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Production and characterization of titanium beryllides for HIDOBE irradiation

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Abstract

Preliminary investigations revealed that beryllides like $Be_{12}Ti$ may be much more suitable for neutron multipliers in future fusion power plants than pure beryllium. With suitable mechanical alloying and hot pressing methods, beryllium and beryllium–titanium tablets with grain sizes of 5 μm have been produced and analyzed by means of scanning electron microscopy (SEM), X-ray diffractometry, transmission electron microscopy (TEM) and plasma emission spectrometry. The fabricated tablets of Be–7.7 at.%Ti, corresponding to a chemical composition of the $Be_{12}Ti$ phase, have been arc-melted in an oxygen-free atmosphere. X-ray diffractometry, scanning and high resolution transmission electron microscopy measurements revealed that the vast majority of the ingots consist of nearly pure $Be_{12}Ti$ phase. Nineteen specimens have been fabricated from the ingots and shipped to HFR-Petten for the HIDOBE benchmark irradiation program that started in June 2005 with an aim to achieve 6000 appm He in Be and Be alloys at blanket relevant irradiation temperatures.

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1. Introduction

The reliable and efficient operation of helium cooled ceramic breeder blankets requires the timely availability of beryllium as an appropriate neutron multiplier. In order to overcome some of the disadvantages of existing beryllium pebbles such as tritium release and He embrittlement at blanket operating temperatures, an attempt has been started to systematically investigate relevant properties by producing beryllium and Be-alloys on a laboratory scale at Forschungszentrum Karlsruhe. In the present EU concept, the main strategy for the reduction of the tritium release temperature is (i) to reduce the grain size of Be pebbles and/or tablets, or (ii) to use a suitable Be-alloy.

Japan Atomic Energy Research Institute and Japanese universities have been investigating Be–Ti, Be–V and Be–Mo intermetallic compounds (beryllides) in the view of their use in the Japanese pebble bed blanket concept. Presently, $Be_{12}Ti$ seems to be the most promising beryllide. Compared with pure beryllium, $Be_{12}Ti$ shows faster tritium release, much smaller swelling, less reactivity with stainless steel, steam and water. Therefore, it is becoming interesting for potential use in the EU-HCPB blanket [1].

This paper describes fabrication of $Be_{12}Ti$ samples based on the combination of powder metallurgical and arc-melting methods. The subsequent

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characterization has been performed by means of X-ray diffractometry, SEM and TEM.

2. Experimental equipment and apparatus

For the production of beryllides, the glove-box named BETINA has been fabricated and assembled. In addition, BETINA contains some auxiliary systems equipped with (i) an arc furnace for ingot production of Be-alloys under controlled atmospheres, and (ii) a 20-ton hydraulic press for compacting of metal powders at temperatures of several hundred degrees to a pellet-shaped form.

Glove-box ANABELL was used for preparation of Be-content samples for optical microscopy, SEM, X-ray and TEM analysis. This glove-box system is supplied with a grinding machine for surface preparation of solid samples (etching, polishing, etc). Also from outer side ANABELL has fully automatic annealing furnace for heat treatment of fabricated probes in vacuum or in an inert-gas atmosphere.

3. Process of fabrication

Technological process is based on the use of the arc-melting facility to obtain Be-alloys from the melt. In this case, a tablet-shaped sample pressed out of the Be–Ti compositional mixture (Be–30.8 wt%Ti corresponding to $Be_{12}Ti$ chemical composition) is used as a target to be melted by electrode [2]. Afterwards, samples for HIDOBE have been cut with appropriate tools out of melted probes. Initial powders used for production of $Be_{12}Ti$ samples were beryllium powder (150–200 μm, Brush Wellman) and titanium powder (less than 44 μm, Alfa Aesar).

3.1. Cold pressing

Pressing is necessary to compact a powder mixture to a tablet-shaped form. With a variation of pressing time and temperature, the density of the pressed sample can be adjusted.

A specific pressing tool has been designed and fabricated for the compaction of standard and microcrystalline powders. It is able to produce tablets with 15 mm diameter and 3 mm thickness at temperatures of up to 600 °C with an integrated ohmic heating system. The experience showed that uniaxial powder compaction at 400–450 °C and 450–500 MPa for 120 min is sufficient to achieve densities of up to 90% T.D. (theoretical density).

3.2. Arc melting process

A relatively high speed of material alloying during process is typical of arc-melting. As the first step, the chamber of the arc-melting facility was filled with argon (0,1 bar pressure). Then the pressed Be–Ti tablet was melted by the electrode on a water-cooled copper plate. The main disadvantage of this process is that the electrode and plate can be sources of some additional impurities in melted samples. In addition, experience during the fabrication of many Be–Ti ingots has shown that it is quite difficult to control the temperature within a narrow window. As a consequence, the amount of beryllium evaporated during arc-melting is not always constant, resulting in slightly different alloy compositions. However, the main advantage of this method is that the vast majority of the sample is a beryllide of type $Be_{12}Ti$ with a theoretical density of practically 100%. Only some voids occur accidentally due to the quick solidification process.

