments are shown on Fig. 2.



Fig. 2

Fig. 3 shows arrangement of the reaction cell for the chamber used for Y_2O_3 sintering. Lithographic stone container (1) with the internal hole with the diameter of 7 mm and graphite heaters (4) with the inner diameter of 5 mm is lathed on a lathe. End heaters made from the mixture of hexagonal graphite and hexagonal boron nitride with the weight ratio 1:1, tablets of hexagonal boron nitride powder (3), and the tablet (5), which consists of either hexagonal boron nitride powder or the mixture of hexagonal boron nitride powder and sodium chloride powder (with the weight ratio 1:1), are pressed in specially manufactured standard molds. Tablet 5 is used as a medium transferring the pressure to sample.

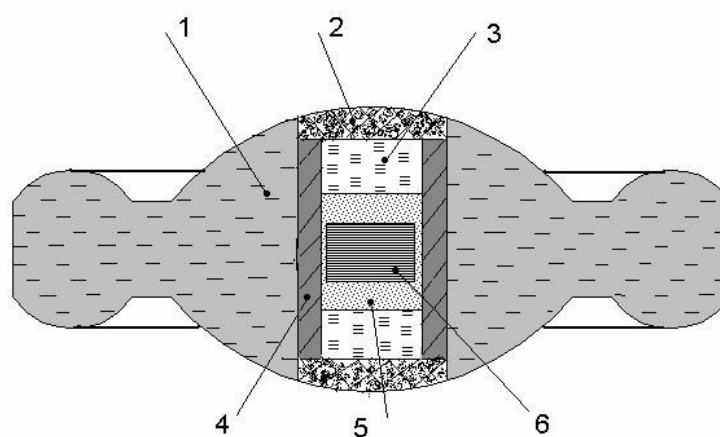


Fig. 3

The above-mentioned mixture of powders for this tablet is selected to facilitate extraction of the sample after its thermobaric treatment. After its extraction from the reaction cell, the tablet with the sample is submerged into hot water and, after a short period of time, it dissolves completely, releasing the sample.

The sample is oriented horizontally in the tablet (to reduce the effects of the thermal gradient). To achieve that, a horizontal hole is drilled in the middle section of the tablet with the diameter equal to the sample diameter. Fig. 4 shows a photo of one of such tablets.

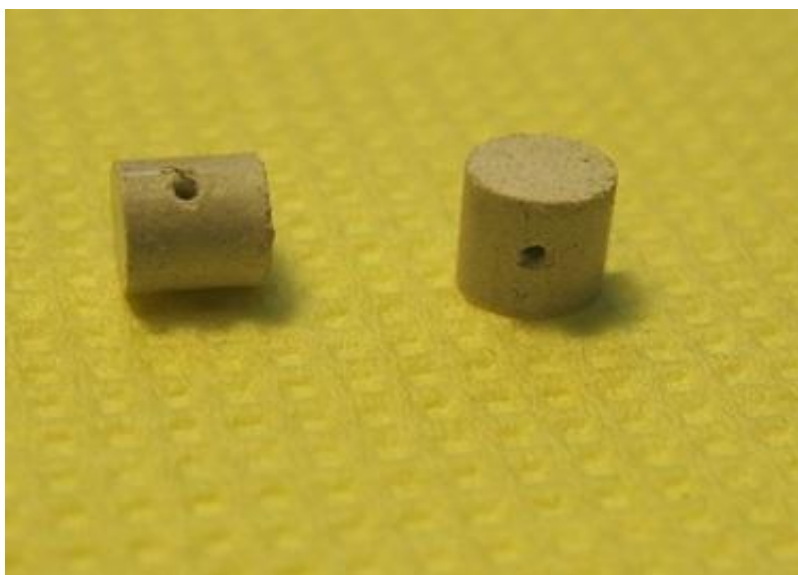


Fig. 4

Our experiments were performed in a 2000 t press. Fig. 5 shows a photo of the working area of the press. Prior to the experiments, high-pressure chambers were calibrated vs. pressure (by using phase transitions in Bismuth) and vs. temperature (by measuring melting temperatures of reference metals with a chromel-alumel thermocouple).

The high pressure sintering technology was tested using the samples formed in a mold with the diameter 3 mm. Fig. 6 shows a photo of two sintered samples formed in this mold.

