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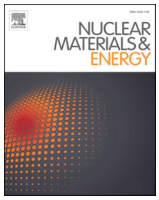

Effect of temperature on mechanical properties of beryllium intermetallic compounds fabricated by plasma sintering

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ABSTRACT

This purpose of study is to establish the material database of neutron multiplier for the JA DEMO design. In the previous study, we reported the effects of sintering conditions as temperature and pressure on hardness and sintered density mainly in the sintering temperature from 1050 °C to 1200 °C. In this study, the microstructure observation and compressive tests were carried out. As results of microstructure observations, the pores were almost disappeared, and the grain size increased with increase in sintering temperature. Compression tests were carried out from room temperature (R.T.) to 1000 °C with the samples sintered at 1200 °C. The compressive strength at R.T. was approximately 1.69 GPa. With the increase in testing temperature, there was a tendency for the strength to decrease from 800 °C to 1000 °C. In addition, the load-compressibility indicated that the yield point appears at 850 °C.

1. Introduction

Beryllium (Be) has been considered as a neutron multiplier material for fusion reactors. however, in terms of hydrogen generation [1] and volumetric expansion as swelling due to the reaction with water vapor under high temperatures and neutron irradiation [2], beryllium intermetallic compounds (beryllides) were suggested which have excellent high-temperature stability. In addition, two blanket design concepts have recently been proposed for a Japanese Demonstration fusion reactor (JA DEMO). Recently, the design of nuclear fusion demonstration reactors is being updated in various parts of the world, including Europe and Japan.

In the case of JA DEMO, according to Someya et al [3], there are two blanket design concepts of JA DEMO reactor suggested. One is a honeycomb rib structure packed with the mixed pebbles of $Be_{12}Ti$ pebbles with 2 mm in diameter and Li_2TiO_3 pebbles with 0.2 mm in diameters. This binary pebble packing design was able to achieve more than 1.05 of the tritium breeding ratio (TBR) with 80 % of the packing fraction.

The other proposal is cylindrical structure. Cylindrical structure is supposed to be loading blocks of beryllide and Li_2TiO_3 pebbles. In which, this blanket design concept has many merits for simplification in terms of engineering feasibility, especially in the case of the cylindrical structure with a block and also, has greater volume of Be atomic density than pebble bed, thus, the local TBR anticipates as approximately 1.21. Furthermore, the results of temperature distribution of the cylindrical structure were revealed. In the case of Li_2TiO_3, the temperature reached about 765 °C. In the case of $Be_{12}Ti$ block, the temperature seemed to reach about 600 °C since they were in contact.

Thus, the EU and Japan are studying on neutron multiplier materials according to their own new candidate materials [4], blankets designs, and there are particularly little data on the evaluation of thermal properties [5–7], mechanical properties [8–11], manufacturing [12–14], and so on. Especially, there are not enough data for $Be_{12}Ti$ beryllide block produced by a plasma sintering method. Therefore, previous studies have investigated the change in density with sintering temperature [15], and the tensile strength [11]. In this study, we aimed to build a database of the mechanical properties of beryllides produced by the plasma sintering by investigating the changes in microstructure with temperature of the sintered material and the changes in compressive strength with temperature after sintering under optimal conditions.

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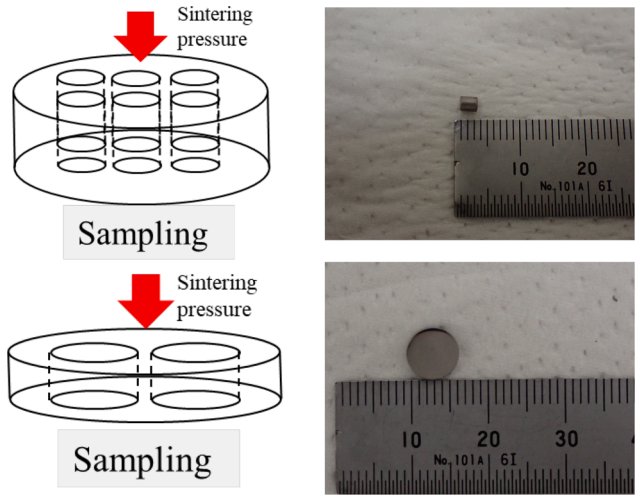

Fig. 1. Sampling direction and samples.