4. Characterization of fabricated $Be_{12}Ti$ samples

4.1. X-ray analysis

The analysis of characteristic X-radiation yields quantitative elemental information from the surface of the sample. In the present investigations, X-ray diffraction analysis was performed by means of a D500 X-ray diffractometer.

Fig. 1 shows X-ray data of one of the Be–Ti probe in the as-pressed condition, and Fig. 2 reveals structural changes in this sample caused after arc-melting of the same tablet. Obviously, the cold pressing in the 400 °C range of Ti and Be powders with grains larger than 40 μm does not produce any measurable intermetallics (Fig. 1). However, the completely different X-ray analysis that appear when the Be–Ti tablet is arc melted (Fig. 2) reveals that the vast majority of the sample is single phase $Be_{12}Ti$ with only small inclusions of Be and some other impurities.

4.2. SEM observation

Fig. 3 shows the surface structure of the arc-melted Be–Ti sample, revealing a flat and dense structure almost without any visible voids and porosity. The black, 'needle type' inclusions are Be-rich, and the small white inclusions that sometimes show up in the vicinity of black inclusions

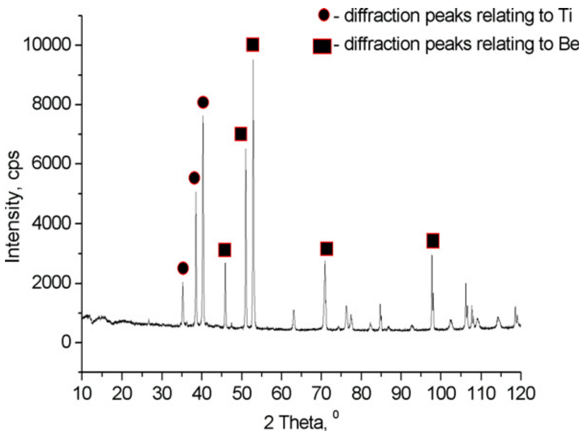

Fig. 1. X-ray diffraction of the cold-pressed Be–Ti sample (1/4 of the page).

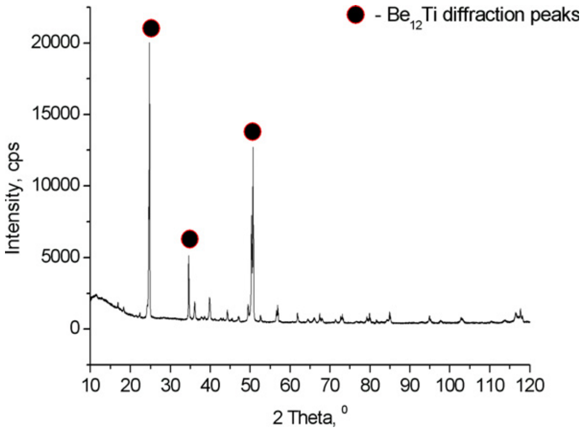

Fig. 2. X-ray diffraction of the arc-melted Be–Ti probe (1/4 of the page).

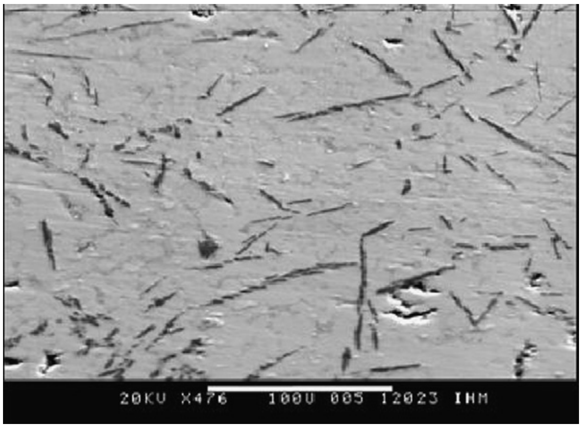

Fig. 3. 'Needle' structure of titanium beryllide (1/4 of the page).

are Ti-rich. The vast majority, however, is uniformly grey, indicating a single phase intermetallic alloy.

4.3. TEM analysis of $Be_{12}Ti$ probes

Several $Be_{12}Ti$ samples were prepared for Transmission Electron Microscopy (TEM) investigation in the form of disks having 300 μm thickness and 3 mm diameter. The TENUPOL device using $H_2SO_4 + 80\%\ CH_3OH$ electrolyte was used for the electro-chemical polishing of the sample. The samples were polished until a small hole was formed in the middle of the disk.

An accurate determination of Be–Ti phase was performed by a High Resolution TEM (HRTEM) analysis. Fig. 4(a) shows a HRTEM micrograph of Be–Ti material. The fast fourier transformation (FFT) image of this HRTEM micrograph presented in the Fig. 4(b) shows three different atomic planes with 0.526 nm, 0.37 nm and 0.37 nm spacing. This corresponds to the [1 1 1] orientation of $Be_{12}Ti$ (tetragonal structure with $a = 0.7278$ nm and $c = 0.424$ nm) perpendicular to the sample surface. All circles and crystallographic indexes were calculated for $Be_{12}Ti$ tetragonal structure by the program Carine Crystallography 3.1. As it can be seen in the FFT image (Fig. 4(b)), the calculated and measured structures are in excellent agreement [3].