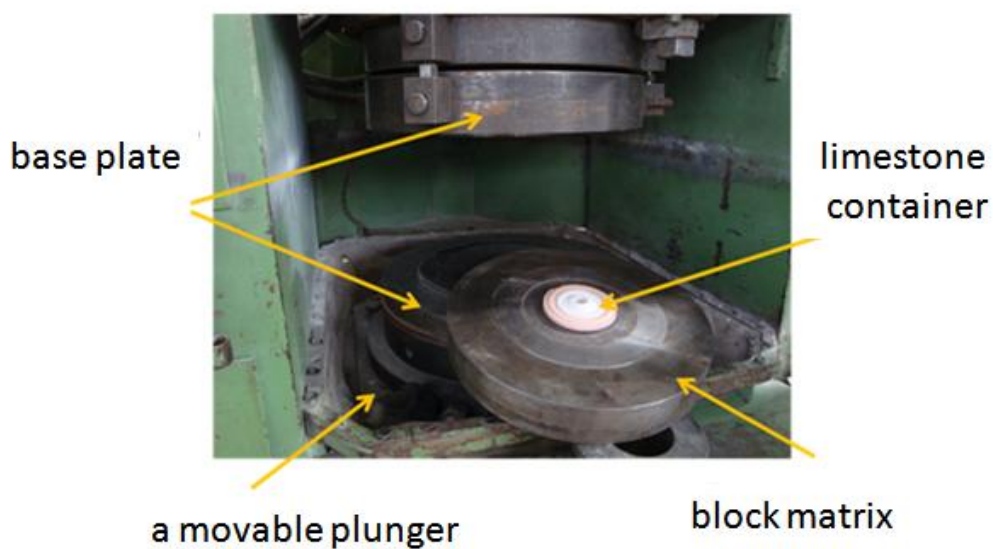


Fig. 5



Fig. 6

The next series of experiments aimed at the improvement of the sintering technology was performed on the samples formed in the molds with the diameter of 1.0 mm or 0.8 mm. A photo of one of these sintered samples (formed in the mold with the diameter of 1.0 mm) is provided on Fig. 7.

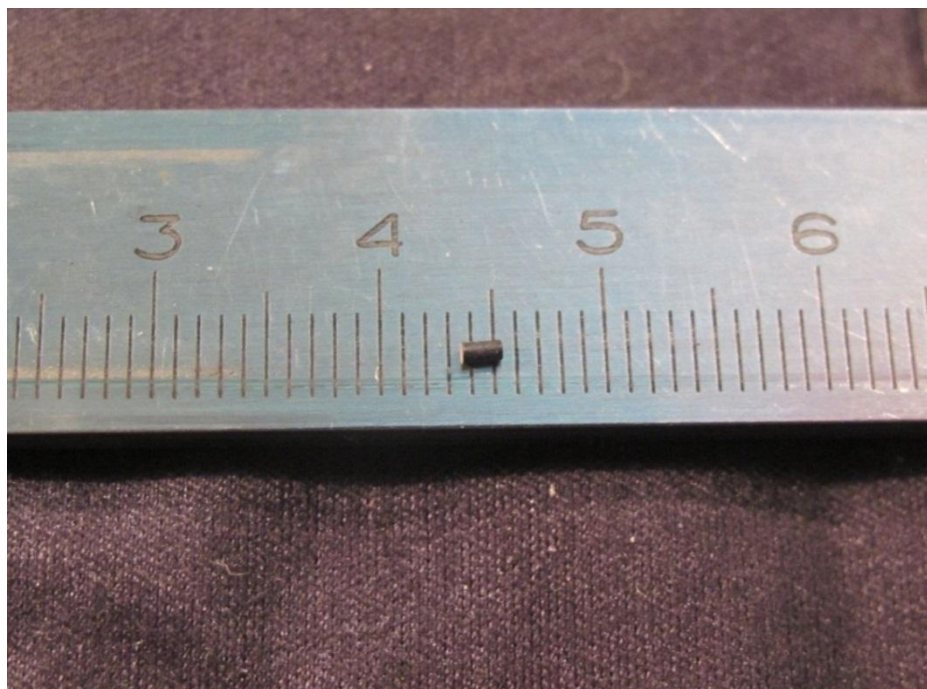


Fig. 7

Usage of high pressures in the sintering process is advantageous because it also allows to produce ytterbia modification with the monoclinic structure (Hoekstra H.R., Gingerich K.A., 1964; Hoekstra H.R., 1966; Herman V.N., Podurets A.M., Tarasova L.F., 1982) with the density 10.04 g/cm^3 , whereas its original modification with cubic structure is 10% more light (9.247 g/cm^3).

