

Fabrication Methods of Zirconia based IMF by Dry Milling and Wet Milling

Chang-Young Joung, Si-Hyung Kim, Su-Chul Lee and Han-Soo Kim

Korea Atomic Energy Research Institute, P.O. Box 105, Yuseong, Daejeon, Korea, 305-600

Tel : 82-42-868-2519, Fax : 82-42-861-3642, E-mail : joung@kaeri.re.kr

1. Introduction

Yttria stabilized zirconia (YSZ) is being studied at the PSI (Paul Scherrer Institute) as an inert matrix for an advanced nuclear fuel [1]. This fuel is used for burning plutonium excesses in light water reactors. Zirconium oxide is a potential inert matrix candidate [2, 3]. Cubic zirconia has a small neutron cross-section, a high melting point, and is stable under an irradiation. In a routine powder process, ZrO₂-based IMF (Inert Matrix Fuel) powder was milled by using a ball mill or attrition mill. In this case, the powder mixture sticks fast to the inner wall of a mill jar. After that the milling process does not go further but the powder sticks harder to the wall. The IMF pellets have been fabricated with a dry continuous type attrition mill which was specially designed to obtain a sinterable, homogeneous powder mixture. The powder mixture was passed through the mill more than 12 times, then compacted and sintered into a pellet. Microstructure was homogeneous but sintered density was less than 92% of the T.D. (therotical density) which did not meet the specification [4].

In this study, the powder mixture was milled by two different methods using a planetary mill. The effects of the powder treatment methods on the sintering behavior were investigated by an estimation of the particle size of the milled powder, the sintered density and the microstructure of the sintered pellet. When the powder was milled by the wet process with two different sizes of ball media, the IMF pellet with a high density, or more than 95%T.D. and a homogeneous microstructure was fabricated.

2. Experimental

Raw materials of the IMF pellet are ZrO₂, Y₂O₃, Er₂O₃ and CeO₂. Zirconium oxide is used as a matrix material and yttrium oxide is a kind of phase stabilizer to obtain a single cubic phase. Erbium is added as a burnable poison absorber to compensate for the reactivity. Cerium oxide is used as a surrogate material of PuO₂. The mixing composition of the starting materials was ZrO₂+10at%Y₂O₃+7at%Er₂O₃

+15at%CeO₂. The powder mixtures were obtained by adding weighed amounts of the raw powders into a turbula mixer for 1h. After that, the powder mixtures were milled by a dry process or a wet process with different sizes of ball media. The milling was performed in a planetary mill for 0.5~6 hours at 300rpm using a zirconia jar containing 20mm zirconia balls for SINGLE or using a zirconia jar containing both 8mm and 20mm balls together for DOUBLE. In the wet milling process, ethanol or CCl₄ was used as a liquid milling media. After the wet milling, the powder was separated from the balls and liquid media and then dried in an oven. The milled powder was pressed with a compaction pressure of 300MPa, by using a double-acting hydraulic press. Sintering of the compacts was carried out in a box furnace at 1650°C for 5 to 10 hours in air. The density of the pellets was measured by the immersion method, and the phases of each pellet were analyzed by XRD. The XRD analysis was performed by using a Ni filtered CuK α radiation. In order to observe the microstructures of the IMF pellets, thermal etching was carried out at 1450°C for 1 hour in air. The average grain size was determined using the linear intercept method.

3. Results

Particle size of the IMF powder milled with a dry milling or wet milling is shown in Fig. 1. In the case of the DOUBLE process, particle size with a dry milling and wet milling is decreased with an increasing milling time.

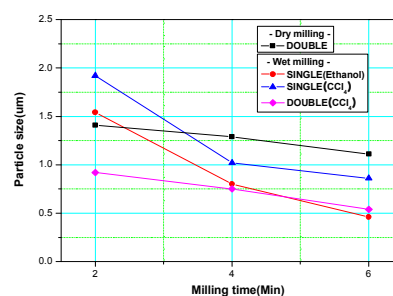


Fig. 1 Particle size of IMF powder treated with (a) dry process, (b) wet process.

