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Journal of Nuclear Materials 329–333 (2004) 112–118
journal of nuclear materials
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Present status of beryllide R&D as neutron multiplier

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Abstract

Since 1997, beryllide application as the neutron multiplier has been proposed in Japan and preliminary investigations revealed that some beryllides such as $Be_{12}Ti$ have excellent properties compared with beryllium metal. Recently, several international R&D programs were performed. Some pebbles of modified $Be_{12}Ti$ that included the Be phase were obtained by the rotating electrode method. Basic characterization using HIPped $Be_{12}Ti$ has also been performed. Preliminary experiments indicate that compatibility with stainless steel and tritium breeder materials, deuterium release, natural oxidation and oxidations by steam interaction were better than the behavior of beryllium. Preliminary neutron irradiation effects were studied in JMTR and by charged particle irradiation. It was observed that $Be_{12}Ti$ has less radiation damage than beryllium. In a future experiment, Japanese $Be_{12}Ti$ samples will be included in a long European irradiation program on beryllium to produce up to 6000 appm He. This is an IEA collaborative experiment, to evaluate neutron irradiation effects.

1. Introduction

Beryllium metal in a form of pebbles is the reference neutron multiplier material for most thermo-nuclear fusion breeding blanket concepts. A suitable semi-industrial pebble bed fabrication technology has been developed and a detailed characterization of the material produced has been performed [1]. However, the result of the characterization experiments indicate that beryllium metal presents some disadvantages for high temperature application like DEMO reactor. The operating temperature is estimated to be 600–900 °C in some Japanese DEMO reactor concepts [2,3]. Beryllium metal has a relatively low melting point and high chemical reactivity at high temperature. Therefore research on advanced materials has been initiated and beryllides have been considered as the candidate material in Japan.

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doi:10.1016/j.jnucmat.2004.04.297

2. Preliminary investigation and material selection

2.1. Target

The application conditions for the DEMO blanket are shown in Fig. 1. DEMO blanket requires the neutron multiplier to withstand high temperature (600–900 °C) and high helium generation (~20 000 appm) by nuclear transmutation [2,3]. Therefore an advanced material for DEMO blanket should have high melting point and low swelling by helium. Before starting a feasibility study, candidate materials were selected by JAERI based on melting point, beryllium content, radio-activation and oxidation. The chosen candidate materials were $Be_{12}Ti$, $Be_{12}V$ and $Be_{12}Mo$ as shown in Fig. 1 [4]. Ti, Mo and V give lower radio activation and high melting point. $Be_{12}X$ structure gives good oxidation resistance and high beryllium content for multiplier function [5]. Also, $Be_{12}Ti$ has the lowest melting temperature in these candidate materials, suggesting that it is the easiest to fabricate. For these reasons, $Be_{12}Ti$ was selected as the first candidate material and R&D has been carried out on this material [6,7].

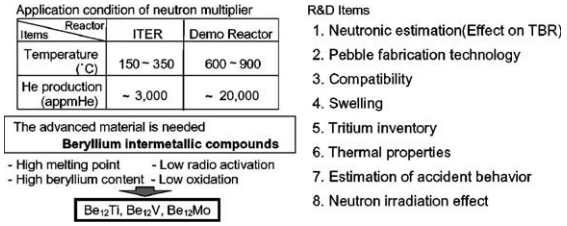


Fig. 1. Outline of beryllide R&D.