Fig. 8 contains a reprint of 1000°C isotherm for rare-earth oxides from the work (Hoekstra H.R., 1966), which clearly demonstrates that ytterbia converts from the cubic phase (C) into the monoclinic phase (B) at this temperature at pressures $>3 \text{ GPa}$.

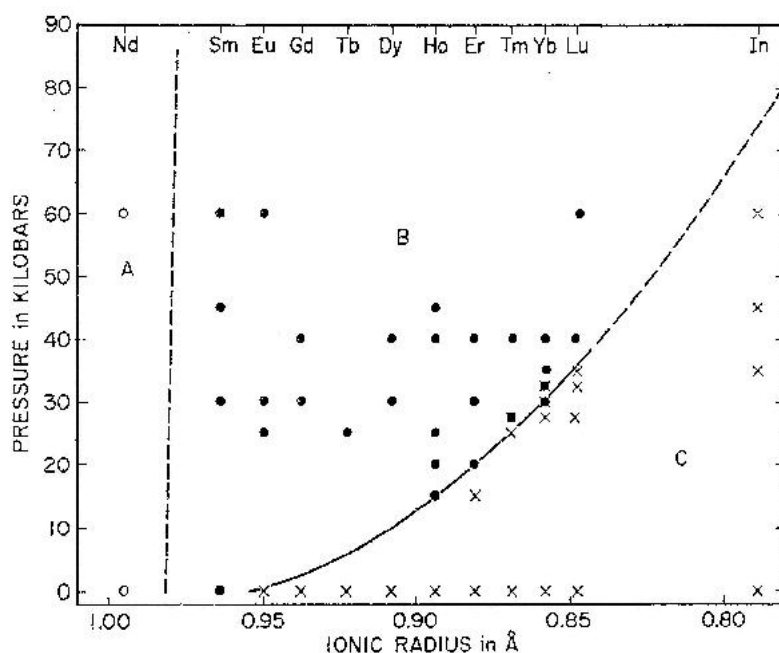


Figure 4.—Rare earth sesquioxides: 1000° isotherm.

Fig. 8

Sintered samples with cubic structure had light color, whereas sintered samples with the more dense monoclinic structure had dark color.

To evaluate the samples after the thermobaric treatment, we measured their density by means of hydrostatic weighing, determined their phase composition by means of X-ray techniques, and analyzed crystalline particle sizes and presence of impurities by means of SEM.

Three examples of ytterbia ceramics production at high pressures are provided below.

Example 1.

A cylinder-shaped sample with the diameter of 1 mm and the length of 2.0 mm was formed from ytterbia powder in a metal mold at 0.5 GPa. The sample was placed into a graphite pot with a cover filled with hexagonal boron nitride, and was annealed in a resistance furnace at 400° C for 2 hours.

After the annealing, the pot containing a tablet of ytterbia was placed in a lithographic stone container, and underwent thermobaric treatment in a high pressure chamber at 4.0 GPa and 1100° C for 60 seconds. The speed of its heating was 50° C/s, and the speed of its cooling was 100° C/s. After the thermobaric treatment, the density of the sample was 9.1 g/cm³; its phase composition was 100% of the cubic phase; the color of the sample was light; the size of its crystalline particles was up to 20 μm (Fig. 9).