The variation of the green density and sintered density of the IMF is shown in Fig. 2.

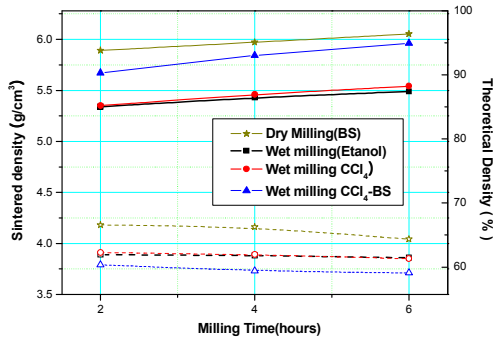


Fig. 2 The change in green density and sintered density of IMF pellets.

The density of the compacts was slightly decreased with an increasing milling time. In the case of a wet milling, the sintered density with the SINGLE method is almost constant regardless of the liquid media. But, the sintered density for the wet milling process with the DOUBLE method was higher than that for the dry milling.

The XRD patterns for each pellet specimen sintered at 1650°C shows the pattern of a homogeneous single cubic phase.

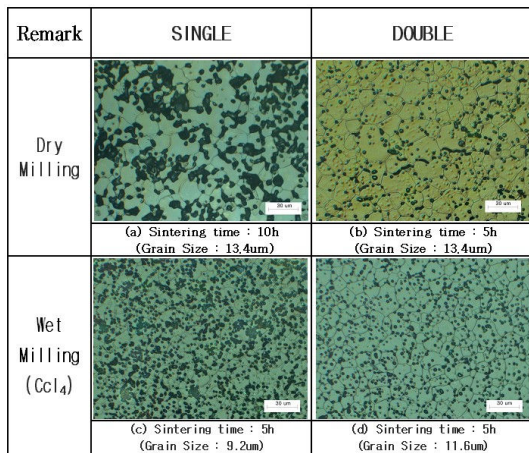


Fig. 3 Microstructures of IMF sintered at 1650°C in air atmosphere.

The microstructures of the IMF pellets sintered at 1650°C in an air atmosphere are shown in Fig. 3. In the case of the dry milling process, the microstructure of the IMF pellets with the DOUBLE method shows larger pores and grains than those of the pellets with the SINGLE method. In the case of a wet milling, the microstructure of the IMF pellets with the DOUBLE method is about two times larger than that of the pellets with the SINGLE method. The average grain sizes of the IMF pellet with a dry milling and a wet

milling with the DOUBLE method are about 13.4 μm and 11.6 μm, respectively. The grain growth rate of the IMF pellets with the dry milling is larger than that of the pellet with the wet milling at the same sintering parameters.

4. Conclusion

In this present work, the effect of a dry milling and a wet milling method on the sintering behavior of IMF powder is investigated by using a planetary mill.

1. The particle size of the powders milled with a dry milling and a wet milling was slightly decreased with an increasing milling time.
2. The sintered density was higher for the pellet fabricated by the DOUBLE method than that for the SINGLE method.
3. The average grain size of the IMF pellets for the dry milling was larger than that for the wet milling.

ACKNOWLEDGEMENT

This study was performed under the auspices of the Korea Ministry of Science and Technology.

REFERENCES

- [1] Ch. Hellwig, M. Pouchon, R. Restani, F. Ingold and G. Bart Eng. J. Nucl. Mater., Vol. 62, pp. 138-166, 1976.
- [2] Ch. Hellwig, U. Kasemeyer, G. Ledergerber, B.-H. Lee, Y.-W. Lee and R. Chawla, Ann. Nucl. Energy Vol. 30, pp. 287, 2003.
- [3] J.M. Paratte and R. Chawla. Ann. Nucl. Energy Vol. 22, pp. 471, 1995.
- [4] Y.W. Lee, H.S. Kim, S.H. Kim, C.Y. Joung, S.H. Na, G. Ledergerber, P. Heimgartner, M.A. Pouchon and M. Burghartz, J. Nucl. Mater. Vol. 274, pp. 7, 1999.