Investigation of Fuel Pellet Interaction with Zirconium Dioxide and Fission Products in LWR

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1. Introduction

In light water reactors (LWR), uranium dioxide (UO_2) is widely adopted as fuel pellet, and zirconium alloys are widely applied as cladding. When a severe accident occurs in a light water reactor, the cladding temperature can increase to 1200 °C, and the oxidation rate of the cladding becomes relatively fast [1]. If the emergency core cooling system (ECCS) does not operate properly during an accident, claddings lose their mechanical integrity due to the extensive oxidation, and they can eventually go through failure. In this kind of situation, it is possible that zirconium dioxide (ZrO_2) reacts with UO_2 [2]. In addition, it is also probable that UO_2 reacts with steam and transforms to U_3O_8 [3]. Hence, it is necessary to investigate how UO_2 will react with ZrO_2 and how U_3O_8 will interact with the fission products that reside within fuel pellet. Therefore, In this study, UO_2/ZrO_2 and $U_3O_8/(Nd,Eu)_2O_3$ mixtures prepared, and the mixtures were annealed in furnaces. After annealing, the samples were analyzed using X-ray diffraction (XRD) and scanning electron microscopy (SEM). Through the analysis, interaction between the materials were studied.

2. Methods

2.1 UO_2/ZrO_2 Mixture Preparation and Heat Treatment

Two types of mixtures were prepared; UO_2-5mol%ZrO_2 and UO_2-10mol%ZrO_2. They were annealed in a furnace following the heating step shown in Fig. 1; The furnace atmosphere was maintained as 10% H_2 and 90% Ar throughout the heat treatment process.

2.2 $U_3O_8/(Nd,Eu)_2O_3$ Mixture Preparation and Heat Treatment

Four types of $U_3O_8/(Nd,Eu)_2O_3$ mixtures were prepared; U_3O_8-20mol%Nd_2O_3, U_3O_8-50mol%Nd_2O_3, U_3O_8-20mol%Eu_2O_3, and U_3O_8-50mol%Eu_2O_3. They were annealed in a furnace following the heating step shown in Fig. 2; The furnace atmosphere was maintained as air throughout the heat treatment process.

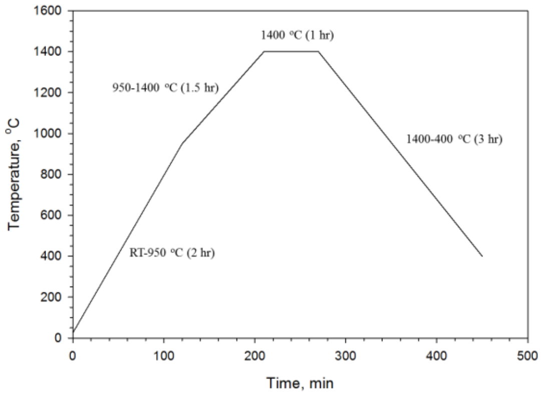

Fig. 1. Heat treatment procedure of UO_2/ZrO_2 mixtures in 10% H_2 and 90% Ar condition.

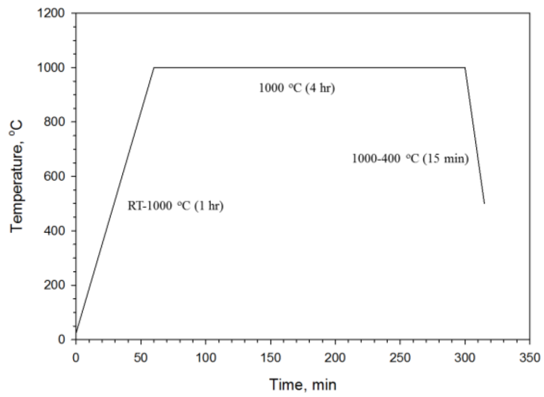

Fig. 2. Heat treatment procedure of $U_3O_8/(Nd,Eu)_2O_3$ mixtures in air.