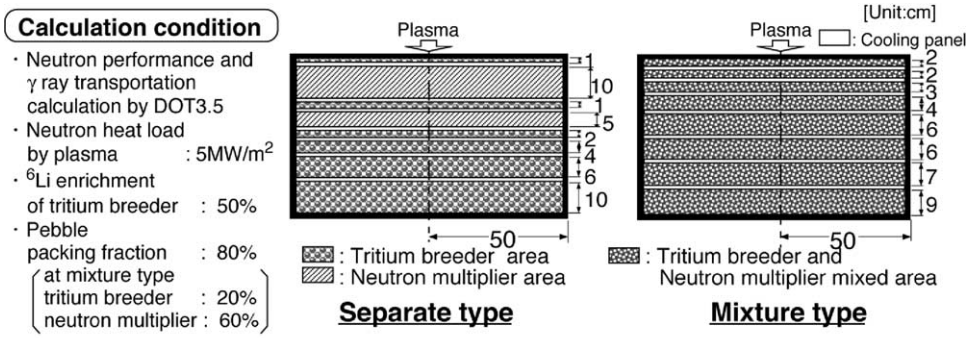
2.2. Neutronic estimation

The evaluation of tritium breeding ratio (TBR) using beryllide as a neutron multiplier was carried out using two models that were separated material packing and mixed material packing (tritium breeder and neutron multiplier). The tritium breeder was Li_2TiO_3 of 85%TD (Theoretical density). and 50 at.% ^6Li enrichment. The packing fraction of pebble beds was 80%. DOT3.5 code and FUSION-40 (based on JENDL3.2) were used for the calculations. The neutron wall load was 5 MW/m^2. Assumed temperatures in the blanket was the same as current blanket designs. The results of the TBR evaluation are shown in Fig. 2. The TBR of the blanket with $Be_{12}Ti$ pebbles was only 10% smaller than that with Be pebbles. It is considered that this value is within the design window and an improvement due to design optimization is expected. The TBR of a mixed pebble bed of tritium breeder and neutron multiplier is estimated to be better than that of the current separated materials blanket design using beryllium metal [8].

3. Technology developments for pebble fabrication

3.1. Phase diagram study

Phase equilibria in the Be–Ti and Be–V binary systems were investigated. Be–(5–23)%Ti and Be–(5–25)%V (at.%) alloys were prepared using an arc melting furnace. Backscattered electron (BSE) images of as-cast and annealed (1200–1450 °C) microstructures were examined and compositions of each phase were measured



Results of calculation

Blanket structure	Separate type			Mixture type		
Neutron multiplier	TBR	atomic number density		TBR	atomic number density	
		Li	Be		Li	Be
Be	1.27	7.4×10^{28}	1.6×10^{29}	1.32	2.9×10^{28}	3.3×10^{29}
$Be_{12}Ti$	1.12	7.4×10^{28}	1.3×10^{29}	1.23	2.9×10^{28}	2.8×10^{29}
$Be_{12}W$	0.77	7.4×10^{28}	1.0×10^{29}	0.72	2.9×10^{28}	2.1×10^{29}

Fig. 2. Results of neutronic estimation – effect on tritium breeding ratio.

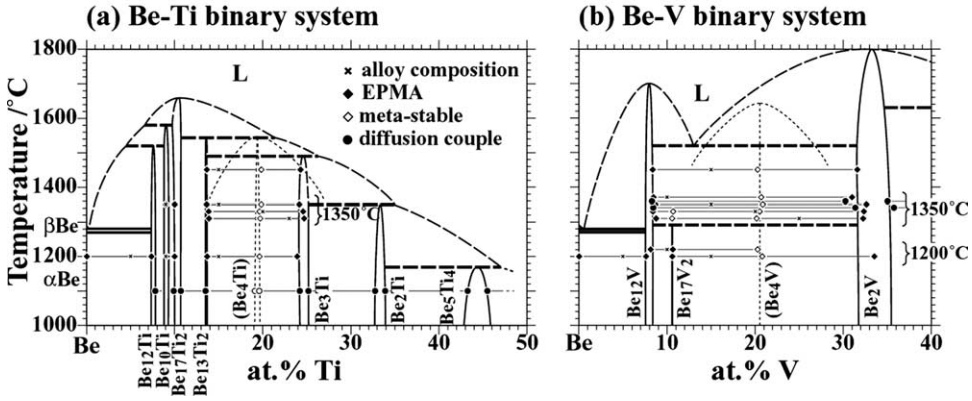


Fig. 3. Phase diagrams of (a) Be–Ti and (b) Be–V binary systems.

by EPMA (JEOL: JXA-8100), where an LDE3H crystal was used for analyzing Be. Samples in capsules were kept in a furnace at 1350 °C for Be/V and at 1100 and 1200 °C for Be–Ti/Be to evaluate liquid/solid diffusion couples. Composition profiles in the diffusion layers were measured by EPMA. Eutectic and peritectic invariant reactions were identified from the as-cast microstructures and equilibrium compositions were determined from the annealed alloys and diffusion couples. The results obtained are summarized in Fig. 3 (a) Be–Ti and (b) Be–V binary systems [9].