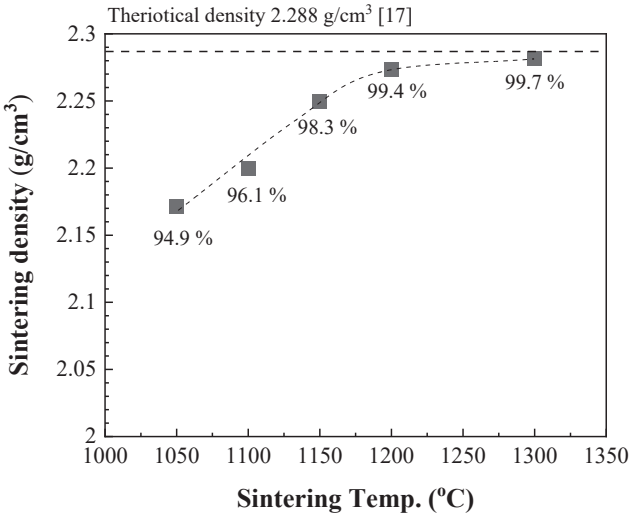

Fig. 2. The results of measuring sintering density with sintering temperature.

2. Experiments

Preparation of beryllide powders was started with mixing beryllium powders (99.4 wt%, <45 μm, Materion, USA) and 7.7 at% titanium powders (>99.9 wt%, <45 μm, Kojundo Chemical Lab.) by using mortar grinder (RM200, Retsch, Germany), and which is the stoichiometric value of $Be_{12}Ti$.

In order to synthesize the single phase of $Be_{12}Ti$ powder as raw material for plasma sintering, the raw powders of Be and Ti mixed in Be-7.7at.%Ti was annealed at 1200 °C for 24 h under argon (Ar) gas flow of 200 mL/min.

For preparing beryllide blocks, the homogenized powders were loaded into graphite punch and die units. Next, plasma sintering (KE-PAS II, Kaken, Japan) was performed at 1200 °C for 20 min at 54.1 MPa in less than 4.0 Pa as low-pressure atmosphere.

After plasma-sintering, a wire EDM (Electrical Discharge Machine) machine was used to prepare the test specimens. The specimens were fabricated in the sintering direction as shown in Fig. 1.

After sintering, specimens were prepared. To observe the microstructure, the surface of the beryllide was physically and electro-polished, analyzed by SEM (Scanning Electron Microscope) and EBSD (Electron Backscatter Diffraction) in IAM, KIT (The Institute for Applied Materials, Karlsruhe Institute of Technology). The calculations and

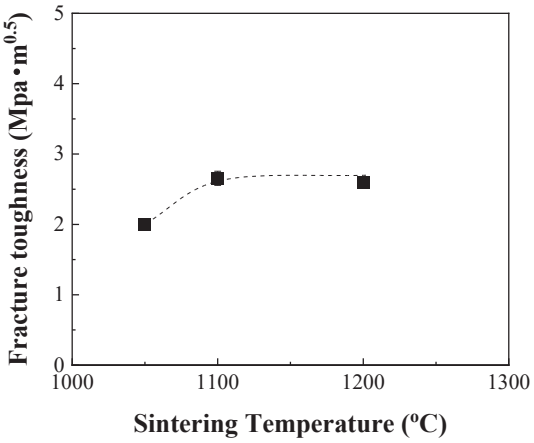

Fig. 3. The results obtained by calculation with if method [15].

measurements were carried out with diameter method by OIM (Orientation Imaging Microscopy) analysis for grain boundary analysis and ASTM E112 [16].

In order to investigate the mechanical properties after sintering, a crack of 5 kgf was generated on the surface in a Vickers hardness tester. The fracture toughness was calculated from the crack length and load using the IF (Indentation fracture) method. As the sintering temperature increased, the destructive toughness test according to the IF method was additionally conducted up to the material conducted at 1200 °C. The calculation used the Tanaka formula in equation 1, which uses the median crack. A wire EDM machine was used to fabricate the specimens. The specimens were machined in the sintering direction as shown in Fig. 1.

To investigate the strength of beryllide blocks sintered at 1200℃ under compressive load, compression tests were performed in KIT IAM, Germany with using cylindrical (L/D ≈ 1.2) specimens of φ2.2 mm × T 2.6 mm diameter. The traverse speed was set as 0.05 mm/min.