3. Results

XRD results of UO_2/ZrO_2 mixtures are shown in Fig. 3 and Fig. 4; reference XRD data of UO_2 and ZrO_2 are shown in the figures. ZrO_2 was not found in the mixture after the heat treatment. Instead, only UO_2 was found from the mixture. Hence, in our experimental range, all ZrO_2 reacted with UO_2 and they existed as solid solution in UO_2 after the heat treatment.

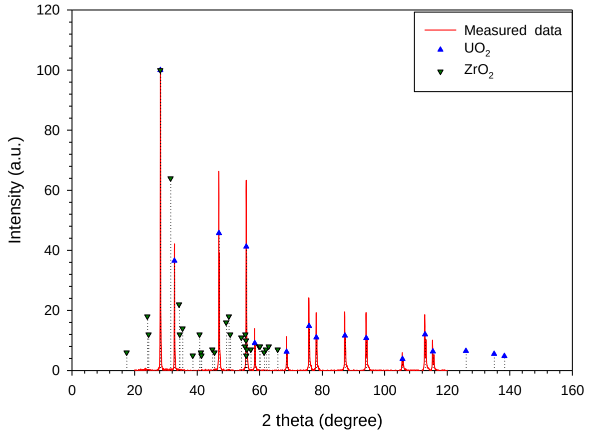

Fig. 3. XRD result of UO_2-95mol%ZrO_2 after heat treatment at 1400 °C for 1 hour in 10% H_2 and 90% Ar condition.

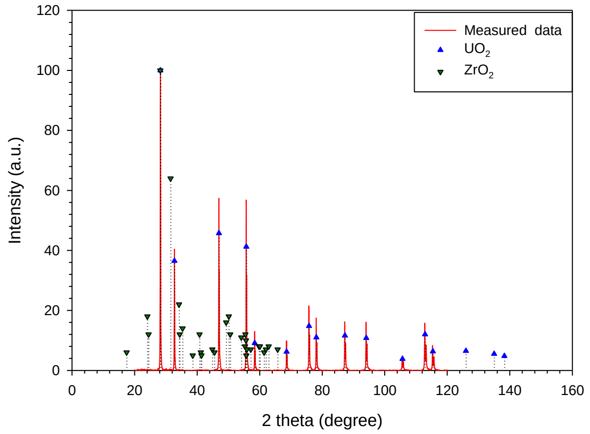

Fig. 4. XRD result of UO_2-90mol%ZrO_2 after heat treatment at 1400 °C for 1 hour in 10% H_2 and 90% Ar condition.

XRD results of U_3O_8/Nd_2O_3 mixtures are shown in Fig. 5 and Fig. 6; reference XRD data of UO_2 and U_3O_8 are shown in the figures. Nd_2O_3 was not found from all the samples after the heat treatment. Instead, materials which have a similar XRD peak with UO_2 was found from all the samples. Hence, it appears that U_3O_8 reacted with Nd_2O_3 and became $(Nd,U)O_{2+x}$.

XRD results of U_3O_8/Eu_2O_3 mixtures are shown in Fig. 7 and Fig. 8; reference XRD data of UO_2 and U_3O_8 are shown in the figures. Eu_2O_3 was not found from all the samples after the heat treatment. Instead, materials which have a similar XRD peak with UO_2 was found from all the samples. Hence, it is highly likely that U_3O_8 reacted with Eu_2O_3 and became $(Eu,U)O_{2+x}$.

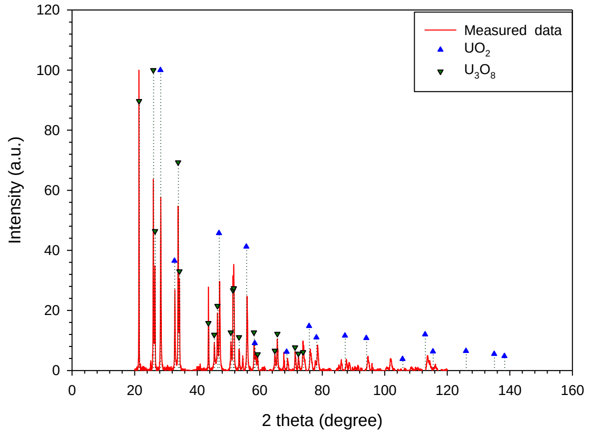

Fig. 5. XRD result of UO_2-20mol%Nd_2O_3 after heat treatment at 1000 °C for 4 hours in air condition.