3.2. Ductility improvement – (1) fabrication test

It was not easy to fabricate beryllide pebbles by the rotating electrode method, the reference fabrication process for beryllium pebbles [10], because of the brittleness of the electrode rod. Previously, $Be_{12}Ti$ rotating electrodes were fabricated by the HIP process (hot isostatic pressing) and trial pebble fabrication was performed with the rotating electrode method. This approach was not successful because the electrode broke by thermal shock during the arc heating because of the brittleness of the electrode. In order to identify the cause of the brittleness i.e., porosity due to the HIP process, large grain size, original brittleness, etc., the relationship between the microstructure and ductility was studied using the arc melting method. Then, it became clear that (1) the structure produced by the melting process had lower porosity than that with HIP process, (2) it was difficult to reduce brittleness by heat treatment for the stoichiometric composition. In order to improve the ductility by structure control, specimens with 5 at.% Ti, 7.7 at.% Ti (stoichiometric), 9 at.% Ti and 15 at.% Ti were fabricated, then microstructure observation and hardness test were performed. Hardness was 650, 1100, 1160 and 1230, respectively. The microstructure showed that Be–5 at.%Ti had finer structure, as a mixture of the

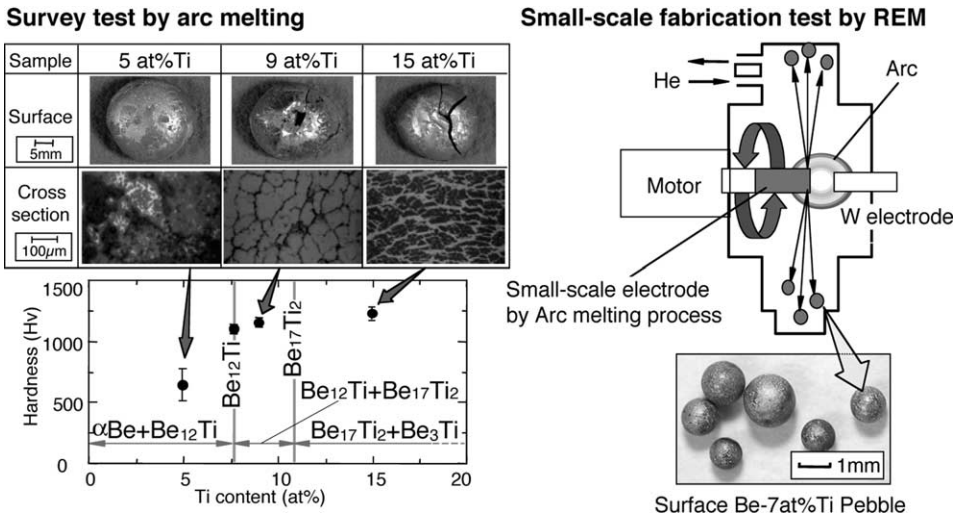


Fig. 4. Results of pebble fabrication technology development.

Table 1
Results of compression tests for $Be_{12}Ti$

Material		RT			1000°C
		0.2% yield (MPa)	Rapture (MPa)	Elongation (%)	0.2% yield (MPa)
Be–3 at.%Ti as-cast	−1	775.4	913.5	1.7	14.9
	−2	678.2	889.0	3.2	24.5
Be–5 at.%Ti as-cast	−1	810.7	888.1	1.1	72.8
	−2	762.8	781.8	0.5	56.1

$Be_{12}Ti$ phase and α-Be phase. Next, small electrodes were fabricated and the rotating electrode method was performed as a thermal shock test. The electrodes withstood the thermal shock from arc heating and some pebbles were obtained. The pebbles were examined and it was clear that the pebbles were dense with a fine structure consisted of $Be_{12}Ti$ phase and α-Be phase, as shown in Fig. 4. These results indicate good prospects for beryllide pebble fabrication [10].

