

# Elemental development of beryllide electrode for pebble production by rotating electrode method

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## Abstract

Beryllide ( $\text{Be}_{12}\text{Ti}$ ) is being considered one of the promising candidates for advanced neutron multipliers. In order to realize the pebble production, a vacuum casting process has to be applied. In the present study, an elemental technology to fabricate beryllide rods by a vacuum casting was developed. There were two major technical subjects to realize this process. The first was to identify a furnace material that shows minimum chemical reaction with beryllide. Another was to develop a procedure to cast durable ingot without any shrinkages and cracks. Reactivity between  $\text{Be}_{12}\text{Ti}$  melt and refractory materials such as  $\text{CaO}$ ,  $\text{MgO}$  and  $\text{BeO}$  was tested. It was clarified that the  $\text{BeO}$  crucible had less reactivity with melt and no contamination. Casting tests with a  $\text{MgO}$  cylindrical mold in a vacuum chamber revealed that the mold dimension was critical to minimize shrinkages and cracks. From this study, elemental technologies to be developed were made clear.

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## 1. Introduction

Beryllium (Be) metal has been extensively studied as a neutron multiplier of a fusion blanket. The pebble fabrication technology of Be metal has been established and characterizations have been conducted [1]. However, Be metal has a disadvantage for high temperature application such as in the Japanese DEMO blanket, which will be operated at temperature up to 900 °C and high neutron dose ( $\sim 20\,000$  appm He,  $\sim 50$  dpa). Thus, beryllide ( $\text{Be}_{12}\text{Ti}$ ), which has higher melting points and higher chemical stability at high temperatures, was selected as one of candidate materials from the viewpoints of low activation, high Be content, low oxidation resistance, etc. [2–4]. The compatibility study of the  $\text{Be}_{12}\text{Ti}$

with a structural material (SS316LN) showed more favorable characteristics was revealed as better than that of Be [5]. The swelling of a  $\text{Be}_{12}\text{Ti}$  disk that was irradiated at 500 °C up to a fast neutron fluence of  $\sim 4 \times 10^{24}$   $\text{nm}^{-2}$  ( $E > 1$  MeV) in the reactor (JMTR) was less than 1/20 of the value for Be [6].

On the other hand, studies to reduce the brittleness of beryllide rod for pebble fabrication have been carried out [7]. As results of several tests, it became clear that duplex metallographic structure with  $\alpha\text{Be}$  was effective to improve the ductility of stoichiometric  $\text{Be}_{12}\text{Ti}$ . Then, the electrode with this structure, which was fabricated by an arc melting method, has withstood the thermal stress from the arc heating in a small-scale rotational electrode method (REM) test, and some pebbles were obtained. In this test, Be-5 at.%Ti and Be-7 at.%Ti pebbles were successfully obtained.

In order to fabricate the  $\text{Be}_{12}\text{Ti}$  pebbles by REM, manufacturing technology of fabrication of the real-size electrode are the necessary, development of vacuum

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melting and vacuum casting is necessary as the manufacturing processes. This paper reports on the result of the trial fabrications.

## 2. Experimental

### 2.1. Melting test

As raw materials for this study, a beryllium ingot with a purity of 99.5 wt%, and a Ti sponge with a purity of 99.99% were used. The raw materials were weighed and blended into desired chemical compositions, then charged into the arc melting furnace with water cooled copper mold and the master alloys were pre-fabricated to make the melting operation in the induction furnace easier. The master alloys were subsequently melted in the vacuum induction furnace as shown in Fig. 1. The target value of chemical composition was 78.1 wt%Be and 21.9 wt%Ti, relevant to Be-5 at.%Ti. This chemical composition is one of the candidates that give the material some ductility because of the existence of  $\alpha$ Be as shown in Fig. 2 [8]. Melting process was carried in the chamber evacuated to  $1 \times 10^{-3}$  Pa, with purging Argon gas of the purity of 99.99 wt%. The induction furnace consists of the induction heater, the inner refractory crucible, MgO stamping powder layer and MgO crucible as shown in Fig. 3. The inside dimension of the inner refractory crucible was  $\varnothing 87 \times 170$  mm. As the inner refractory crucible, MgO, CaO, and BeO were tested.