The differences in the calculated and experimental positions of the diffraction spots were in the range of 1–2%.

5. Fabrication of $Be_{12}Ti$ specimens for HIDOBE Program

The high dose irradiation program HIDOBE was started in June 2005 in the High Flux Reactor in Petten within the frame of the European Program for the development of the helium cooled pebble bed (HCPB) to study the irradiation behavior and tritium release properties of various international beryllium products at blanket relevant irradiation temperatures. Long irradiation campaigns of 2 and 4 years are foreseen, providing a helium gas concentration of 3000 and 6000 appm He, respectively, corresponding to about one third of the lifetime of a DEMO reactor blanket [4].

Nineteen $Be_{12}Ti$ specimens with 3 mm diameter and 3 mm thickness were cut out of intermetallic ingots. These ingots were produced by combining powder metallurgy and arc-melting technology:

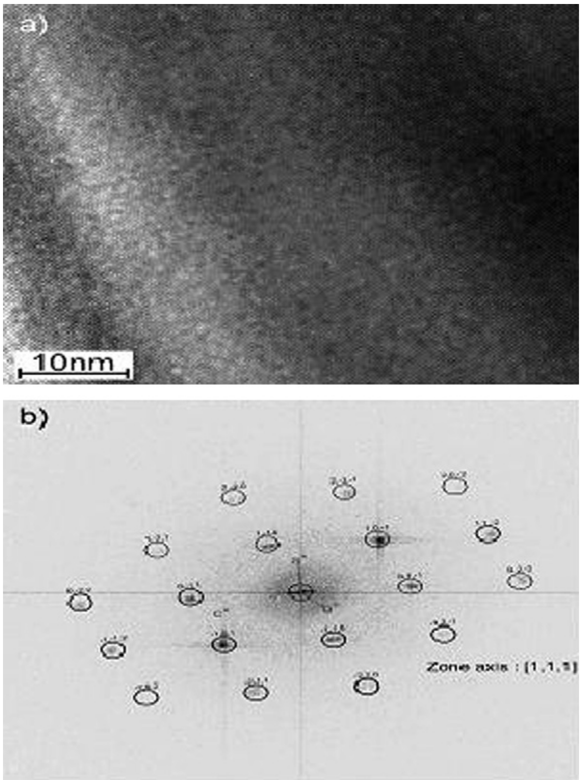

Fig. 4. HRTEM micrograph micrograph of a $Be_{12}Ti$ grain oriented with [1 1 1] zone axis (a) and FFT image of this micrograph with calculated reciprocal lattice pattern of $Be_{12}Ti$ tetragonal structure, (b) (1/4 of the page for both images).

- cold uniaxial pressing of Be–Ti powder mixture (Be–30.8 wt%Ti);
- arc-melting in inert gas atmosphere of cold-pressed Be–Ti tablets.

Before shipping to the HFR Reactor in Petten, porosities and weight of the fabricated samples have been measured. X-ray analysis has also been performed to make qualitative and quantitative analysis of the obtained Be–Ti phases (mainly $Be_{12}Ti$). A picture of the quartz tube filled with some $Be_{12}Ti$ probes delivered to HFR Reactor is shown in Fig. 5.

Fig. 5. $Be_{12}Ti$ specimens sealed in the quartz tube delivered to High Flux Reactor, Petten, Netherlands (1/4 of the page).

6. Conclusions

In this study, the process of fabricating $Be_{12}Ti$ samples, corresponding to an atomic concentration of Be–7.7 at.%Ti, has been carried out by cold pressing of powders followed by arc-melting in an oxygen-free atmosphere. According to the requirements of the HIDOBE rigs, cylindrical samples with 3 mm diameter and about 2–3 mm thickness have been cut from the arc-melted ingots. X-ray analysis, scanning and transmission electron microscopy measurements revealed that the dense cylinders consist mainly of $Be_{12}Ti$ phase.

References

[1] A. Cardella, E. Rigal, L. Bedel, Ph. Bucci, J. Fiek, L. Forrest, L.V. Boccaccini, E. Diegele, L. Giancarli, S. Hermsmeyer, G. Janeschitz, R. Laesser, A. Li Puma, J.D. Lulewicz, A. Möslang, Y. Poitevin, E. Rabaglino, J. Nucl. Mater. (2004) 133, 329.

[2] H. Okamoto, P.R. Subramanian, L. Kacprzak (Eds.), Second edition, Binary Alloy Phase Diagrams, 1, ASM International, 1991, p. 704.

[3] E. Gillam, H.P. Rooksby, Acta Cryst. 17 (1964) 762.

[4] J.B.J. Hegeman, J.G. van der Laan, et al. in: Proceedings of the 6th IEA International Workshop on Beryllium Technology for Fusion, Miyazaki, Japan, p. 141.