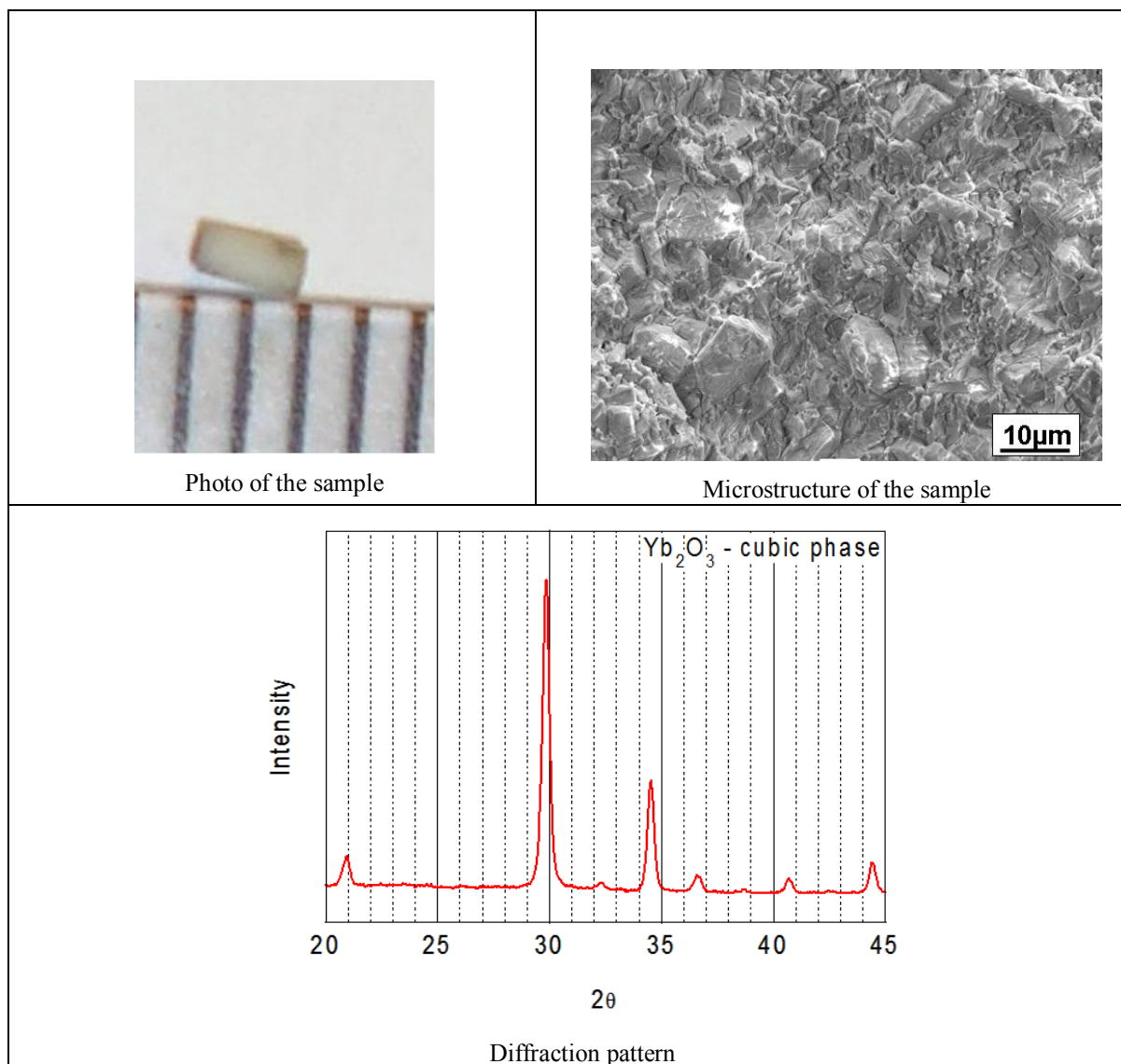


Fig. 9

Example 2.

A sample was prepared for thermobaric treatment by the same method as described in Example 1.

The treatment was performed at 8.0 GPa and 1200° C for 15 seconds, with the heating speed 20° C/s, and the cooling speed 100° C/s. After the thermobaric treatment, the density of the sample was 9.4 g/cm³; its phase composition was 60% of the monoclinic phase + 40% of the cubic phase; the color of the sample was grey; the size of its crystalline particles was up to 5 µm.

Example 3.

A sample was prepared for thermobaric treatment by the same method as described in Example 1.

The treatment was performed at 8.0 GPa and 1000° C for 5 seconds, with the heating speed 100° C/s, and the cooling speed 100° C/s. After the thermobaric treatment, the density of the sample was 10.0 g/cm³; its phase composition was 100% of the monoclinic phase; the color of the sample was black; the size of its crystalline particles was 0.05-0.15 μm (Fig. 10).

