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Development of materials and fabrication of porous and pebble bed beryllium multipliers

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Abstract

Beryllium is considered to be a neutron multiplier material for the reference ITER breeding blanket. The main requirements for the porous beryllium multiplier for the breeding blanket are: (1) inherently open porosity within 20 ± 2% for easy removal of radioactive gases; (2) high thermal conductivity; (3) close contact with a stainless steel (SS) shell to provide high heat transfer. A beryllium multiplier can be fabricated by two different techniques: by manufacturing porous or pebble bed beryllium. The method designed (patent 2106931 RU) in SSC RF-VNIINM (Russia) provides for the production of porous beryllium conforming to the requirements mentioned above. For comparative fission tests and the optimization of breeding zone functional capabilities, porous (21.9%) and binary pebble bed (density = 78%) beryllium multipliers were fabricated. DEMO breeding blanket models and a mock-up of fission (IVV-2M reactor) tests have been manufactured at SSC RF-VNIINM. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

One of the basic problems in the development of a helium-cooled, ceramic breeder blanket for the DEMO or of the blanket for ITER is the experimental verification of the design, material-science and technological solutions, including reactor tests of the models. For these purposes, in the Russian State Unitary Enterprise Research and Development Institute of Power Engineering and the SSC of the Russian Federation All-Russia Scientific Research Institute of Inorganic Materials (SSC RF-VNIINM), a model was constructed of a unit of the breeding zone of the helium-cooled blanket of DEMO with tritium-breeding ceramics and a beryllium neutron multiplier [1,2]. This report presents the properties of materials and methods of production used by the SSC RF-VNIINM for manufacture of the models of the breeding zone.

2. Some performance requirements of a material for a porous beryllium multiplier

A beryllium multiplier can be manufactured by two methods:

1. low-temperature pressing (LTP) of porous beryllium with open porosity, designed and licensed in SSC RF-VNIINM;
2. filling of the neutron multiplier zone with granules of beryllium with a binary size distribution.

2.1. Manufacture of porous beryllium

Investigations and technological experiments have demonstrated the feasibility of manufacturing porous beryllium in one stage by filling with a mixture of $Be + BeH_2$ to obtain a finished product with preset geometry and porosity [3]. We designed and tested a two-stage method of manufacturing comparison samples and a porous product. In the first stage, pressing was used to make separate elements with a preset geometry from a $Be + BeH_2$ mixture. In the second stage, pressing of separate elements was done at a particular temperature to obtain a finished product with open

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porosity. As a result of decomposition of BeH_2 at temperatures between 250 and 350°C during the second stage, highly chemically active dispersed beryllium was formed. This dispersed beryllium plays the role of a binder between larger particles of powdered beryllium metal, and the hydrogen released ensures a homogeneous microporous structure with fully open pores [3].

We produced comparison test samples that were taken from mock-ups of the manufactured models and measured their functional properties.

2.2. Structure

With the use of a scanning electronic microscope (SEM), the fracture surfaces of porous beryllium samples were explored after tensile tests (Fig. 1). From the fractographs, it was found that pores are uniformly distributed in the samples and mock-ups, and that the porosity formed is characterized by a complex combination of pores of different sizes from 1–2.5 μm up to 25–30 μm and a system of channels joining the pores and providing open porosity.

2.3. Mechanical properties

Samples for tension and compression tests were produced from comparison test samples. Samples for the compression test were 6 mm in diameter and 9 mm in height. Testing of the samples took place at room temperature at a displacement rate of 2 mm/min. Under compression, the average values of UTS and fracture deformation (ε_f) of porous beryllium at room temperature were 315 MPa and 2.2%, respectively.

As in a previous investigation [3], beryllium with open porosity behaves like a typical sintered material and there was a good correlation of mechanical properties with the volume of open porosity.