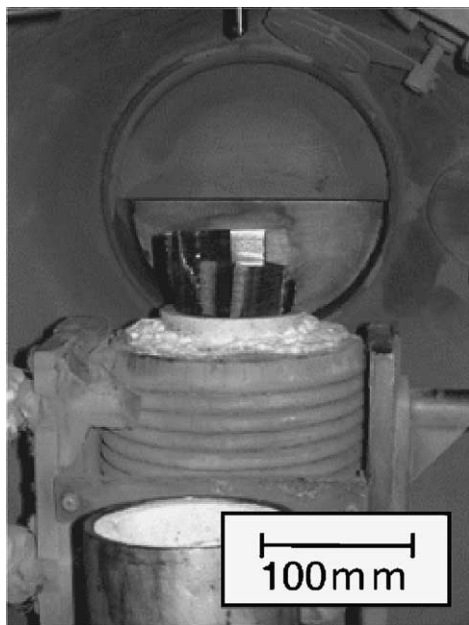


Fig. 1. Overview of vacuum induction furnace and casting mold.

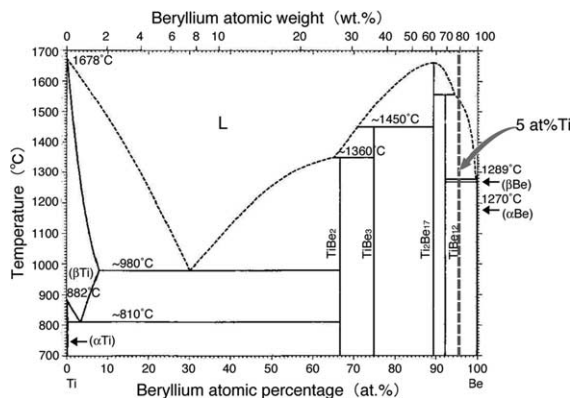


Fig. 2. Assessed Be-Ti binary phase diagrams.

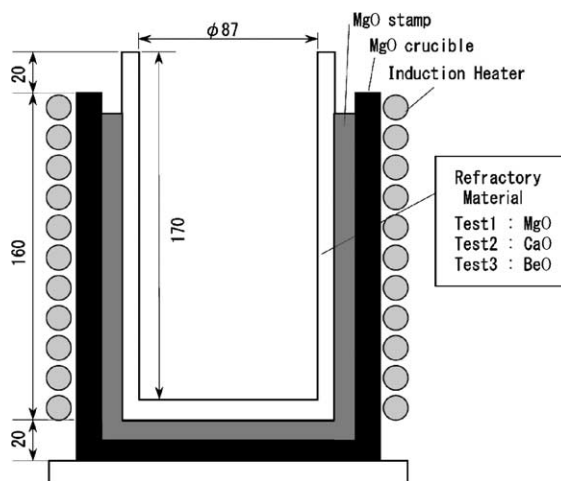


Fig. 3. Design of melting crucible.

These materials were selected because their free energies of oxidation are close to that of Beryllium. On the Ellingham diagram as shown in Fig. 4, MgO is unstable with Beryllium at all temperatures while CaO is stable under  $\approx 1700$  °C [9]. The melting temperature was measured with Pt-Rh thermocouple.