3. Results and discussion

As shown in Fig. 2, we previously reported the change in sintered density as a function of sintering temperature [11]. The result was about 94.9 % of the theoretical density [17] (2.288 g/cm^3) when sintered at 1050 ℃. The sintered density increased as the sintering temperature increased, and about 99.4 % of the sintered density was expressed at 1200 ℃. There was little difference between the results at 1300 °C, and the report showed that the workability of the material deteriorated at 1300 °C.

The fracture toughness was calculated by the result of Vickers hardness using the conversion formula of Tanaka. Tanaka's formula as the IF method [15] was selected from the viewpoint of conservative evaluation because Tanaka's formula was showed the lowest value in previous study [15].

The results of calculation indicates in Fig. 3. The values of about 2.8 MPa·m$^{0.5}$ and 2.6 MPa·m$^{0.5}$ were obtained at 1100 °C and 1200 °C, respectively. These values appear to exhibit higher fracture toughness compared to the previously reported value of 2.0 MPa·m$^{0.5}$ [15]. These results indicate that the values are almost the same after 1100 °C. This result shows a similar trend to the results of the Vickers hardness test conducted previously, suggesting that the density increases slightly after the sintering temperature of 1100 °C, but the hardness and fracture toughness are not affected to such a large extent.

To investigate microstructure behaviour, microstructural observations were made by SEM and EBSD on sintered materials at sintering temperatures of 1050 °C, 1100 °C, and 1200 °C. The results are shown in the following figures: a) is an SEM observation of the surface of sintered material at 1050℃, b) is a surface image of sintered material at 1100 °C.

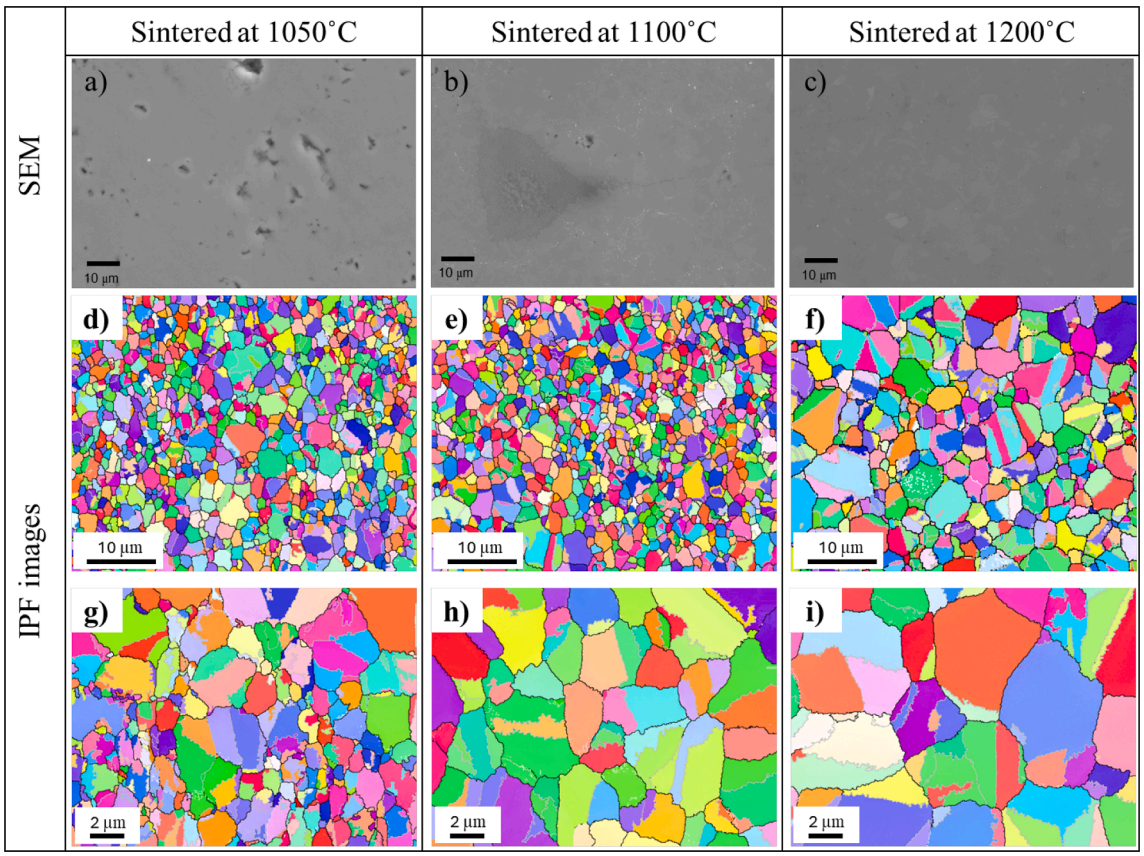

Fig. 4. Results of observation for the microstructures of be_{12} Ti which sintered at various temperature, (a,d,g) sintered at 1050 ℃; (b,e,h) 1100 ℃; (c,f,i) 1200 ℃.

