2.5 Status of Beryllium R&D in Japan

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Recently, several R&D programs of beryllium for fusion are being promoted in Japan and the community of beryllium study is growing up. In the R&D area of beryllium for solid breeding blanket, major subjects are beryllide application for prototype reactor, lifetime evaluation of neutron multiplier, impurity effect of beryllium and recycling of irradiated beryllium. Especially, the study of beryllide application has significant progress in these two years. The basic properties such as tritium inventory, oxidation behavior, steam interaction for stoichiometric $Be_{12}Ti$ fabricated by HIP have been studied and some advantages against beryllium were made clear. For manufacturing technology development, phase diagram and ductility improvement have been studied. And, $Be_{12}Ti$ pebbles with the improved microstructure were successfully fabricated by Rotating Electrode Process. In order to enhance the R&D activities, the R&D network consisted of industries, universities and laboratories in all Japan have been organized. Many collaboration and information exchange strongly promotes the R&D and some projects for commercial application have been launched from these activities. Also international collaborative project such as IEA and ISTC have been launched or planed. Recent results of R&D in Japan is described on this paper.

1. INTRODUCTION

Beryllium metal in a form of pebbles is the reference neutron multiplier material of most thermonuclear fusion breeding blanket concepts. A suitable semi-industrial pebble bed fabrication technology has been developed and a detailed characterization of the produced material has been performed [1]. However, the result of the characterization experiments indicates 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. 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.

2. BERYLLIUM STUDY

As to beryllium metal, beryllium pebbles that focus the ITER test module condition has been developed as shown in Fig.1. Rotating electrode method was established as fabrication process. And characterizations were carried out up to 3000 appmHe at 400°C. It was confirmed that the developed pebbles shows good performance under ITER test module condition. Remained subjects to be studied for beryllium are lifetime evaluation and recycling of used beryllium. As to the recycling of used beryllium, basic concept has been established as shown in Fig.2. Based on this concept, detailed process has to be established. Also, international scheme to recycle used beryllium has to be made.

