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Deuterium retention and desorption properties of ternary beryllide pebbles as advanced neutron multipliers

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HIGHLIGHTS

- The ternary beryllide pebbles were successfully fabricated by the novel process.
- The ternary pebbles exhibited the better deuterium desorption properties and lower total retention than Be.
- The ternary beryllide pebbles indicated the lower activation energy for the deuterium desorption compared to Be sample.

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ABSTRACT

Beryllium intermetallic compounds (beryllides) such as $Be_{12}Ti$ and $Be_{12}V$ are the most promising advanced neutron multipliers in demonstration (DEMO) power reactors because of their low swelling and high stability at high temperature. In this work, single-phase $Be_{12}Ti$ and $Be_{12}V$ pebbles produced by a novel process combining plasma sintering and rotating electrode methods were successfully fabricated. In parallel to the successful fabrication of binary beryllide pebbles, new research and development on ternary beryllides was begun to determine preferable pebble compositions in terms of mass production and improved properties. The desorption properties and retention of hydrogen isotopes (deuterium) using the newly developed ternary beryllide pebbles indicated that the ternary pebbles exhibited better deuterium desorption properties, such as lower starting-up temperature, total retention, and activation energy for deuterium desorption, than these for beryllium.

1. Introduction

As an advanced neutron multiplier for demonstration (DEMO) fusion reactors, beryllium intermetallic compounds (beryllides) have received considerable attention. Because of their high stability at high temperature, studies on beryllide fabrication have been performed, focusing not only on the development of pebble fabrication methods [1] but also on the characterization of these materials [2].

The authors have developed a novel process for beryllides employing plasma sintering [3] and rotating electrode [4] methods as a framework for Broader Approach activities in DEMO R&D for the International Fusion Energy Research Centre (IFERC) project. For Be–7.7 at.% Ti pebbles fabricated using this novel process, the variation of the phase composition was unexpectedly confirmed by the peritectic reaction due to re-melting, such that it was necessary to add a homogenization heat treatment for single-phase $Be_{12}Ti$ beryllide pebbles. For the preferable composition from the viewpoints of tritium breeding ratio, mass production, and stability, Be–7.7 at.% V pebbles with a stoichiometric ratio of $Be_{12}V$ were fabricated, without the need for any additional treatment. However, even though single-phase $Be_{12}V$ pebbles were successfully fabricated without homogenization using the rotating electrode method with plasma-sintered rod beryllide, their collapse strength at room temperature was relatively low [5]. This low strength may be problematic because of the physical vibration and purge gas pressure in the blanket. To overcome this mechanical disadvantage, Be–Ti–V ternary beryllides were investigated, and it was demonstrated that $Be_{12}Ti_{0.3}V_{0.7}$ and $Be_{12}Ti_{0.1}V_{0.9}$ pebbles consist of dual phases, $Be_{12}Ti$ and $Be_{12}V$, which are much more stable at high temperature than the Be phase. Concerning the collapse strength [5], the ternary beryllide pebbles clearly exhibited higher strength than the single-phase $Be_{12}V$ pebbles.

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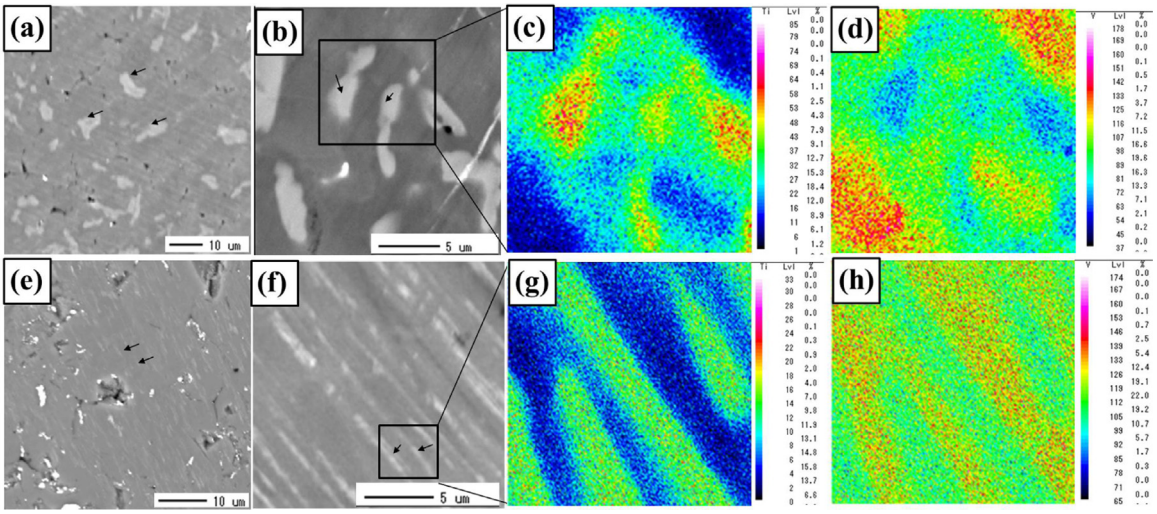

Fig. 1. Cross-sectional SEM composition images of (a, b) $Be_{12}Ti_{0.3}V_{0.7}$ and (e, f) $Be_{12}Ti_{0.1}V_{0.9}$ and those areal analyses for each element, (c, g) Ti, (d, h) V, respectively. The black arrows indicate $Be_{12}Ti$ while others do $Be_{12}V$.