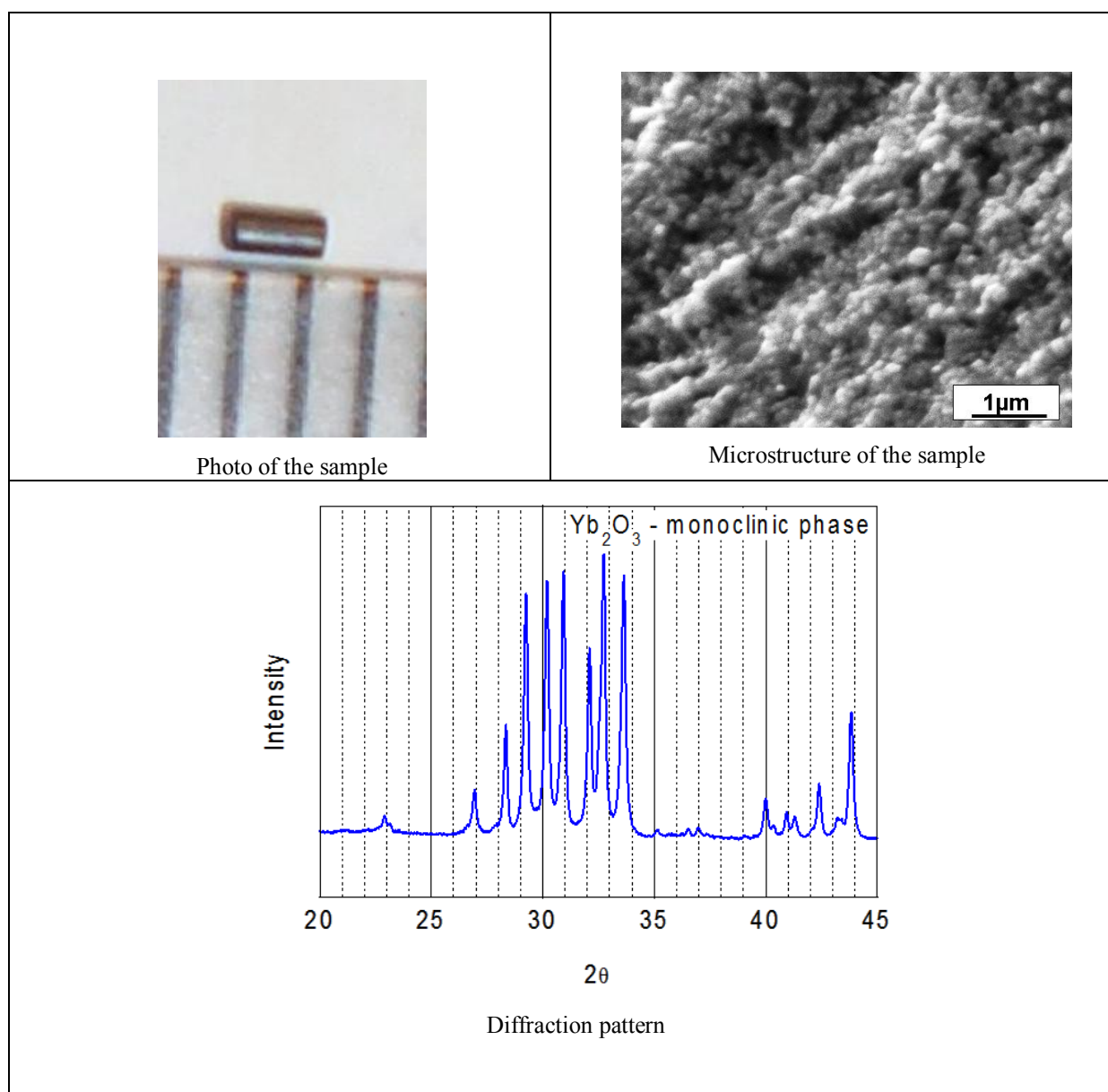


Fig. 10

3. CONCLUSION

We demonstrated viability of the high pressure method for production of high-density ceramics from ytterbia.

ACKNOWLEDGEMENTS

This work was supported by the Russian Foundation for Basic Research, project no. 11-03-00308.

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