A Comparison of Li₄SiO₄ Pebble Characteristics according to Slurry condition

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1. Introduction

Tritium is one of the fuels needed for nuclear fusion energy generation, and it is rarely exist in nature (about 10⁻¹⁸ times that of natural hydrogen) so, needs the fabrication process additionally. The tritium breeder (TB), a material that produces tritium, is a lithiumbased material, and neutrons generated by nuclear fusion and lithium of TB reacted with the neutrons generated by the fusion reaction to produce tritium in the Test Blanket Module (TBM). In order to increase the efficiency of tritium production, the maximum amount of TBs should be stored in TBM. Therefore, the most basic requirement for TB is to form a spherical pebble shape with a diameter of 1mm or less, and to stably deliver the TB to TBM, it has to have high mechanical strength [1]. Recently, Li₄SiO₄ and Li₂TiO₃ were being considered intensively as TB materials. Among them, Li₂TiO₃ is a promising material because it has high phase stability and mechanical strength, so it is easy to handle [2]. But, Li₂TiO₃ has a lower lithium density than Li₄SiO₄, and thus the tritium production efficiency is relatively low when made of pebble having the same size and density [3].

In this study, research on the fabrication of Li_4SiO_4 TBs with high mechanical strength and high sphericity was conducted. A freeze-drying method was used to fabricate the Li_4SiO_4 TB, and the characteristics of the pebble were evaluated according to the concentration of the Li_4SiO_4 slurry.

2. Methods and Results

In this section, some of the techniques using freezedrying are described. Freeze-drying is a process that obtain the Li₄SiO₄ green pebble after drop the Li₄SiO₄ slurry into liquid nitrogen.

2.1 Li₄SiO₄ synthesize

Li₄SiO₄ was synthesized by solid-state synthesis method. Lithium Hydroxide and Silicon dioxide were used as precursor, and their molar ratio was 4:1. Precursors were mixed by planetary milling for 3hours. And then, the mixed precursors were calcined at 600°C for 7 hours.

2.2 Li₄SiO₄ pebble fabrication

The Li₄SiO₄ pebble was fabricated by dropping method. First, Li₄SiO₄ slurry was fabricated by Li₄SiO₄ powder and polyvinyl alcohol (PVA) into deionized water according to each weight percentage condition (1, 3, 5wt% PVA and 50wt% Li₄SiO₄ powder). At this time, the weight percentage of Li₄SiO₄ powder and PVA is calculated by deionized water weight. Li₄SiO₄ pebbles were fabricated by dropping the Li₄SiO₄ slurry into liquid nitrogen using a syringe and froze Li₄SiO₄ pebbles were dried in a freeze-dryer for 24hours. Li₄SiO₄ green pebbles were sintered at 900°C for 3 hours. Second, after the concentration of the PVA solution was determined, Li₄SiO₄ slurry was prepared according to each weight percentage of Li₄SiO₄ powder in the determined concentration PVA solution for determining the mixing ratio of Li₄SiO₄. The fabrication method of Li₄SiO₄ pebble and sintering method was the same as the first step.

2.3 Results

The XRD patterns of the synthesized Li_4SiO_4 powder by solid-state method was shown in Fig.1. The XRD patterns was compared with the reference (PDF #10-076-1085). As shown in Fig. 1, only sharp diffraction peaks of Li_4SiO_4 were confirmed without an impurity peak.

The morphology of sintered Li4SiO4 pebble according to PVA wt% (1-50LSP, 3-50LSP, 5-50LSP) was shown in the optical microscopy (OM) pictures, Fig.2. It was confirmed that the size of Li₄SiO₄ pebbles gradually increased as the PVA wt% increased. The average diameters of pebbles measured through OM were 0.94, 1.035, and 1.27mm, respectively. In addition, the crush load of the pebble was analyzed at 7.83, 14.94, 11.25N. It was confirmed that the crush load of 3-50LSP was 1.91 and 1.328 times higher than 1-50LSP and 5-50LSP, respectively. The crush load was measured by 10 pebbles for increasing reliability of the measured value, and Fig. 3 shows the average diameter and average crush load of the pebbles according to the PVA content in a graph. The error bar was set by the average of amount of maximum and minimum. Through the graph analysis in Fig. 3, it was determined that 3wt% of the PVA solution concentration was the optimal condition.

After determining the concentration of the PVA solution, the mixing ratio of Li_4SiO_4 was determined. The amount of Li_4SiO_4 powder was calculated according to the mass of deionized water calculated during slurry preparation, and 50, 75, and 100wt% of Li_4SiO_4 powder were mixed, respectively, and each pebble was named 3-50LSP, 3-75LSP, and 3-100LSP. The morphology of the pebble prepared according to the Li_4SiO_4 powder wt% is shown in Fig. 4. In each

Li₄SiO₄ pebble, the sphericity improved as the Li₄SiO₄ powder content increased, and the measured average diameters were 1.035, 1.05, and 1.07 mm, respectively, and the size of the Li₄SiO₄ pebble did not change significantly. The compressive strengths were 14.94, 14.78, and 18.13N, respectively, and it was confirmed that the compressive strength of 3-100LSP was 1.21 and 1.22 times higher than those of 3-50LSP and 3-75LSP, respectively. [Fig.5]



Fig. 1. XRD result of synthesized Li₄SiO₄ powder at 600°C for 7hours.



Fig. 2. The optical microscopy images of Li_4SiO_4 pebbles according to PVA wt% (a) 1wt%, (b) 3wt%, and (c) 5wt%



Fig. 3. The average crush load and diameter of $\rm Li_4SiO_4$ pebble according to PVA wt%



Fig. 4. The OM images of Li4SiO4 pebbles according to Li4SiO4 powder wt% (a) 50wt%, (b) 75wt%, and (c) 100wt%



Fig. 5. The average crush load and diameter of Li₄SiO₄ pebble according to Li₄SiO₄ powder wt%

3. Conclusion

Li₄SiO₄ pebble was successfully prepared by Freezedrying. To fabricate a Li₄SiO₄ pebble with high mechanical strength, the wt% of PVA was determined, and the wt% of Li₄SiO₄ powder was determined based on the determined wt% of PVA. As a result, it was analyzed that the Li₄SiO₄ pebble prepared as a slurry containing 3wt% PVA and 100% Li₄SiO₄ based on distilled water, respectively, was the best in terms of size and mechanical strength.

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REFERENCES

[1] R. Knitter, P. Chaudhuri, Y.J. Feng, T.Hoshino, and I.K. Yu, Recent developments of solid breeder fabrication, J. Nucl. Mater. 442, 420-424, 2013

[2] C.L. Yu, F.Wang, A.L. Zhang, D.P. Gao, S.Y. Cao, Y.Y Guo, H.B. Hui, X. Hao, D.Y. Wang, and K. Yanagisawa, Preparation of β -Li₂TiO₃ pebbles by a modified indirect wet chemistry method, Fusion Sci. Technol. 101, 73-79, 2015

[3] M. Xiang, Y. Zhang, Y. Zhang, S. Liu, H. Liu, C. Wang, and C. Gu, Preparation of Li₂TiO₃-Li₄SiO₄ core-shell ceramic pebbles with enhanced crush load by graphite bed process, J. Nucl. Mater. 466, 477-483, 2015