3.3. Ductility improvement – (2) mechanical properties

In order to impart some room temperature ductility to intrinsically brittle $Be_{12}X$ type intermetallic compounds, two-phase alloys including the Be solid solution, hereafter (Be), are prepared in Be–Ti systems and the microstructure-mechanical property relationships are examined. In these alloy systems, the Ti concentration is initially chosen as 3 and 5 at.% and the microstructures are observed for the as-cast condition. The morphology of $Be_{12}Ti$ in Be is irregular in shape. The room temperature compressive strength ranges from 700 to 850 MPa in both alloy systems and naturally it is higher with 5 at.% Ti addition than with 3 at.% Ti. The compressive 0.2% flow stress at 1000 °C is less than 100 MPa in all the alloys but the alloys are fully ductile at the temperature as shown in Table 1 [11].

4. Characterization

4.1. Chemical properties – (1) compatibility

The compatibility of $Be_{12}Ti$ with structural material (SS316LN) and tritium breeder (Li_2TiO_3) at was evaluated out 600, 700 and 800 °C up to 1000 h. The results of the compatibility tests with SS316LN are shown in Fig. 5. It was obvious that the compatibility between $Be_{12}Ti$ and SS316LN was much better than that between Be and SS316LN. The thickness of the reaction layer

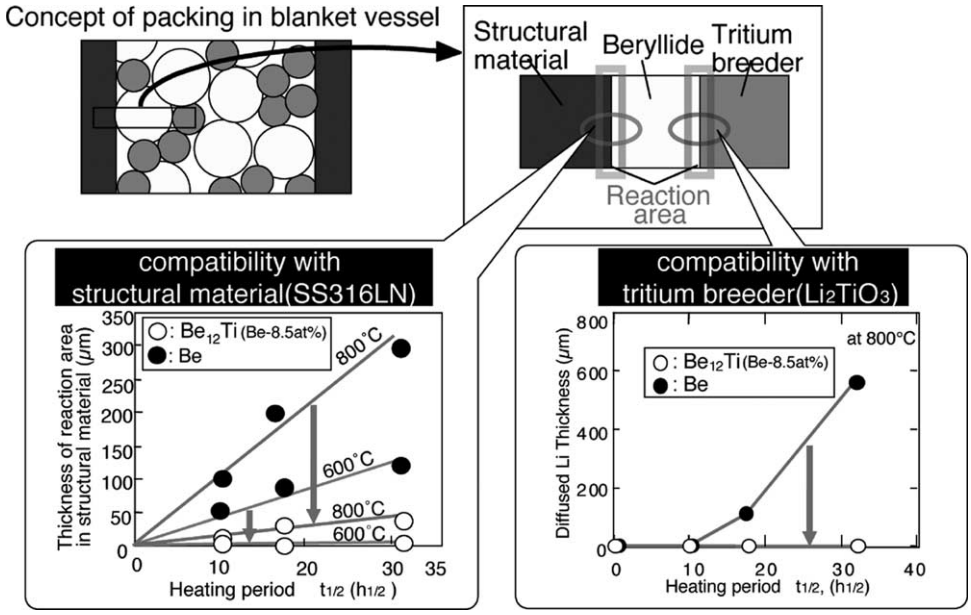


Fig. 5. Results of compatibility test.

between $Be_{12}Ti$ and SS316LN at 800 °C was one tenth of that of Be. For the compatibility between $Be_{12}Ti$ and Li_2TiO_3, reaction products on the $Be_{12}Ti$ and Be in contact with Li_2TiO_3 were not found at any temperatures up to 1000 h. On the other hand, the diffusion of Li into Be was identified at 800 °C for 300 and 1000 h. The results of these compatibility evaluations showed that $Be_{12}Ti$ had the advantages over Be for high temperature use [12,13].