The tritium retention property of beryllium generated by the following reactions is of great importance; Chen estimated the production of tritium to be several tens of kilograms [6], with reports of the production of 24 kg of tritium from 390 tons of beryllium:

$$^{9}Be + n \rightarrow 2^{4}He + 2n(En > 2.7\,MeV) \quad (1)$$

$$^{9}Be + n \rightarrow {}^{7}Li + {}^{3}H\,(En > 10.5\,MeV)$$
$$^{7}Li + n \rightarrow {}^{4}He + {}^{3}H + n \quad (2)$$

$$^{9}Be + n \rightarrow {}^{6}He + {}^{4}He\,(En > 0.6\,MeV)$$
$$^{6}He \rightarrow {}^{6}Li + \beta^{-} + v$$
$$^{6}Li + n \rightarrow {}^{4}He + {}^{3}H \quad (3)$$

Considering the importance of the hydrogen retention property, the deuterium retention and desorption properties of $Be_{12}Ti_{0.3}V_{0.7}$ and $Be_{12}Ti_{0.1}V_{0.9}$ pebbles were investigated in this study using deuterium ion irradiation and thermal desorption spectrometry.

2. Experimental

A novel process for pebble fabrication was reported in previous studies [1,3,4]. For the fabrication of beryllide rods, Be, Ti and V powders (purity: 99.5, 99.9%, and >99.9%, respectively) were mixed with $Be_{12}Ti_{0.3}V_{0.9}$ and $Be_{12}Ti_{0.1}V_{0.9}$ and plasma-sintered at 1073 K for 5 min. These rods were granulated into pebbles of approximately 1 mm in diameter using the rotating electrode method with a rotating speed of 6000 rpm and a discharge current of 60 A. Scanning electron microscopy (SEM) and an electron probe micro-analyzer (EPMA, JXA-8530F, JEOL, Japan) were used for micro-structure observation and quantitative/qualitative analyses by using analyzing crystal (LDE2H) in the WDS (wavelength dispersive x-ray spectrometer) apparatus.

To evaluate the deuterium retention and absorption properties of the ternary beryllides, thermal desorption spectroscopy (TDS) was performed on D^{+}-irradiated beryllides, irradiated with 3-keV $D_2{}^{+}$ ions with a fluence of 1×10^{20}–1×10^{23} ions/m^2 at room temperature. For deuterium implantation range, it was estimated to approximately 70 nm, which is near surface. The pebbles were set in a tantalum sheet for easy handling for the TDS. To evaluate the apparent activation energy for deuterium desorption in the pebbles [7], the dependence of the heating rates with 0.2–5 K/s was evaluated, and the activation energy (E) is determined from the following relation:

$$\ln(\beta/Tp^2) = -E/kTp - \ln(E/kv), \quad (4)$$

where β is the heating rate (K/s), Tp is the desorption peak temperature (K), E is the apparent activation energy (eV), k is the Boltzmann constant, and v is the rate constant.

3. Results and discussion

SEM examination and qualitative analyses were performed to confirm the phase composition, as shown in Fig. 1. Both pebbles consisted of dual phases, $Be_{12}Ti$ and $Be_{12}V$ while $Be_{12}Ti_{0.1}V_{0.9}$ (Fig. 1(e)) contained large fraction of $Be_{12}V$ with fine and long-shaped $Be_{12}Ti$ phase. Mapping analyses of the composition using the EPMA are also presented, indicating that the $Be_{12}Ti$ and $Be_{12}V$ phases were well dispersed without phase agglomeration, which is effective for increasing the

mechanical strength. In previous studies [8–10], both $Be_{12}Ti$ and $Be_{12}V$ were shown to be much more stable than Be at temperatures above 973 K. Furthermore, the superiority of their collapse strength was clearly demonstrated by compressive experiments, which revealed that the collapse strength of the dual-phase pebbles was twice as high as that of single-phase $Be_{12}V$ pebbles [5]. Regarding to the dual phases in the pebbles, Fig. 2 shows that the grain size near the surface was considerably fine. In evaluating material strength, the Hall–Petch law is predominantly used to explain the relationship between grain size and strength. Not only the mechanical properties but also the hydrogen retention properties are apparently important because beryllium and its compounds as neutron multipliers produced tritium, which corresponds to several tens of kilograms when loading several hundreds of tons of multipliers [6].

To evaluate the hydrogen isotope behavior, in the present study, $D^{+}{}_2$ irradiation of ternary beryllides and thermal desorption spectroscopic measurements were performed. Figs. 3 and 4 show the deuterium desorption rate of the irradiated $Be_{12}Ti_{0.3}V_{0.7}$ and $Be_{12}Ti_{0.1}V_{0.9}$ beryllide pebbles, respectively. The main peaks are located near 600 K, which is relatively low compared with the peaks of Be metal [11]. Moreover, it is not surprising that as the fluence increased, the peak intensity increased. A secondary peak is observed in the shape of the peaks, which have a long tail in the high-temperature region; this result differs greatly from that of

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