### 2.2. Casting test

Casting tests were carried out with MgO cylindrical mold in a vacuum chamber. The mold dimension (the ratio of diameter to height) was surveyed to understand its impact on the solidification behavior. The melt was prepared with the same procedure described in Section 2.1. The mold design is shown in Fig. 5. The inside diameters of the molds are 50 mm in Test A and 85 mm in Test B, respectively. The height of the mold is 170 mm in both tests. The molds were set on the water cooled copper mold, which enhanced the cooling rate from the

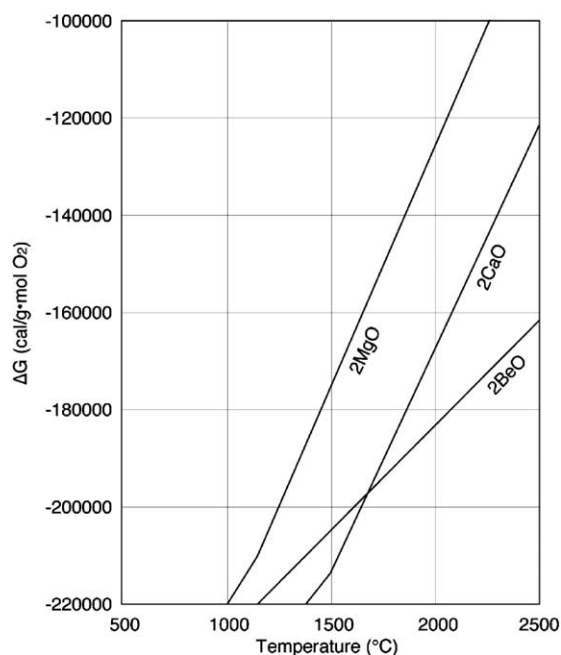


Fig. 4. Ellingham diagram.

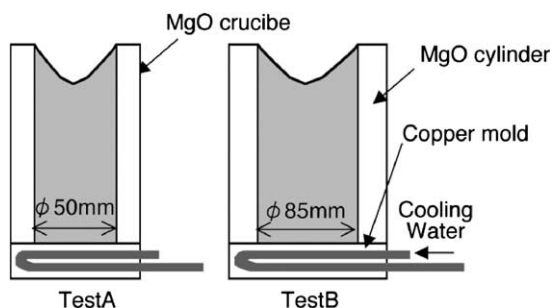


Fig. 5. Design of casting mold.

bottom. After casting, mold was disassembled and the cast ingot was removed. The ingots were cut by electrical wiring cutting method and the cross section was observed. The chemical composition was analyzed with the sample taken from the ingots by ICP method.

### 3. Results and discussion

#### 3.1. Melting test

Reactivity between  $\text{Be}_{12}\text{Ti}$  melt and refractory materials such as  $\text{CaO}$ ,  $\text{MgO}$  and  $\text{BeO}$  was tested. One kilogram of  $\text{Be}_{12}\text{Ti}$  was melted in the crucible with each refractory material by a 20 kW vacuum induction furnace. The melt was heated up to 1700 °C and cooled

Table 1  
Results of melting test

No.	Material	Purity (%)	Results
Test 1	MgO	>98.0	Heavy reaction
Test 2	CaO	98.0–99.0	Heavy reaction, heavy cracks
Test 3	BeO	>99.5	No reaction, small crack

down. After cooling to room temperature, the reactivity was evaluated by observation of the melt/crucible interface and by chemical analysis of  $\text{Be}_{12}\text{Ti}$ . The results are shown in Table 1.  $\text{MgO}$ , which is usually used for beryllium melting at 1300 °C, shows severe reaction with the melt and many heavy holes (indication of  $\text{MgO}$  losses) were observed after melting. Although  $\text{CaO}$  had less reactivity, heavy cracks were observed on the surface of crucible after melting. It was found that the  $\text{BeO}$  crucible had less reactivity with melt.