4.2. Chemical properties – (2) oxidation

High temperature oxidation of $Be_{12}Ti$ was investigated at 800 °C by thermogravimetric technique, and compared with Be and Ti. The atmosphere was dry air and oxidation time was up to 24 h. Fig. 6 shows the mass gain curve of $Be_{12}Ti$ at 800 °C. Though Be and Ti show larger mass gain, the mass gain of $Be_{12}Ti$ after 24 h was very small, as small as the mass gain for superalloys or stainless steels at 800 °C. Fig. 6 also shows the surface morphologies of oxidized Be and $Be_{12}Ti$. Be formed a porous and powdery scale. On the other hand, $Be_{12}Ti$ formed a thin and compact scale. X-ray diffraction measurement revealed these scales are BeO. From these experimental results, it is obvious that $Be_{12}Ti$ has good oxidation resistance in air at high temperature. Furthermore, it is assumed that Ti would form a protective BeO film on $Be_{12}Ti$ [14].

4.3. Chemical properties – (3) steam interaction

The reactivity of $Be_{12}Ti$ with water vapor was investigated. In the experiments, sample disks of $Be_{12}Ti$ were placed in a reactor. Argon gas with measured amounts of water vapor was introduced into the reactor, and the reactor temperature was raised to around 1000 °C at constant rates. The concentrations of water vapor, hydrogen and oxygen in the outlet stream of the reactor were measured with a mass spectrometer. Fig. 7 shows an experimental result, indicating that the reaction between $Be_{12}Ti$ and water vapor began to take place at 600 °C and hydrogen was produced. However, the catastrophic breakaway reaction, which is known to take place in the case of beryllium, was not observed. The analysis of the result reveals that the amount of water which reacts with $Be_{12}Ti$, is far smaller in comparison to similar experiments with beryllium. Thus, it can be said that titanium beryllides are less reactive with water vapor [15].

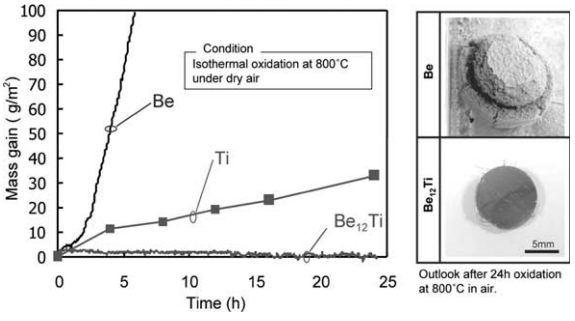


Fig. 6. Results of oxidation properties test.

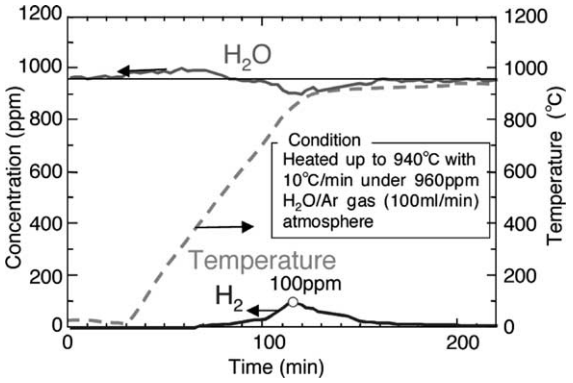


Fig. 7. Results of interaction test of $Be_{12}Ti$ with steam.

4.4. Tritium inventory

The desorption behavior of deuterium from $Be_{12}Ti + Be$ was evaluated by a heating test after deuterium implantation. Some results are shown in Fig. 8. Deuterium was implanted up to 1×10^{21} ions/m^2 at room temperature. The profile of the desorption rate for $Be_{12}Ti$ has a peak at about 100 °C. On the other hand, the peak temperature of desorption rate for Be is higher (350–700 °C) than that for $Be_{12}Ti$. About 20% of the implanted deuterium is retained in Be around 700 °C. These results showed that the deuterium desorption properties of $Be_{12}Ti$ was superior to that of Be. This could indicate that a blanket using $Be_{12}Ti$ would have a tritium inventory much smaller than that for Be. Suitable neutron irradiation experiments should be performed to confirm these indications [16].

