Thermophysical Properties of Rare-Earth-Stabilized Zirconia and Zirconate Pyrochlores as Surrogates for Actinide-Doped Zirconia

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Zirconate pyrochlore, $A_2Zr_2O_7$, where A = Pu or a minor actinide (MA), is expected to be a nuclear fuel and a form of waste. Zirconium is inactive for thermal neutrons. Furthermore, zirconium oxide is stable in a radioactive environment and also has chemical stability. Therefore, such zirconate pyrochlores can burn out recycled Pu and MA effectively. In this study, the lattice structure and thermophysical properties were evaluated for the system $A_2Zr_2O_7$ using rare-earth metals (La, Nd, Sm, Gd, Dy, and Y) as surrogate actinides.

The results of X-ray diffractometry (XRD) and Raman spectroscopy showed that $La_2Zr_2O_7$, $Nd_2Zr_2O_7$, $Sm_2Zr_2O_7$, and $Gd_2Zr_2O_7$ had pyrochlore structures, whereas $Dy_2Zr_2O_7$ and $Y_2Zr_2O_7$ had defect fluorite structures. The smaller the difference of ionic radius between A and Zr, the more stable the defect fluorite structure is compared to the pyrochlore structure [1].

For zirconate pyrochlores, the thermal expansion measured by high temperature XRD decreased in the order: $Gd_2Zr_2O_7$, $Sm_2Zr_2O_7$, $Nd_2Zr_2O_7$, and $La_2Zr_2O_7$, with the increasing ionic radius of A. On the other hand, elastic moduli and Debye temperatures were obtained from ultra-sound velocities (longitudinal and transverse) and densities of the samples. For example, Young's modulus decreased in the order: $Nd_2Zr_2O_7$, $Sm_2Zr_2O_7$, and $Gd_2Zr_2O_7$, contrary to the thermal expansion. These thermophysical properties can be explained by the binding energy for the pyrochlore crystal [2].

[1] Y. Tabira, R.L. Withers, J.C. Barry, and L. Elcoro, *J. Solid State Chem.* **159** 121 (2001).
[2] K.V.G. Kutty, S. Rajagopalan, and C.K. Mathews, *Mat. Res. Bull.* **29** 759 (1994).