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Oxidation behavior of Am-containing MOX fuel pellets in air

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Abstract

Americium-containing MOX (Am-MOX) fuels were subjected to heating tests using thermogravimetric and differential thermal analysis measurements in a flowing gas atmosphere of dry air to investigate the effect of Am addition on oxidation behavior of MOX fuel. Three kinds of Am-MOX fuel pellets containing 3, 5 and 10 wt.% Am were prepared for the examination together with MOX fuel and UO_2 fuel pellets as references. Sintered fuel pellets were heat-treated to adjust the oxygen-to-metal ratio to 2.00 and were crushed into small pieces. The weight gain due to oxidation was monitored and the pulverization behavior of the fuel pellets was observed. The specimens were analyzed by X-ray diffraction in order to investigate the change of the phase relation. The specimens were subjected to programmed rate heating test (6 K/min) from room temperature to 1073 K. The UO_2 pellet specimen was oxidized rapidly to an O/M ratio of 2.67 (i.e. U_3O_8 was formed) and it was pulverized easily. The specimens of MOX fuel and Am-MOX fuel pellets, however, were oxidized gradually to O/M ratio around 2.3 (i.e. MO_{2+x} and/or M_4O_9 were formed) and there was no pellet crumbling. Although the oxidation rate slightly decreased with increasing the Am content in the Am-MOX fuels, the oxidation curve shapes of Am-MOX fuels were similar to the curve of MOX fuel. The isothermal heating test was carried out for the specimens of MOX, 3 wt.% Am-MOX and 5 wt.% Am-MOX fuels. The kinetic analysis of the oxidation in the isothermal heating test was evaluated by the Johnson-Mehl equation.

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1. Introduction

Uranium and plutonium mixed oxide (MOX) fuels containing several percent of minor actinides (MAs) and fission products are promising candidates for a closed nuclear cycle system based on a fast reactor [1]. Many novel and practical findings have been obtained for MA-containing MOX (MA-MOX) fuels related to items such as development of their fabrication process, measurement of several of their thermophysical and themochemical properties, and observation of their irradiation behavior [2]. In addition to the knowledge noted above, the oxidation properties of the fuels are also important for understanding the fundamental mechanism of oxidation of actinide oxides, for the design of suitable schemes for the fuel fabrication and reprocessing, and for purpose of reactor safety. Understanding of the pulverization behavior due to the change of crystal structure resulting from oxidation of the fuel is necessary to design suitable schemes for the utilization of dry recovery powders from the scrap pellet and for expediting the removal of volatile and gaseous fission products before the fuel is dissolved in nitric acid (adoptability of voloxidation). From the aspect of safety, it is desirable that the fuel pellets maintain its mechanical integrity if it is exposed to an oxidized environment at elevated temperature in the case of accident.

Although oxidation properties of UO_2 and MOX fuels in air were reported so far [3-11], no research has yet been carried out to investigate the oxidation behavior of the MA-MOX fuels. In order to accumulate the basic data and knowledge concerning the oxidation behavior of MA-MOX fuel, Am-containing MOX (Am-MOX) fuels were subjected to heating tests under the flowing gas atmosphere of dry air in the present study. The weight gain due to oxidation was monitored and the pulverization behavior of the fuel pellets was observed. The change of the phase relation of the specimens was also analyzed. The experimental results of oxidation behavior of Am-MOX fuels were compared with those of the data for UO_2 and MOX fuels.

2. Experimental

2.1. Specimens

Am-MOX fuel pellets were fabricated adopting the conventional powder metallurgy method using a remote handling technique in the shielded air-tight hot cell of the Alpha-Gamma Facility (AGF) at JAEA's Oarai Research and Development Center [12]. UO_2 powder, Am-containing UO_2 powder and two kinds of Am-containing PuO_2 powders were used as raw materials. Before the sintering step, these powders were annealed at 873 K for 2 hours in a furnace with a kanthal heater to remove absorbed moisture. After the heat pre-treatment, each powder type was weighed on an electronic balance in order to adjust the amount to the predetermined weight ratio. This was followed by mixing the desired powders in an alumina ball mill for about 5 hours without solvent (dry condition). Zinc stearate was added as a binder and the powders were further mixed for 30 minutes before cold-pressing into green pellets at a pressure of about 380 MPa. The green pellets were pre-sintered at 1073 K for 2.5 hours in the furnace with the kanthal heater to remove the binder and then were sintered at 1973 K for 3 hours in a furnace with a tungsten mesh heater. The densities of the fuel pellets were determined from dimensional and weight measurements and were found to be almost the same values of 93 ± 3 % T.D.(Theoretical Density). As references, UO_2 and MOX fuel pellets were also prepared by the same fabrication procedure. Sintered fuel pellets were heat-treated to adjust the oxygen-to-metal (O/M: M= U, Pu, Am) ratio to 2.00. The ratio was confirmed gravimetrically at room temperature. All of the heat treatments and sintering processes were carried out under a flowing gas atmosphere of Ar-5 % H_2 or Ar-0.05 % H_2. The composition of the atmosphere was controlled by adding an appropriate amount of moisture.

2.2. Heating tests

After crushing the pellets into small specimen pieces, about 50 μg of each specimen of UO_2, MOX, 3 wt.% Am-MOX, 5 wt.% Am-MOX and 10 wt.% Am-MOX fuels (Pu content : 30 wt.%) was subjected to the programmed rate heating test (6 K/min) from room temperature to 1073 K under the flowing gas atmosphere of dry air. The isothermal heating test was also carried out for the specimens of MOX, 3 wt.% Am-MOX and 5 wt.% Am-MOX fuels. Both the programmed rate and isothermal heating tests were carried out by using a thermogravimetric and