2.4. Coefficient of linear thermal expansion

Measurement of the coefficient of linear thermal expansion (CLTE) was conducted in a high-temperature dilatometer ('Adamel', France, model DHT-60 with furnace DHT-60) in helium. The heating rate of the samples during measurement was 5–6°C/min. The measurement precision was 1–3%. Measurements were conducted in the temperature range 20–820°C. In this temperature range, three measurements for each sample were conducted. For samples with a porosity 22 ± 0.5%, the average values of CLTE for two temperature ranges were

$$\alpha_{20\text{–}400^\circ\mathrm{C}}\ (1/^\circ\mathrm{C}) = 13.5 \pm 0.5 \times 10^{-6},$$

$$\alpha_{20\text{–}800^\circ\mathrm{C}}\ (1/^\circ\mathrm{C}) = 17.6 \pm 0.5 \times 10^{-6}.$$

As noted earlier, the value of CLTE for porous beryllium is close to the values for a consolidated material [3].

2.5. Thermal conductivity

Thermal conductivity of the porous beryllium was determined in a vacuum of 10^{-4} Pa by measuring the temperature gradient while heating one face of a sample by an electron beam. To obtain a reference point, we measured the thermal conductivity of a sample of hot-pressed, S-65B grade Be produced by Brush Wellman (USA), for which the thermal conductivity is known.

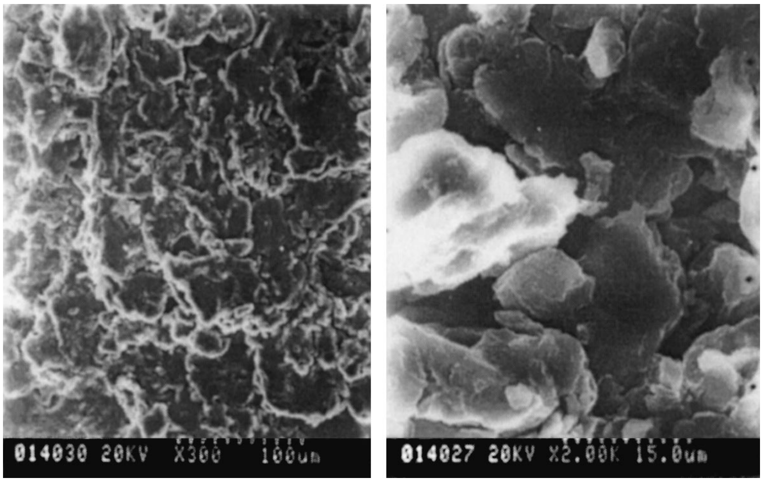

Fig. 1. SEM picture of the structure of fracture surfaces for the comparison of test samples of porous beryllium tested in tension.

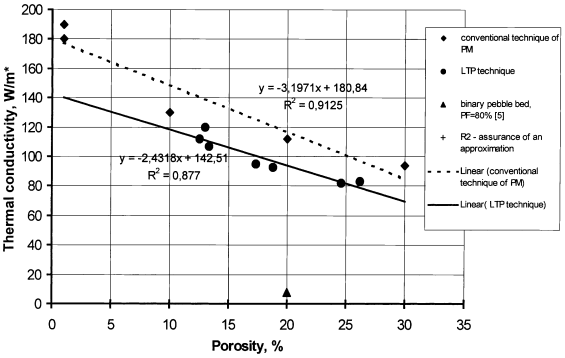

Fig. 2. Thermal conductivity of porous beryllium obtained by conventional methods of powder metallurgy (PM), LTP and binary filling [4], depending on the level of porosity at temperature of 50°C.

The thermal conductivity of porous beryllium depends on the method of manufacture and level of porosity, and is presented in Fig. 2.

3. Manufacture of neutron multiplier from porous beryllium for the model of the breeding zone of a blanket

To manufacture the porous breeder, we used a two-stage technology including heat treatment of pressed intermediate products from a mixture of beryllium powders and beryllium hydride. As a result, we demonstrated the feasibility of manufacturing products from sintered porous beryllium with the following properties:

1. Composite shapes, including those containing channels.
2. Full sintering between separate intermediate products making up the full-scale product.
3. A tight contact between the product and tubular steel shells.
4. A preset level of open porosity within ±5%.