#### 3.2. Casting test

The melt was prepared with  $\text{BeO}$  crucible in the induction furnace. The casting temperature was 1650–1750 °C in Test A, respectively. The results were shown in Table 2. In Test A, shrinkage in the center of the ingot as well as surface crack were observed as shown in Fig. 6. The chemical composition was analyzed in four portions, and the results were shown in Table 3. Segregation was detected at near shrinkage of center portion. Except this portion, no significant segregation was detected, and it can be said that  $\text{Ti}$  is within 1.2 wt%, which means 0.4 at.% in 10–110 mm height from bottom. This variation of  $\text{Ti}$  is considered acceptable. It is noticed that there is some contamination of  $\text{Mg}$  near the mold area. In center area, no contamination was observed. The microstructure is observed by Scanning Electron Microscopy (SEM) at the same portion where the chemical composition was analyzed. The results are shown in Fig. 7. The microstructure consists of  $\text{Be}_{12}\text{Ti}$  as white and  $\alpha\text{Be}$  as white. In the center at 110 mm from the bottom, a big grain was observed and it is considered to be formed due to a slow cooling rate by shrinkage.

Table 2  
Results of the casting test

No.	Cast temperature (°C)	Mold diameter (mm)	Results
Test A	1650	ø50	Heavy shrinkage
Test B	1750	ø85	Sound portion: 40 mm height, small cracks

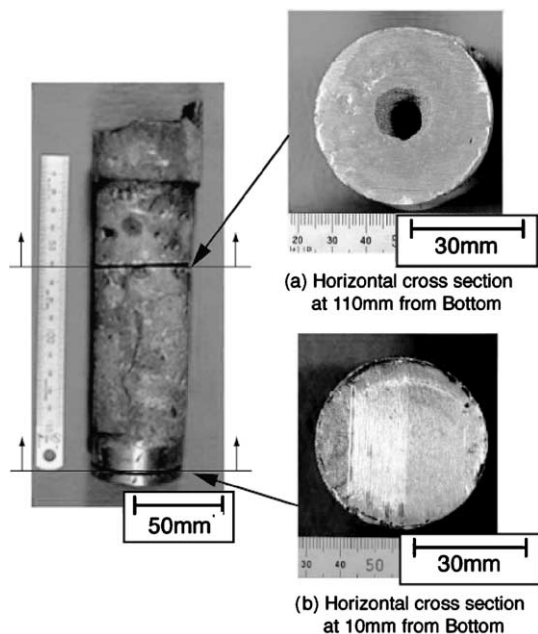


Fig. 6. Observation of the cast ingot Test A.

With the exception of this area, it can be said that the microstructure is uniform.

Next, Test B with a bigger mold, which had 80 mm inside diameter, was carried out to evaluate its effect on shrinkage and crack reductions. Fig. 8 shows outside view of the ingot and Fig. 9 shows a cross section view. Vertical cracks were observed on the side surface and on the bottom. Shrinkage was observed over 40 mm from the bottom. Needle type dendritic crystal was observed in the shrinkage area. It is considered that shrinkage occurred with stronger cooling from the sidewall against the cooling from the bottom. From this mechanism, (volume of ingot/surface of wall) ratio played a critical role on shrinkage formation. By comparing the values in Test A and Test B, (that is  $V_A/S_A:V_B/S_B$ , equals 1:1.7), it can be said that Test B has better cooling balance between side and bottom to reduce shrinkage.

As to the casting procedure, further studies to reduce shrinkage, crack and contamination are desired.

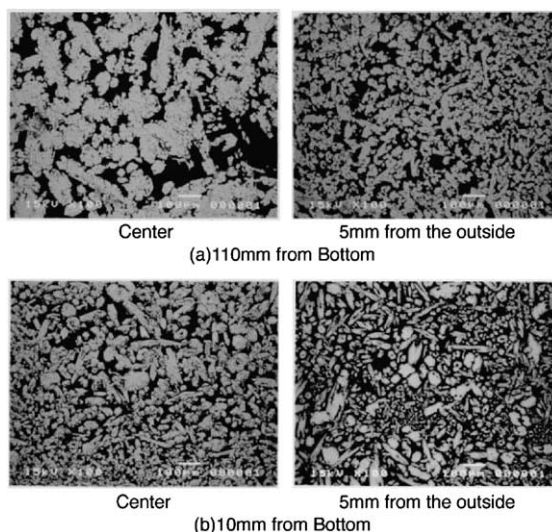


Fig. 7. Microstructure of the cast ingot Test A.