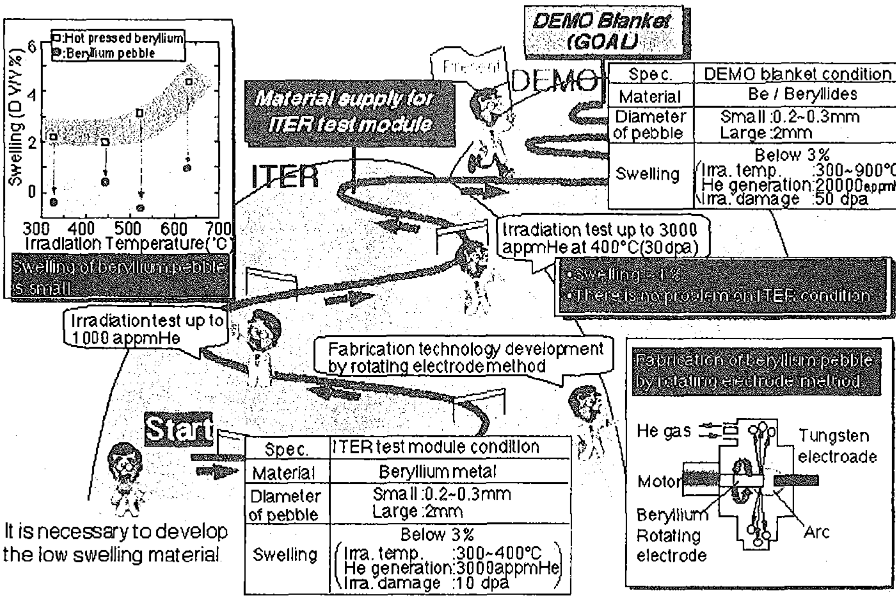


Fig.1 Development of neutron multiplier for DEMO blanket

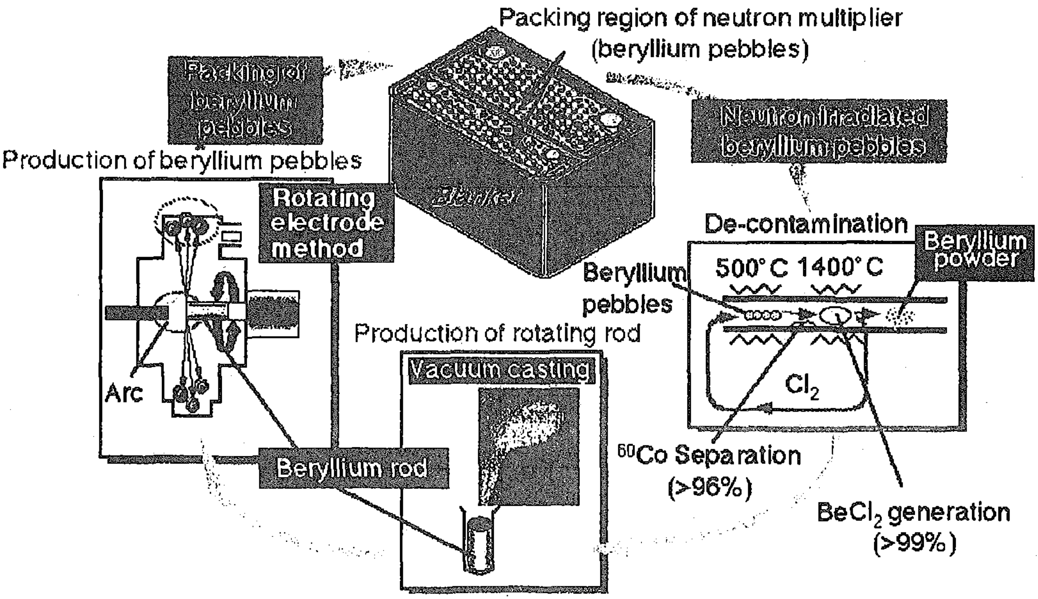


Fig.2 Concept on recycle of beryllium pebbles

3. BERYLLIDE STUDY

3.1 Target and R&D in Japan

The application condition for the DEMO blanket is shown in Fig.3. DEMO blanket requires the neutron multiplier to withstand high temperature (600-900°C) and high helium generation (~20,000appm) 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, the candidate materials have been selected by JAERI from the viewpoint of 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.3 [4]. Ti, Mo and V give the low 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, and it was considered that it is the easiest to be fabricated. From these points, $Be_{12}Ti$ was selected as the first candidate material and several R&D has been carried out on this material [6, 7]. The R&D for beryllide application is studied by many organizations as shown in Fig.4.

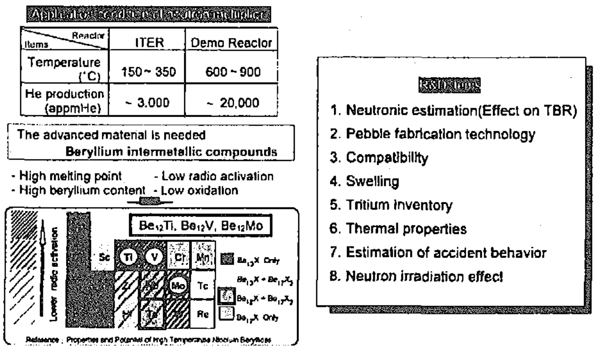


Fig.3 Outline of beryllides study

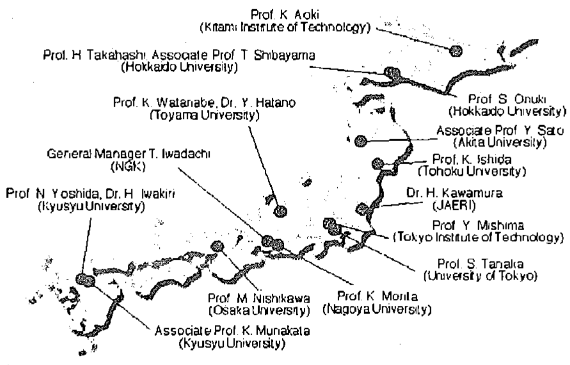
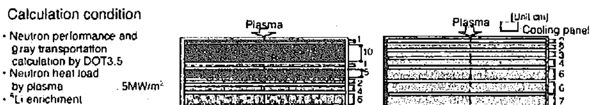


Fig.4 Organization of beryllide study in Japan

3.2 Neutronic estimation

The evaluation of Tritium Breeding Ratio (TBR) using beryllide as a neutron multiplier was carried out using two models that were mono material packing and mixed material packing (tritium breeder and neutron multiplier). The tritium breeder was Li_2TiO_3 of 85%T.D. and 50at% ^6Li enrichment. The packing fraction of pebble beds was 80%P.F. DOT3.5 code and FUSION-40 (based on JENDL3.2) were used for the calculation. The neutron wall load was $5MW/m^2$. Assumed temperature in the blanket was the same as current blanket design. The result of the TBR evaluation is shown in Fig.5. TBR of blanket with $Be_{12}Ti$ pebbles resulted only 10% smaller than that with Be pebbles. It is considered that this value is within design window and an improvement by raising temperature is expected. The TBR of a mixed pebble bed of tritium breeder and neutron multiplier is estimated better than that of current separate blanket design using beryllium metal [8].



