

# Synthesis and reactivity of single-phase Be<sub>17</sub>Ti<sub>2</sub> intermetallic compounds



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## HIGHLIGHTS

- Preliminary synthesis of single-phase Be<sub>17</sub>Ti<sub>2</sub> was succeeded.
- Reactivity difference between beryllium and beryllides may be caused by a lattice strain.
- Oxidation of Be<sub>17</sub>Ti<sub>2</sub> at high temperatures results in the formation of TiO<sub>2</sub>.
- Simulation results reveal that a stable site for hydrogen at the center of tetrahedron exists.

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## ABSTRACT

To investigate feasibility for application of Be<sub>17</sub>Ti<sub>2</sub> as a neutron multiplier as well as a refractory material, single-phase Be<sub>17</sub>Ti<sub>2</sub> intermetallic compounds were synthesized using an annealing heat treatment of the starting powder and a plasma sintering method. Scanning electron microscopic observations and X-ray diffraction measurements reveal that the single-phase Be<sub>17</sub>Ti<sub>2</sub> compounds were successfully synthesized. We examined the reactivity of Be<sub>17</sub>Ti<sub>2</sub> with 1% H<sub>2</sub>O and discovered that a larger stoichiometric amount of Ti resulted in the formation of TiO<sub>2</sub> on the surface at high temperatures. This oxidation may also contribute to an increase in both weight gain and generation of H<sub>2</sub>. This suggests that the formation of the Ti-depleted Be<sub>17</sub>Ti<sub>2-x</sub> layer as a result of oxidation facilitates an increased reactivity with H<sub>2</sub>O. To evaluate the safety aspects of Be<sub>17</sub>Ti<sub>2</sub>, we also investigated the hydrogen positions and solution energies based on the first principle. The calculations reveal that there are 10 theoretical sites, where 9 of these sites have hydrogen solution energies with a positive value (endothermic) and 1 site located at the center of a tetrahedron comprising two Be and two Ti atoms gives a negative value (exothermic).

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## 1. Introduction

Much attention has been given to the application of Be<sub>13</sub>M, Be<sub>12</sub>M, and Be<sub>17</sub>M<sub>2</sub> compounds as refractory materials because of their high temperature strength, good thermal conductivity, excellent oxidation resistance, and so on. Furthermore, single-phase intermetallic compounds, Be<sub>12</sub>Ti, Be<sub>17</sub>Ti<sub>2</sub>, Be<sub>12</sub>V, Be<sub>13</sub>Zr are considered to be the promising neutron multiplying material in a demonstration power plant reactor because of its nuclear properties and its reactivity at high temperature. Several attempted synthetic methods have been reported thus far on the fabrication of beryllides. We have suggested a new synthetic process that employs a combination of a plasma sintering method and a rotating

electrode method (REM) [1–4]. We have also reported on the prospects of mass-producing beryllide pebbles with a 1 mm diameter [2]. However, it was obvious that the fabrication of beryllide pebbles (Be–7.7 at.% Ti) with the REM resulted in formation of the Be<sub>17</sub>Ti<sub>2</sub> and Be<sub>12</sub>Ti phases on the surface whereas that of Be, Be<sub>12</sub>Ti and Be<sub>17</sub>Ti<sub>2</sub> [1] in cross section. This result was best explained by the occurrence of a peritectic reaction from re-melting during the REM process [1]. In the case of a multiplier for neutrons related to the tritium breeding rate, the beryllide pebble fabricated by the REM should not pose any significant problems even though the pebble itself has a different phase composition than that of the rod used for the REM. However, since the phase composition on the surface of the pebble predominately influences its reactivity at high temperatures, further evaluation of a single-phase Be<sub>17</sub>Ti<sub>2</sub> compound is inevitable. Furthermore, it is important to investigate the compound's multiplying abilities and its role as a refractory material at high temperatures.

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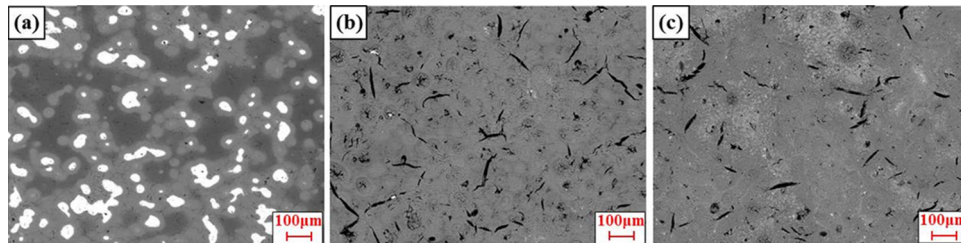


Fig. 1. SEM images of Be–10.5 at.% Ti beryllide that was plasma-sintered at 1273 K for 20 min (a) as received, (b) annealed at 1473 K for 1 h, and (c) annealed at 1473 K for 5 h.

