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Development of beryllide pebbles with low-hydrogen generation as advanced neutron multipliers

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HIGHLIGHTS

- Prototypic pebbles of beryllides, $Be_{12}Ti$ and $Be_{12}V$, were successfully fabricated.
- It was possible to fabricate prototypic $Be_{12}V$ pebbles without homogenization.
- Beryllides pebbles was much more resistant to water vapor than pure Be pebbles.
- The surface BeO layer was shown to act as a protective barrier against H_2 generation.

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ABSTRACT

Hydrogen generation via an oxidation reaction of Be with water vapor at high temperatures should reduce the safety associated with fusion reactors. Advanced neutron multipliers with higher stability at high temperatures are desired as replacements for the existing Be neutron multipliers for the pebble bed blanket design using a water coolant. Beryllium intermetallic compounds (beryllides) are the most promising material for advanced neutron multipliers. In this study, prototypic beryllide pebbles, $Be_{12}Ti$ and $Be_{12}V$, were successfully fabricated using the rotating electrode granulation method with a plasma-sintered beryllide electrode. The beryllides pebbles were much more resistant to water vapor than pure Be pebbles. The surface BeO layer on the beryllide pebbles was shown to act as a protective barrier against hydrogen generation reaction.

1. Introduction

Beryllium (Be) is a candidate material for neutron multipliers for fusion reactors. However, some issues concerning the use of this material are anticipated, such as the hydrogen generation reaction with vapor and volumetric swelling at high temperatures [1]. Hydrogen generation via an oxidation reaction of Be with water vapor at high temperatures which may result in a hydrogen explosion accident should reduce the safety hazards associated with fusion reactors. This safety issue would have a substantial impact on fusion reactors such as demonstration fusion power plant (DEMO) reactors, especially at the blanket operating temperature. Therefore, the use of advanced neutron multipliers with improved high-temperature stability instead of the existing Be neutron multipliers is desired for pebble bed blanket design with a water coolant.

Be intermetallic compounds (beryllides) are the most promising materials for advanced neutron multipliers and exhibit high stability at high temperatures for the pebble bed blankets of DEMO reactors [2–4]. However, beryllides are too brittle to allow the production of pebbles. Establishing synthesis and granulation techniques for beryllides is a key issue facing advanced neutron multiplier development. A study on advanced neutron multipliers has been started between Japan and the EU in the DEMO R&D of the International Fusion Energy Research Centre (IFERC) project as a part of the Broader Approach (BA) activities. The authors have successfully established the beryllide fabrication process by a plasma sintering method for not only beryllide block but also the rod [5,6].

In this study, the recent progress in the development of beryllide synthesis, granulation techniques and the characterization of these materials is described.

2. Experimental

Firstly, two processes of direct beryllides granulation were evaluated. One involved sintering granulation under pressure-

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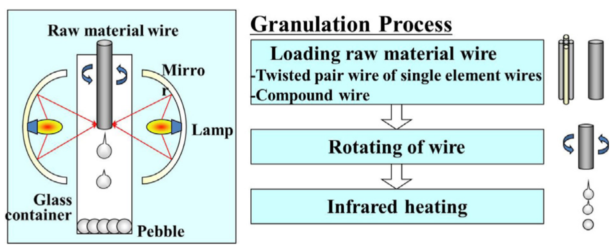

Fig. 1. Schematic of infrared heating granulation process.

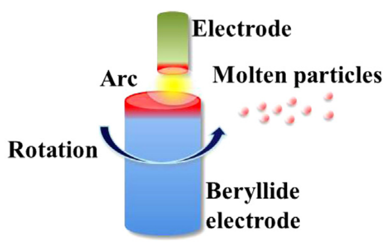

Fig. 2. Schematic of rotating electrode granulation method.

less sintering condition using mixed raw materials powder, and the other involved melting granulation by infrared heating using twisted raw material wire. The structural composition of $Be_{12}Ti$ was selected as the trial composition of beryllides.

For the trial experiment involving direct sintering granulation under pressureless conditions, the preparation of the raw materials for granulation started with mixing elemental powders. Be powder (Materion Brush, USA, 99.4 wt.%) and Ti powder (Kojundo Chemical Laboratory, Japan, 99.9 wt.%) were mixed using an automatic mortar (RM200, Retsch, Germany) to obtain a composition of 92.3 at.% Be and 7.7 at.% Ti (the stoichiometric composition of $Be_{12}Ti$). The particle sizes of the Be and Ti powders were less than 45 μm. The raw mixed elemental powder was granulated by hand using polyvinyl alcohol (PVA) as a binder paste. Sintering without pressure was performed at 1473 K for 3 h.