Results of calculation

Blanket structure	Separate type			Mixed type		
neutron multiplier	TBR	atomic number density(n) Li	atomic number density(n) Be	TBR	atomic number density(n) Li	atomic number density(n) Be
Be	1.27	7.4x10^[illegible]	1.6x10^[illegible]	1.32	7.9x10^[illegible]	3.3x10^[illegible]
$Be_{12}Ti$	1.12	7.4x10^[illegible]	1.3x10^[illegible]	1.23	2.9x10^[illegible]	2.8x10^[illegible]
$Be_{12}W$	0.77	7.4x10^[illegible]	1.0x10^[illegible]	0.72	2.9x10^[illegible]	2.1x10^[illegible]

Fig.5 Neutronic estimation - Effect on TBR -

3.3 Fabrication technology improvement

It has been not easy to fabricate beryllide pebbles by rotating electrode method that had been reference fabrication process as to beryllium pebble, because of the brittleness of electrode rod. Previously $Be_{12}Ti$ rotating electrode was fabricated by HIP process (Hot Isostatic Pressing) and trial pebble fabrication was performed with the rotating electrode method. But, it was not successful because electrode broke by thermal shock during the arc heating because of the brittleness of the electrode. In order to clarify 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 the ductility was studied with arc melting method. Then, it became clear that 1) the structure with melting process had few porosity than that with HIP process, 2) it was difficult to improve brittleness by heat treatment for stoicheometric composition. In order to improve the ductility by structure control, the specimens with 5at%Ti, 7.7at%Ti (stoicheometric), 9at%Ti and 15at%Ti were fabricated, then microstructure observation and hardness test were performed. Hardness was 650, 1100, 1160 and 1230 respectively. The microstructure showed that Be-5at%Ti had finer structure with $Be_{12}Ti$ phase and α-Be phase. Next, the small electrodes were fabricated and the rotating electrode method was performed as thermal shock test. The electrode withstood against the thermal shock from arc heating and some pebbles were obtained. The pebbles were observed and it was clear that pebbles were dense and had fine structure consisted of $Be_{12}Ti$ phase and α-Be phase as shown in Fig. 6. From these results, bright prospect for beryllide pebble fabrication was obtained [9].

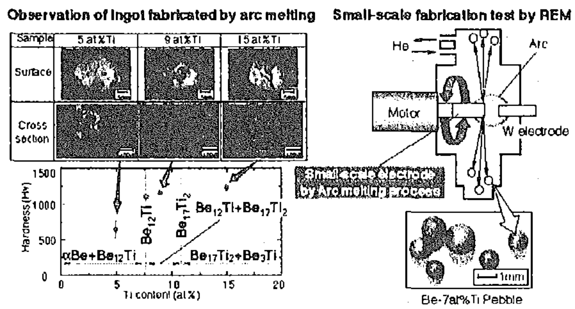


Fig.6 Pebble fabrication technology

3.4 Chemical property (1) -Compatibility-

The compatibility test of $Be_{12}Ti$ was carried out with structural material (SS316LN) and tritium breeder (Li_2TiO_3) at 600°C, 700°C and 800°C up to 1000h by annealing. The results of the compatibility for SS316LN are shown in Fig.7. It was obvious that the compatibility between $Be_{12}Ti$ and SS316LN was much better than that between Be and SS316LN. The thickness of reaction layer 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 1000h. On the other hand, the diffused Li into Be was identified at 800°C for 300h and 1000h. The results of these compatibility evaluations showed that $Be_{12}Ti$ had the advantages for high temperature use [10, 11].