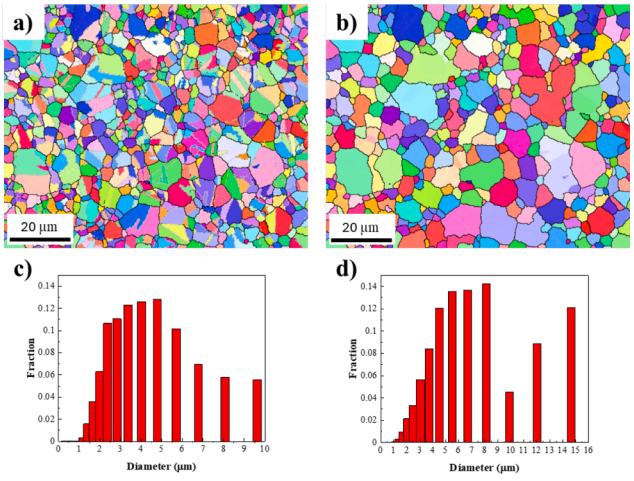

Fig. 5. The microstructures and results of grain size analysis by oim for be_{12}Ti which sintered at 1200 ℃; (a,c) including twins, (b,d) disregarding twins.

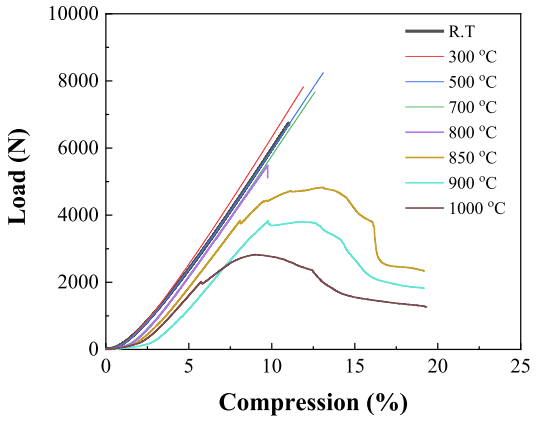

Fig. 6. Relationship between load-compression which tests at various temperatures.

In case c), shows the surface of the sintered material at 1200 °C. From the SEM observation, as the sintering temperature increased, fewer pores were found on the surface.

The Fig. 4 displays the EBSD analysis results. Fig. 4 d) and g) depict the analysis outcomes for materials sintered at 1050 °C, panels e) and h) at 1100 °C, and Fig. 4 f) and i) at 1200 °C. The IPF (Inverse Pole Figure) illustrates the analysis results obtained with the standard for major angle grain boundaries set at 15° as usual. The black and white lines on the maps indicate high angle grain boundaries, respectively, while silver lines correspond to twin boundaries.

The results of grain size analysis showed that the average values were between 1.06 and 2.44 μm with Be_{12}Ti sintered at 1050 °C, 1.10 and 4.15 μm with Be_{12}Ti sintered at 1100 °C, and 1.62 and 5.34 μm with Be_{12}Ti sintered at 1200 °C when including twins. In addition, the calculation of grain size also was conducted based on ASTM E112 [16] with the sample which sintered at 1200 ℃, the grain size No. was nearly 12.9. Thus, the average diameter as grain size was approximately 4.0 μm sintered at 1200 ℃. In addition, according to the OIM analysis as shown in Fig. 5, the interpreted average grain size was 4.38 ± 2.16 μm. However, in the case of degrading twins, a larger average size of 7.29 ± 3.80 μm was obtained.