Concerning the trial experiments for direct melting granulation, as a first step, a preliminary examination of the pebble fabrication of Ti–Al intermetallics as a simulating material of beryllide was started using an infrared heating method. A schematic illustration of the infrared heating granulation process is presented in Fig. 1. The main processes of this melting method comprise loading the raw material wire, rotating the wire and applying infrared heating. A twisted pair of single-element wires or a compound wire is loaded to the top of chamber as the raw material. For the twisted pair wire, synthesis of the intermetallic compounds and granulation were performed simultaneously. This method was also applied for the granulating method with reduced thermal shock and contamination. The purity and wire diameter of Ti and Al were 99.9 wt.% and ø1 mm (Nilaco Corporation, Japan), respectively. The infrared melting using this twisted pair wire of Ti and Al was performed at a rotating speed of 20 rpm under an Ar atmosphere.

From the viewpoints of the inhibition of Be evaporation, melting granulation with a rapid heating rate was considered suitable for beryllide granulation. Thereafter, a rotating electrode granulation method was selected because the experience base for its use is broad, not only for Be pebbles but also for metallic pebbles in industry in general. A schematic of the rotating electrode granulation method (REM) is presented in Fig. 2. In this granulation process, molten beryllide particles were produced at the top of a heated beryllide electrode that was rotated along its longitudinal axis. The molten beryllide was centrifugally ejected and formed droplets that solidified into spherical particles.

However, a beryllides electrode in the shape of a rod was necessary for granulation using the rotating electrode method. Powder metallurgy, including hot isostatic pressing (HIP) and casting, has been proposed for the synthesis of beryllides [4]. However, these methods face some issues such as complicated process, difficulty in achieving composition control, contamination of impurities and low reproducibility. In particular, a HIPed beryllide electrode is brittle with less tolerant for thermal shock during granulation using the rotating electrode method. These previous synthesis methods are not suitable for beryllide pebble fabrication.

Thus, a plasma sintering synthesis method was selected for fabrication of the beryllide electrode. This method is simple and easy to control. Plasma sintering is a non-conventional consolidation process that consists of plasma generation, resistance heating and pressure application [7]. The plasma discharge results in particle surface activation that enhances sinterability and reduces high-temperature exposure. Pressure application assists the densification process by enhancing sintering, thus further reducing the high-temperature exposure of the consolidating powders. Consequently, the plasma-sintered material has higher ductility than its HIPed counterpart because of particle surface activation [8].

Beryllides are very brittle; however, the ductility of Be is higher than that of beryllides. Therefore, improvement of the ductility of the plasma-sintered electrode using the Be phase was evaluated. To determine the effects of the plasma sintering conditions on the compositional structure of the plasma-sintered electrode, plasma sintering was performed at 1273 K for 40 min, 1273 K for 5 min, 1173 K for 5 min, and 1073 K for 5 min. Plasma sintering apparatus (KE-Pas III) made by KAKEN Co. Ltd. was used.

Beryllide electrodes of a Be–Ti intermetallic compound were synthesized from mixed pure Be and Ti powders. Be powder (Materion Brush Inc., 99.4 wt.%) and Ti powder (Kojundo Chemical Laboratory Co. Ltd., 99.9 wt.%) were mixed using an automatic mortar (RM200, Retsch, Co. Ltd.). The diameters of the Be and Ti powders were less than 45 μm. The mixed powder composition was 92.3 at.% Be and 7.7 at.% Ti, which is the stoichiometric composition of $Be_{12}Ti$. After fabrication of the plasma sintered beryllide rods, those were granulated by the REM with rotating speed and discharge current of 6000 rpm and 70 A, respectively. In addition to this, Be-V pebble was granulated by the rotating electrode method using by the plasma sintered Be-7.7 at.% V electrode.

To evaluate the reactivity of the beryllide pebbles with water vapor at high temperatures compared with that of pure Be pebbles, hydrogen generation reaction experiments at 1273 K under an Ar gas atmosphere with a H_2O concentration of 10,000 ppm under a flow of 300 cm^3/min using thermo-gravimetric apparatus (TG; TG-8110, Rigaku, Japan) in conjunction with gas chromatography (GC; CP-4900, Agilent, USA) were conducted. The TG analysis was conducted at a heating rate of 10 K/min. The Ar gas containing 10,000 ppm H_2O was controlled by water vapor generation equipment (HUM-1E, Rigaku, Japan) and injected at 1273 K.

3. Results and discussion

Fig. 3 presents photographs of the pebbles before and after sintering. Pebbles could not be fabricated by direct sintering under pressureless conditions because of evaporation of Be and its nature not to consolidate.

A photograph of Ti–Al intermetallic pebbles obtained using the infrared heating process is presented on the left side of Fig. 4. Infrared melted pebbles have metallic luster. This preliminary fabrication indicates that Ti–Al intermetallics could be directly synthesized and granulated using the infrared melting method. As a next step, a trial fabrication experiment of Be–Ti pebbles was performed using the infrared heating granulation process. The purity

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