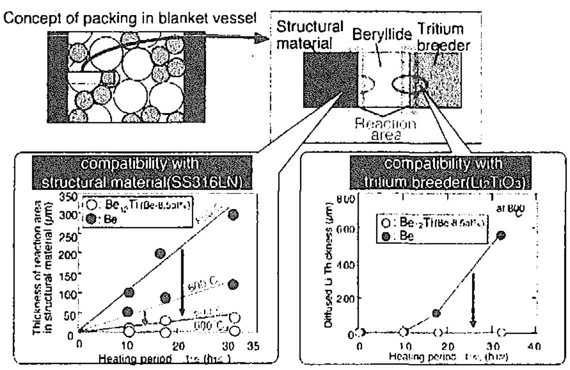


Fig.7 Compatibility

3.5 Chemical property (2) -Oxidation -

High temperature oxidation property of $Be_{12}Ti$ was investigated at 800°C by thermogravimetric technique in contrast with Be and Ti. The atmosphere was selected to dry air and oxidation time was within 24h. Fig.8 shows mass gain curve of $Be_{12}Ti$ at 800°C. Though Be and Ti show lager mass gain, mass gain of $Be_{12}Ti$ after 24h was very small. It is as small mass gain as that of superalloys or stainless steels at 800°C. Fig.8 also shows surface morphologies of oxidized Be and $Be_{12}Ti$. Be formed porous and powdery scale. On the other hand, $Be_{12}Ti$ formed 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 a good oxidation resistance in air at high temperature. Furthermore, it is supposed that Ti would take a role of forming a protective BeO film on $Be_{12}Ti$ [12].

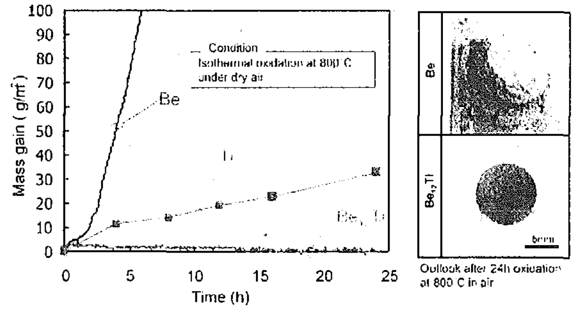


Fig.8 Oxidation property

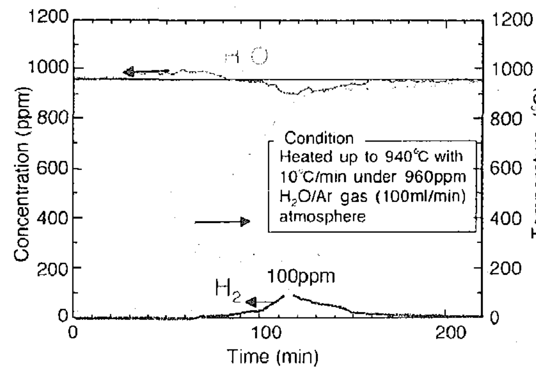


Fig.9 Interaction with steam

3.6 Chemical property (3) -Steam interaction-

The reactivity of $Be_{12}Ti$ with water vapor was investigated. In the experiments, the sample disks of $Be_{12}Ti$ were placed in a reactor. Argon gases with certain amounts of water vapor were introduced to the reactor, and the reactor temperature was raised up to around 1000 °C at constant rates. The concentrations of water vapor, hydrogen and oxygen in the outlet stream of the reactor were traced with a mass-spectrometer. Fig.9 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 chaotic 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 with beryllium. Thus, it can be said that titanium beryllides have less reactivity with water vapor [13].

3.7 Tritium inventory

A desorption property of deuterium was evaluated by the heating test after deuterium implantation. Some results are shown in Fig.10. Deuterium was implanted up to $1x10^{21}$ ions/m^2 at room temperature. The profile of 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°C-700°C) than that for $Be_{12}Ti$. The amount of 20% in implanted deuterium is retained in Be around 700°C. These results showed that the deuterium desorption property of $Be_{12}Ti$ was more superior than that of Be which could indicate that $Be_{12}Ti$ tritium inventory could be much smaller than that for Be. Suitable Neutron irradiation experiments should be performed to confirm these indications [14].