Some examples of the porous breeder produced are presented in Fig. 3. As a structural material for the mock-up and models, ferritic–martensitic steel EP-450 (1Cr12Mo2NbVB) was selected. It possesses thermal fatigue strength, radiation and corrosion resistances, low activation under irradiation and a high level of serviceability in industrial and experimental fast-neutron reactors BN-600, BN-350, BOR-60, etc. [1]. The thin-walled pipes for the outer and inner shells of the models were manufactured from this steel. Methods have been designed for assembling, soldering, welding and checking lithium and beryllium blocks. A series of welding alloys containing no silver were explored, and one of them with a Cu–Ni–Mn composition was used in manufacturing models for high-temperature (1050–1070°C) vacuum soldering. A series of procedures was designed to check the operations and the quality of the models, including hermeticity. Models were certified before reactor tests.

Fig. 3. Neutron multiplier (on the right) and a trial mock-up (on the left) in steel tubes without trailer flanges manufactured by the method of LTP of porous beryllium.

4. Manufacture of materials for the multiplier of neutrons with a filler

To compare the porous multiplier and the European filler multiplier [4], methods were developed to manufacture initial granules of different particle sizes and a pebble-filled beryllium multiplier with a filling density of ≅78–80%. The density was reached by filling with two sizes of beryllium particles, 1.5–2.5 and 0.2–0.3 mm.

Filling a volume with a single size of spherical particles can achieve a maximum density of 67%. However in practice, neither ideal spheres nor absolutely identical size particles, nor their ideal packing can be achieved.

To obtain large particles of beryllium with a typical structure of a dense hot-pressed material without shrinkage cavities, we used conventional pelletizing of beryllium grit. Filling tubes with these particles by Vibrocompaction produced a maximum density of 58% of the theoretical one.

By calculation and experiment it was found that to obtain a more dense filling, it was necessary to use two sizes: 1.5–2.5 mm pelletized particles and 0.25 mm particles. This results in a density gain to ≈78% of the theoretical one.

To manufacture small beryllium particles 0.2–0.3 mm in size, we used centrifugal atomization of beryllium

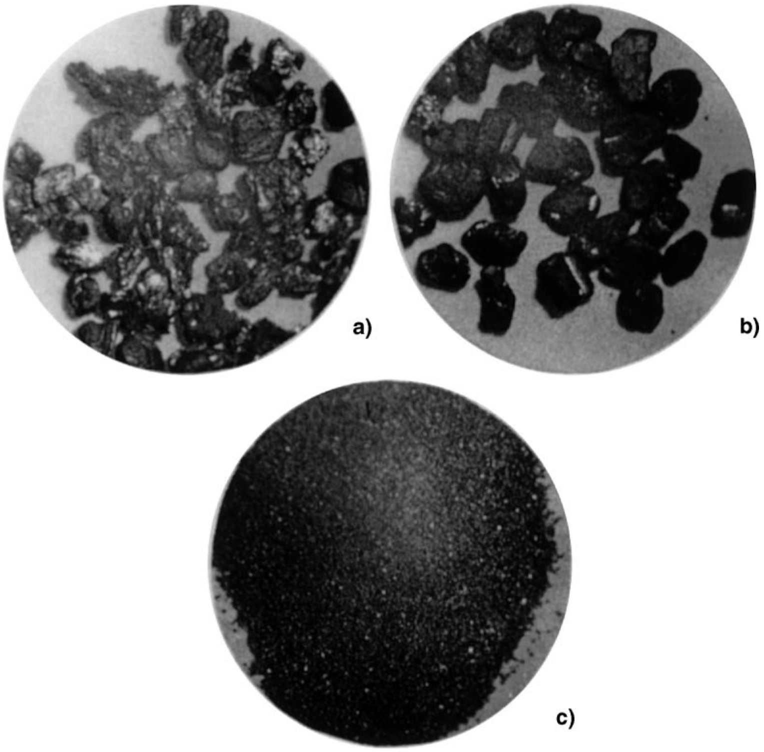

Fig. 4. Initial beryllium granules for binary filling of the zone of neutron multiplier in the model of a breeding blanket: (a) initial grit of beryllium; (b) pelletized granules of beryllium with a particle size of 1.5–2.5 mm; (c) spheroidized powder of beryllium with a particle size of 0.2–0.3 mm.

melted in a rotating crucible in an atmosphere of inert gas, continuously fed with crushed beryllium grit. This resulted in obtaining the initial materials for binary filling of the breeder, i.e., pelletized beryllium granules of 1.5–2.5 mm and spherical granules of 0.2–0.3 mm. Photos of these materials are presented in Fig. 4.