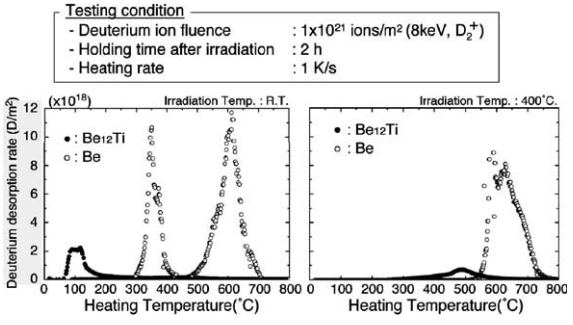


Fig. 8. Results of deuterium release test on heating of implanted $Be_{12}Ti$ and Be.

4.5. Neutron irradiation in JMTR

$Be_{12}Ti$ and Be specimen were irradiated to $\sim 4\times10^{20}$ n/cm^2 ($E > 1$ MeV) at 500 °C in JMTR. Helium production rate and dpa for Be were about 70 appm He and 0.5 dpa, respectively. Swelling was calculated from the dimension and weight measurements for the neutron irradiated $Be_{12}Ti$ disk heated at 1100 °C for 1 h after irradiation. The swelling of $Be_{12}Ti$ was less than 3%. On the other hand, the swelling of Be was ~60%. From these results, the swelling of $Be_{12}Ti$ under high temperature neutron irradiation can be expected to be smaller than that of Be [17]. The tritium release from neutron irradiated $Be_{12}Ti$ disk specimens was studied. The results were (1) tritium inventory of $Be_{12}Ti$ was lower than that of beryllium, and (2) the characteristic phenomenon concerning tritium release might depend upon the surface oxidation and the oxidation was less than that of beryllium (visual inspection). It is desirable to study the surface structure to establish the mechanism of tritium release from $Be_{12}Ti$. According to the results obtained, the swelling of $Be_{12}Ti$ might be much lower than that of beryllium [17]. Thermal conductivity for un-irradiated and irradiated $Be_{12}Ti$ was evaluated and the thermal properties needed for the DEMO blanket design were obtained. The preliminary results show: (1) the degradation of thermal conductivity by neutron irradiation was not recovered by annealing up to 600 °C, but recovered on heating up to 1000 °C, and (2) the effective thermal conductivity of a $Be_{12}Ti$ pebble bed was evaluated with SZB correlation from measured data, and was 10–20% lower than that of Be at 800 °C [18].

4.6. Charged particle irradiation

To evaluate the microstructural response to $Be_{12}Ti$ to fusion equivalent environments, in situ charged particle irradiation experiments were conducted. The experiments used a function of dose and simultaneous He irradiation effects at elevated temperatures up to 700 °C, using the Multi Beam High Voltage Electron Microscope (MBHVEM) in CARET, Hokkaido University. Several precipitates in $Be_{12}Ti$ were analyzed by EDP and EDS and found to be BeO and titanium oxide. After He/electron irradiation at 500 °C, tiny bubbles were observed along the grain boundary in Fig. 9. However, it was hard to detect black dots or dislocation loops in $Be_{12}Ti$ for the same irradiation conditions. Simultaneous He irradiation is found to enhance the nucleation of bubbles. On increasing the irradiation temperature, $Be_{12}Ti$ demonstrated the same trends of the microstructure response as seen for He irradiation at 500 °C. Mechanical property measurements at elevated temperatures are still underway [19].

5. Future plans – IEA irradiation program and air/steam reaction rates

High dose neutron irradiation tests are needed to evaluate the lifetime of $Be_{12}Ti$. Future plan for the irradiation tests are considered as shown in Fig. 10. In the frame of a IEA collaborative action between Japan and Europe, $Be_{12}Ti$ small disks (8 mm diameter, 2 mm thickness) will be irradiated starting in 2004 in the HFR reactor in Petten (NL). The objective of the experiment is to obtain irradiation data (swelling, creep, tritium retention) at DEMO relevant conditions in terms of dpa/He ratio and temperatures. The chosen irradiation temperatures range between 400 and 800 °C. A 30% DEMO end-of-life helium production (6000 appm in four years) should be achieved with an intermediate unloading of some of the samples at 3000 appm. At the maximum dose (40 dpa Be), maximum tritium production of 850 appm is expected. The $Be_{12}Ti$ reaction rate with air/steam at temperatures between 450 and 900 °C will also be measured out-of-pile in Europe in order to have independent verification with another experimental apparatus [20].

