Contribution ID: **245** Type: **POSTER**

Thermal conductivity of non-stoichiometric (Pu0.928Am0.072)O2-x

Tuesday 27 June 2017 17:30 (1h 30m)

Am-bearing oxide fuel is considered as the fuel candidate for Fast Reactor to reduce the amount of high level radioactive waste. Thermo-physical properties of oxide fuel such as thermal conductivity and diffusion coefficient are very sensitive to the change of O/M ratio. Am is one of high produced minor actinide in irradiated nuclear fuel and its dioxide (AmO_{2-x}) has higher oxygen potential than PuO_2. Therefore, in the development of Am-bearing MOX fuel, it is very important to evaluate the effect of Am on thermo-physical properties. In this study, thermal diffusivity of $(Pu_{0.928}Am_{0.072})O_{2-x}$ was measured to evaluate the Am effect on thermal conductivity in the hypo-stoichiometric region.

Thermal conductivity of non-stoichiometric $(Pu_{0.928}Am_{0.072})O_{2-x}$ (x = 0.000 −0.058) was evaluated using experimentally measured thermal diffusivity by laser flash method, the bulk density and the heat capacity.

The obtained thermal conductivity was analyzed using oxygen potential data of $(Pu_{0.928}Am_{0.072})O_{2-x}$ to evaluate the effect of Am on thermal conductivity in the hypo-stoichiometric region. The oxygen potential data shows that Am and Pu in $(Pu_{0.928}Am_{0.072})O_{2-x}$ is reduced to trivalent over O/M = 1.964 (x = 0.036), and below O/M = 1.964, respectively. Therefore, it is considered that the effect of anion on thermal conductivity switches from Am to Pu, and the thermal conductivity discontinuously decrease at O/M = 1.964. However, the experimental results showed that thermal conductivity continuously decreased with reduction of the O/M ratio. This suggests that Am has almost comparable effect on the thermal conductivity with Pu in the hypo-stoichiometric region.

The O/M ratio dependence of thermal conductivity was well explained by the evaluation with the phonon transport model and slack model. This evaluation showed that the decrease of thermal conductivity was caused by the increase of oxygen vacancy and ionic radius differences between Pu^{4+} and Pu^{3+} or Am^{4+} and Am^{3+}. In addition, it is considered that Am has almost identical effect on the thermal conductivity with Pu due to the similar ionic radius of Pu^{4+} and Am^{4+} or Pu^{3+} and Pu^{3+}.

Country/Int. Organization

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Session Classification: Poster Session 1

Track Classification: Track 5. Fast Reactor Materials (Fuels and Structures) and Technology