Thus, using the above methods of obtaining porous beryllium and beryllium granules by filling and compacting, we manufactured beryllium multipliers with a density of 78%: two models of a sector of the breeding zone of a blanket, and a mock-up for testing. At present, irradiation experiments of the model are being carried out in the reactor IVV-2M.

5. Summary

Development of methods and manufacture of breeders from porous beryllium and by binary filling have allowed us to estimate the advantages and deficiencies of materials and technologies for their production.

From [3] and results of the present work, the technology of LTP of porous beryllium has advantages when contrasted with the technology for obtaining granulated beryllium. These include the low temperatures, small number of operations, opportunity to conduct the process in air using simple production equipment, high reproducibility of the characteristics of the product, and relatively low price of porous beryllium.

The porous beryllium has advantages compared with filling: the thermal conductivity is 10–15 times higher; close contact with the steel shell is accomplished leading to more effective heat transfer through the contact; maintenance of the structure of the material during vibrations is ensured, and an opportunity for specifying product characteristics is provided.

The method of rotated welding rod atomization to obtain granules requires more complex production equipment and a separate technology to manufacture welding rods with a high accuracy. It has poor efficiency and high cost [4]. The process to manufacture the 1–2 mm particles similar to the process used by Brush Wellman (USA) requires further investigation and has not been completed yet. The cost of manufacturing the large particle fraction is estimated by Brush Wellman to be 670 US \$/kg [4].

Obtaining spherical powder 0.1–0.2 mm in size by atomization has been developed to achieve a production volume up to 45 ton/yr. But the method requires complex equipment, resulting in costs of the small-sized fraction of 500–900 US $/kg [4].

6. Conclusions

1. A waste-free method of low-temperature (250–300°C) pressing of porous beryllium products in air has been designed. The process achieves a preset level of porosity (≈20%) within ±5%.

1.1. The porosity formed in porous beryllium is characterized by a complex combination of pores of different sizes from 1–2.5 μm up to 25–30 μm and a well-developed system of channels connecting these pores to provide open porosity.

1.2. Good reproducibility of structure and properties has been achieved in LTP of porous beryllium. We have demonstrated the feasibility of manufacturing products of complex and tubular shapes, also containing technological channels.

1.3. The average values of UTS and fracture deformation (ε_f) of porous beryllium at room temperature under compression were 315 MPa and 2.2%, respectively.

1.4. For samples with $22 \pm 0.5\%$ porosity, the average values of CLTE for two temperature ranges correspond to values for a dense material and are

$$\alpha_{20\text{–}400^\circ\mathrm{C}}\ (1/^\circ\mathrm{C}) = 13.5 \pm 0.5 \times 10^{-6},$$

$$\alpha_{20\text{–}800^\circ\mathrm{C}}\ (1/^\circ\mathrm{C}) = 17.6 \pm 0.5 \times 10^{-6}.$$

1.5. For a porosity of 19–20%, thermal conductivity of porous beryllium is 95–97 W/m K, which is 11–12 times higher than the thermal conductivity of a binary filler with similar porosity.

2. Two methods have been developed to manufacture granulated beryllium of different particle sizes (0.2–03 and 1.5–2.5 mm), and the technology to achieve a binary filler with a density of 78–80% for the zone of neutron multipliers zone has been demonstrated.

3. Using the designed materials and methods, we have made:

- neutron multiplier from porous beryllium (21.9%) and filler (with density of binary filling 78%);
- two models of a breeding zone of the DEMO, blanket composed of tritium breeding material and beryllium neutron multiplier;
- a mock-up for testing.

At present, irradiation experiments are underway for the model with a binary filling of ceramics and beryllium in the IVV-2M reactor.

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