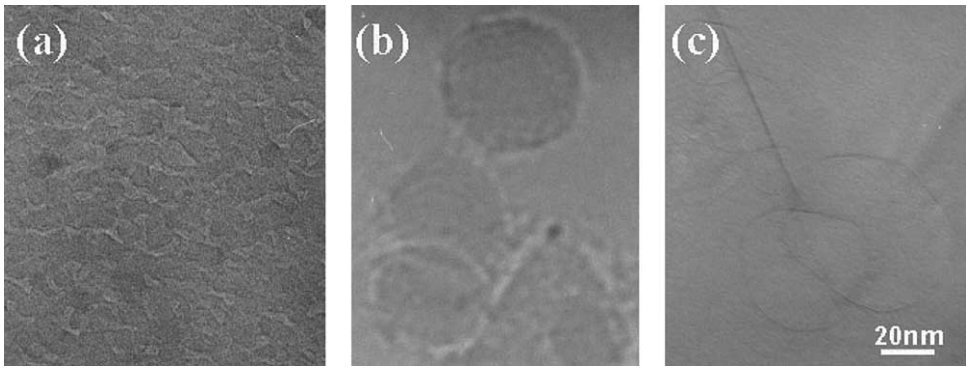


Fig. 9. TEM after dual irradiation in (a) pure Be, (b) $Be_{12}Ti$ at RT and (c) $Be_{12}Ti$ irradiated at 500 °C.

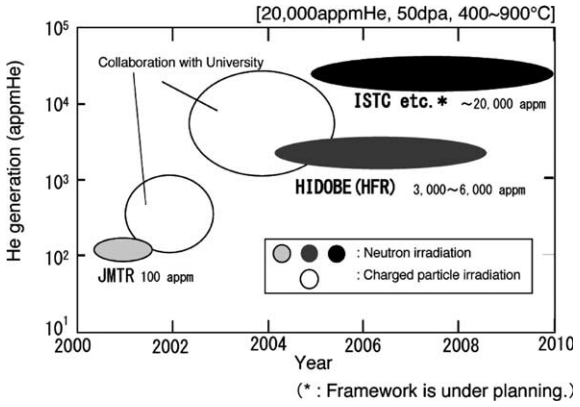


Fig. 10. Scope of HIDOBE test.

6. Conclusion

These results show that there are bright prospects for the application of a beryllide as neutron multiplier for the DEMO blanket. This follows from the good chemical, thermal, mechanical and irradiation properties.

The status on the R&D is as follows:

(1) $Be_{12}Ti$, $Be_{12}V$ and Be_{12} Mo were pre-selected as candidate materials for their low radio activation and high melting point, good oxidation resistance and high beryllium content. Now $Be_{12}Ti$, which was believed to have the easiest production, is being emphasized in continuing work. Estimation of the tritium breeding ratio for a blanket using beryllide was carried out and the TBR of the blanket with $Be_{12}Ti$ pebbles was only 10% smaller than that with Be pebbles. It is considered that this value is within the design window.

(2) Phase equilibria of Be–Ti and Be–V system were investigated and the phase diagrams for high beryllium compositions were obtained. The relationship between the Ti content in Be–Ti and the mechanical property was studied and it was established that a mixture including some α-Be phase is effective to add some ductility. Pebbles with this phase were obtained by small scale REM trial. Also, basic study of the ductility improvement of the $Be_{12}Ti + Be$ using compressive tests is being performed.

(3) Characterization of the compatibility with structural materials and ceramic breeders, oxidation, steam reaction, tritium inventory, neutron irradiation effects in JMTR and the structure evolution during charged particle irradiation have been carried out and the advantages of $Be_{12}Ti$ compared to beryllium metal were made clear.

(4) In future, $Be_{12}Ti$ small disks (8 mm diameter, 2 mm thickness) will be irradiated up to 6000 appm He starting in 2004 in the HFR reactor to obtain irradiation data (swelling, creep, tritium retention) at DEMO relevant conditions of dpa/He ratio and temperatures.

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