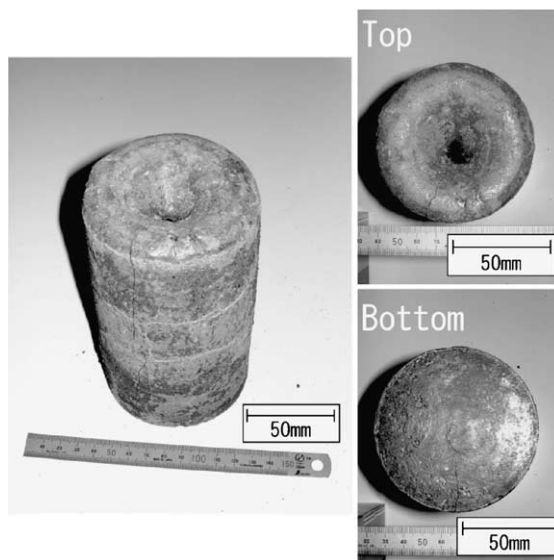


Fig. 8. Observation of the outside of the ingot Test B.

Table 3  
Analyzed chemical composition of the ingots Test A (wt%)

Lot number		Be	Ti	V	Mg	Al	Si	Fe	Co	Cu
Target		78.1 (95.0) <sup>a</sup>	21.9 (5.0) <sup>a</sup>	–	–	–	–	–	–	–
110 mm from bottom	Center	75.8	23.6	0.23	0.009	0.072	0.036	0.049	0.029	0.022
	5 mm from the outside	79.8	19.6	0.19	0.049	0.095	0.034	0.074	0.022	0.044
10 mm from bottom	Center	80.7	18.7	0.18	0.056	0.099	0.032	0.071	0.022	0.031
	5 mm from the outside	80.9	18.4	0.17	0.13	0.10	0.038	0.075	0.021	0.036

<sup>a</sup> Atomic percentage.

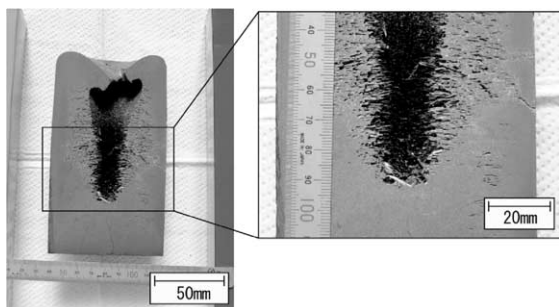


Fig. 9. Cross section of the cast ingot Test B.

#### 4. Conclusion

The results of this study can be summarized as follows:

- It was made clear that the BeO crucible had less reactivity with melt. And, the melt of Be-5 at.%Ti was successfully fabricated.
- It was made clear that the ingot of Be-5 at.%Ti, which has  $\alpha$ Be phase and enough dimension for the

electrode, was able to be fabricated with Vacuum Induction melting and Vacuum casting process.

- The technical subjects such as undesirable shrinkage, the crack formation and contamination from the mold require further study in order to realize an economical production of the electrode.

#### References

- [1] S. Tanaka et al., in: International Symposium, vol. 51&52, 2000, p. 299.
- [2] H. Kawamura et al., Fusion Eng. Des. 61&62 (2002) 391.
- [3] J.M. Marder et al., in: Second International SAMPE Metals Conference, 2–4 August 1988, p. 357.
- [4] S.M. Bruemmer et al., in: Proceedings of the Material Research Society Symposium, vol. 288, 1993, p. 799.
- [5] H. Kawamura et al., J. Nucl. Mater. 307–311 (2002) 638.
- [6] M. Uchida et al., J. Nucl. Mater. 307–311 (2002) 653.
- [7] M. Uchida et al., Fusion Eng. Des. 69 (2003) 491.
- [8] H. Okamoto et al., ANS International, 1960, ISBN:0-87170-303-3.
- [9] J.F. Elliott, in: Thermochemistry for Steelmaking, vol. 1, 1960.