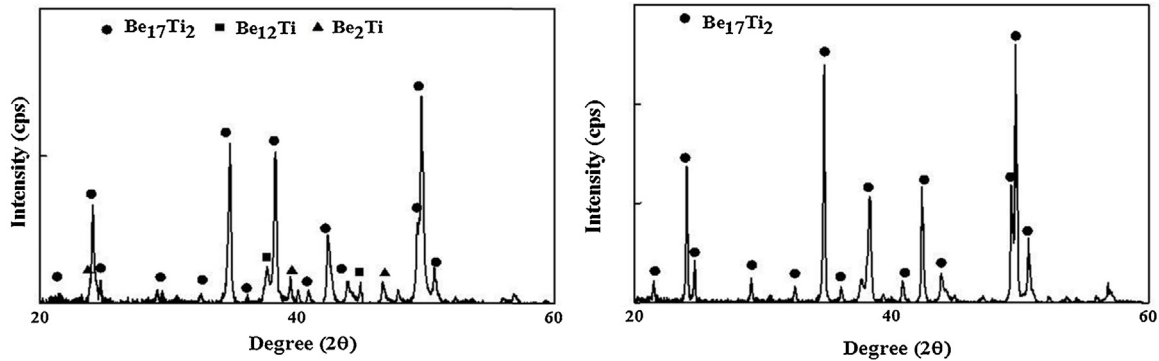


Fig. 2. XRD profiles of as-received Be–10.5 at.% Ti beryllide (left) and as-annealed at 1473 K for 1 h (right).

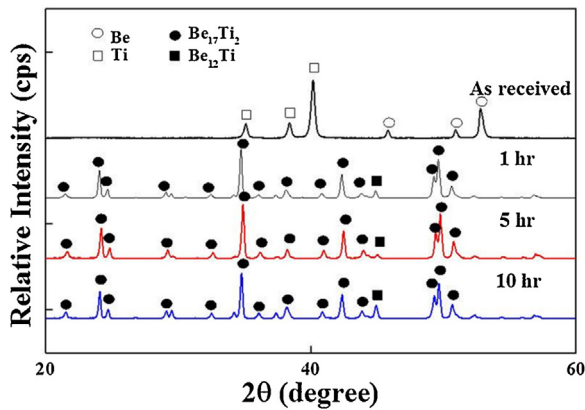


Fig. 3. XRD profiles of Be–10.5 at.% Ti powder in its as-received and as-annealed forms at 1473 K for 1, 5, and 10 h.

Beryllium and lithium have also been reported as neutron-producing targets in an accelerator-based boron neutron capture therapy (BNCT) [5]. Because of its relatively high melting point and excellent thermal conductivity, beryllium metal has attracted significant interest as an effective neutron-producing target. However, the water cooling system that is used to control the temperature contributes to a problematic generation of  $H_2$  gas. This is attributed to an unexpected reaction between beryllium and leaked  $H_2O$  from the coolant pipe lines. Therefore, beryllium-based intermetallic compounds such as single-phase  $Be_{12}Ti$  and  $Be_{17}Ti_2$  would be a better alternative for a potential beryllium target because of its higher melting point and higher stability in the presence of  $H_2O$ . Although some studies on  $Be_{12}Ti$  have been broadly carried out, however, studies on the fabrication and stability of a single-phase  $Be_{17}Ti_2$  are currently insufficient.

In this study, the single-phase  $Be_{17}Ti_2$  was fabricated through a combinational process involving an annealing and plasma sintering method to generate the powder. To evaluate its reactivity at high temperatures, the weight gain and the amount of generated  $H_2$  were investigated at 1073, 1273, and 1473 K under 1%  $H_2O/Ar$ .

Additional simulation analysis results on the hydrogen positions and solution energies for  $Be_{17}Ti_2$  will also be reported with regard to its safety aspects.

## 2. Material and methods

The starting powders for Be ( $<45 \mu m$ ) and Ti ( $<45 \mu m$ ) used in this study had high purity percentages of 99.5% and 99.9%, respectively. In the first step, the plasma sintering method [6] was used to prepare Be–10.5% Ti beryllide, which was further annealed at 1473 K for 1 h and 5 h to investigate the possibility of homogenization into the single-phase  $Be_{17}Ti_2$ . The qualitative analysis was conducted with X-ray diffraction measurements (XRD, UltimaIV, Rigaku, Japan) and electron probe micro-analysis (EPMA, JXA-8530F, JEOL, Japan). In the next step, the preliminary synthesis of single-phase  $Be_{17}Ti_2$  was attempted through the calcination of the starting powder and the plasma sintering method, which was derived from the existing synthetic technologies for  $Be_{12}Ti$  [7]. The powder was weighed with Be–10.5 at.% Ti in a stoichiometric ratio, mixed by mortar for 1 h, and then calcinated at 1473 K for 10 h under Ar gas. To confirm the presence of the phase, the treated powder was examined using XRD and scanning electron microscopy (SEM). The powder was ground until the particle sizes were less than  $45 \mu m$  to match the particle size of the starting powder. After this pre-treatment method, we attempted to fabricate a disk type of the  $Be_{17}Ti_2$  sample using the plasma-sintering method. The powder was loaded into a graphite punch and die. A pulse current of 500 A was applied for 20.2 ms with the application of pressure to activate the powder surface. A direct current was then applied to facilitate the heating of the sample to 1273 K for 20 min using a heating ratio of 100 K/min.

In fact, to verify that  $Be_{17}Ti_2$  was synthesized in this process, the compound was examined by XRD. Furthermore, an electron probe micro-analyzer was used for surface observation and qualitative analysis. The weight gain and the amount of generated  $H_2$  for single-phase  $Be_{17}Ti_2$  were evaluated using thermogravimetry (TG, TG-8110, Rigaku, Japan) and gas chromatography (GC4900, Agilent, USA) at 1073, 1273, and 1473 K for 24 h in 1%  $H_2O/Ar$ .

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