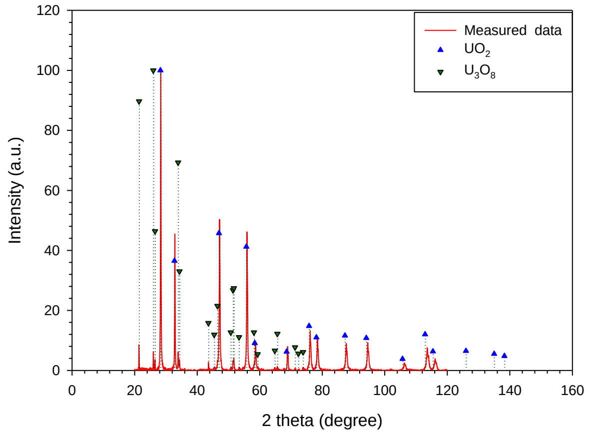

Fig. 6. XRD result of UO_2-50mol%Nd_2O_3 after heat treatment at 1000 °C for 4 hours in air condition.

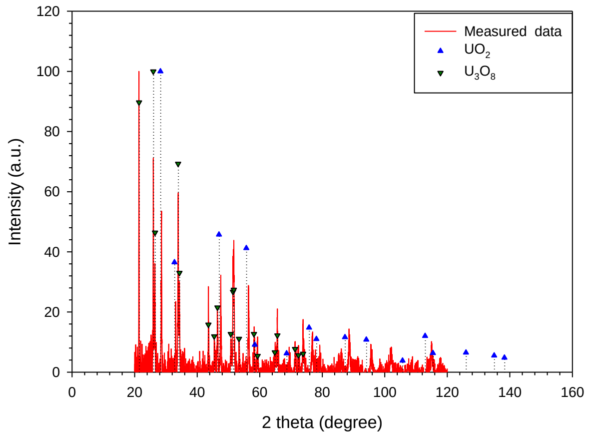

Fig. 7. XRD result of UO_2-20mol%Eu_2O_3 after heat treatment at 1000 °C for 4 hours in air condition.

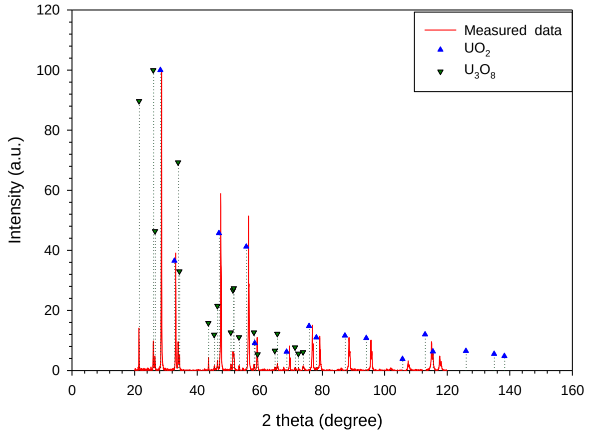

Fig. 8. XRD result of UO_2-50mol%Eu_2O_3 after heat treatment at 1000 °C for 4 hours in air condition.

4. Conclusions

Through the analysis, it was found that UO_2-5mol%ZrO_2 and UO_2-10mol%ZrO_2 mixtures became $(U,Zr)O_2$ after the heat treatment at 1400 °C for 1 hour in 10% H_2 and 90% Ar condition. In addition, after the heat treatment of U_3O_8-20mol%Nd_2O_3, U_3O_8-50mol%Nd_2O_3, U_3O_8-20mol%Eu_2O_3, and U_3O_8-50mol%Eu_2O_3 mixtures at 1000 °C for 4 hours in air condition, it was found that U_3O_8 reacted with Nd_2O_3 or Eu_2O_3 and became $(Nd,U)O_{2+x}$ or $(Eu,U)O_{2+x}$, respectively.

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