Based on the results, sintering the material at 1200 °C is considered optimal for achieving high sintered density and fracture toughness. Therefore, compression tests were performed on the materials sintered

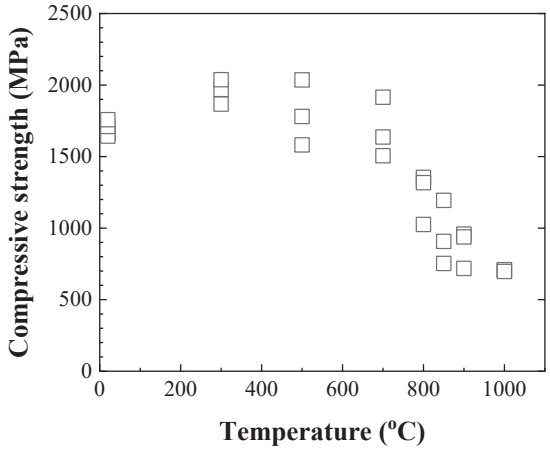

Fig. 7. The results of compressive tests in the temperature range from r.t to 1200 ℃.

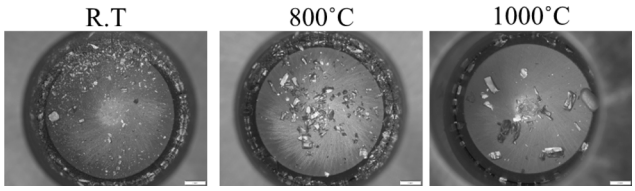

Fig. 8. The fragments after compression tests at R.T, 800 ℃, 1000 ℃.

at this temperature, and the results are depicted in the accompanying figure.

Fig. 6 shows the relationship between load and compression at various temperatures. This figure showed the relationship of compressive load from R.T. to 700 °C with no plastic deformation peculiar to brittle materials, but there was no deformation at 800 °C, but the strength decreased, and from 850 °C and above, a clear yield point appeared. The deformation region appeared to widen as the temperature increased.

Fig. 7 shows the relationship between test temperature and compressive strength. When the temperature was 25 °C, the compressive strength ranged from 1650 to 1760 MPa, with an average of about 1692 MPa; when tested at 300 ℃, the results ranged from 1866 to 2037 MPa, with an average of about 1957 MPa; and when tested at 500 °C, the results ranged from 1581 to 2036 MPa, with an average of 1799 MPa; 700 °C results ranged from 1505 to 1914 MPa; at 800 °C and 850 °C, compressive strength decreased, with values ranging from 1025 to 1354 MPa and 752 to 1193 MPa, respectively.

At 900 and 1000 °C, the compressive strength decreased further, with values ranging from 718 to 958 MPa and 697 to 709 MPa, respectively.

According to new analysis results [18], the maximum operating temperature for $Be_{12}Ti$ was reported to be approximately 800 °C. Additionally, it was reported that the F82H surrounding $Be_{12}Ti$ generates a thermal stress of approximately 478 MPa. Conversely, in the case of $Be_{12}Ti$, it is affected by the expansion of F82H. Regarding the pressure due to the expansion of F82H, when only the internal compression load is considered, it is about twice as much as 476 MPa at the operating temperature of 800 ℃, reaching approximately 1000 MPa. From this, it is considered that the fragment using the data so far is satisfactory.

After compression tests the fragments were shown in Fig. 8, the fragments became larger as the temperature increased. It was also found that the remaining fragments was about 2.2 mm high, as it appeared to have shrunk before breaking rather than breaking completely.

However, since the N number is considered to be extremely important when evaluating the mechanical properties of brittle materials, we plan to continue collecting mechanical property data.

4. Summary

As a result of the $Be_{12}Ti$ microstructure development behavior depending on plasma sintering temperature and compression tests, the following findings were obtained.

1) As increase of sintering temperature, the grain size increased obviously.
2) The fracture toughness results were higher when sintered at 1100 and 1200 ℃ than the previously reported results at 1050 ℃, but there was almost no difference between 1100 ℃ and 1200 ℃.
3) Compression test results showed a tendency for compressive strength to decrease with increasing temperature from room temperature above 700 ℃. A tendency for plastic deformation to occur with a yield point appearing at temperatures above 800 ℃ was also observed.

CRediT authorship contribution statement

Taehyun Hwang: Investigation, Writing – original draft, Writing – review & editing, Resources. **Jae-Hwan Kim:** Supervision, Methodology, Project administration, Conceptualization. **Yutaka Sugimoto:** Resources. **Ramil Gaisin:** Investigation, Validation. **Rolf Rolli:** Investigation, Resources, Validation. **Pavel Vladimirov:** Project administration. **Yoshiaki Akatsu:** Resources. **Shota Yokohama:** Resources. **Suguru Nakano:** Resources. **Masaru Nakamichi:** Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The authors do not have permission to share data.

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