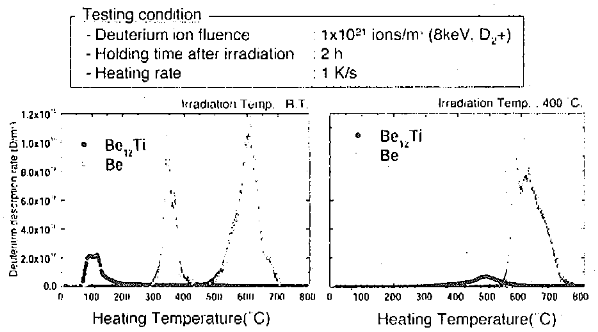


Fig.10 Deuterium desorption test

3.8 Charged particle irradiation

To evaluate microstructure response of $Be_{12}Ti$ in fusion equivalent environments, in-situ charged particle irradiation experiments were done as a function of dose and spontaneous He irradiation effects at elevated temperatures up to 700°C using Multi Beam High Voltage Electron Microscope (MBHVEM) in CARET, Hokkaido University. Several precipitates in $Be_{12}Ti$ were found to be BeO and titanium oxide by EDP and EDS. After He/electron irradiation at 500°C, tiny bubbles were observed along the grain boundary in Fig.11. However it was hard to observe black dots or dislocation loops in $Be_{12}Ti$ at same irradiation condition. Spontaneous He irradiation is enhanced to the nucleation of bubbles. Increasing the irradiation temperature, $Be_{12}Ti$ demonstrated same trend of the microstructure response following He irradiation at 500°C. Mechanical property at elevated temperatures is still undergoing [15].

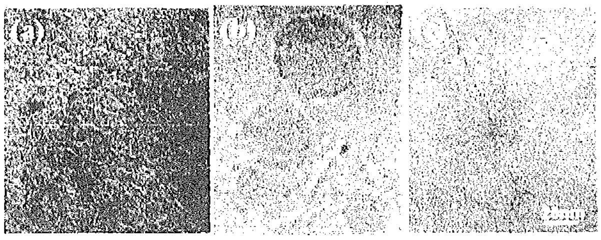


Fig.11 TEM after dual irradiation in (a) pure Be, (b) $Be_{12}Ti$ at R.T. and (c) $Be_{12}Ti$ irradiated at 500°C

3.9 IEA Irradiation program

It is necessary to have high neutron dose irradiation tests to evaluate the lifetime. Future plan for the irradiation tests is considered as shown in Fig.12. In the frame of an 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, T-retention) at DEMO relevant conditions in terms of dpa/He ratio and temperatures. The chosen irradiation temperature ranges between 400 and 800 °C. A 30% of DEMO end of life helium production (6000 appm in 4 years) should be achieved with an intermediate unloading of part of the samples at 3000 appm. At the maximum dose (40 dpa Be) a maximum T 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 an independent verification with another experimental apparatus [16].

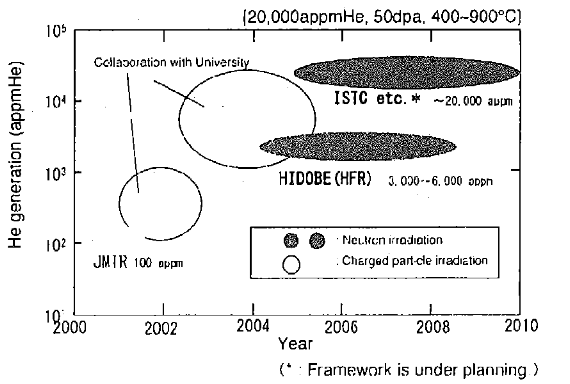


Fig.12 HIDOBE Test

4. CONCLUSION

From these results, there is a bright prospect of the application of beryllide as neutron multiplier for DEMO blanket due to the good chemical, thermal, mechanical, and the irradiation property.

The conclusion of the status on the R&D is as follows:

(1) Beryllium
- Beryllium pebbles for ITER test module condition have developed. The subjects to be studied are lifetime evaluation and recycling of used beryllium.

(2) Beryllide
- $Be_{12}Ti$, which was considered to have the easiest productivity, is being concentrated to be studied and developed. TBR estimation of the blanket using beryllide was carried out and TBR of the blanket with $Be_{12}Ti$ pebbles resulted only 10% smaller than that with Be pebbles and it is considered that this value is within design window.
- The relationship between the Ti content in Be-Ti and the mechanical property was studied and it was clarified that α-Be phase is effective to add some ductility. The pebbles with this phase were obtained by small scale REM trial.
- Characterization concerning the compatibility with structural material and ceramic breeder, oxidation, steam reaction, tritium inventory and the structure evolution by charged particles has been carried out and the advantage comparing with beryllium metal was made clear.
- In future plan, $Be_{12}Ti$ small disks (8 mm diameter, 2 mm thickness) will be irradiated up to 6000 appmHe starting in 2004 in the HFR reactor to obtain irradiation data (swelling, creep, T-retention) at DEMO relevant conditions in terms of dpa/He ratio and temperatures.

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