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Physical mechanism analysis of burnup actinide composition in light water reactor MOX fuel and its application to uncertainty evaluation

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ABSTRACT

In designing radioactive waste management and decommissioning facilities, understanding the physical mechanisms for burnup actinide composition is indispensable to satisfy requirements for its validity and reliability. Therefore, the uncertainty associated with physical quantities, such as nuclear data, needs to be quantitatively analyzed. The present paper illustrates an analysis methodology to investigate the physical mechanisms of burnup actinide composition with nuclear-data sensitivity based on the generalized depletion perturbation theory. The target in this paper is the MOX fuel of the light water reactor. We start with the discussion of the basic physical mechanisms for burnup actinide compositions using the reaction-rate flow chart on the burnup chain. After that, the physical mechanisms of the productions of Cm-244 and Pu-238 are analyzed in detail with burnup sensitivity calculation. Conclusively, we can identify the source of actinide productions and evaluate the indirect influence of the nuclear reactions if the physical mechanisms of burnup actinide composition are analyzed using the reaction-rate flow chart on the burnup chain and burnup sensitivity calculation. Finally, we demonstrate the usefulness of the burnup sensitivity coefficients in an application to determine the priority of accuracy improvement in nuclear data in combination with the covariance of the nuclear data. In addition, the target actinides and reactions are categorized into patterns according to a sensitivity trend.

1. Introduction

The present design of facilities for radioactive waste management and decommissioning demands quality assurance, verification and validation, accountability, transparency, traceability, and reproducibility. To satisfy these requirements, the physical mechanisms in the burnup composition changes of actinides need to be analyzed in more detail. To guarantee the required design accuracy, quantitative analysis of the uncertainty arising from physical quantities, such as nuclear data, is most important. In this situation, burnup sensitivity coefficients that are calculated based on the generalized depletion perturbation theory (Williams, 1979; Takeda and Umano, 1985) are available for the uncertainty breakdown.

In the present study, we aim to illustrate a methodology to analyze the physical mechanisms of burnup actinide composition with sensitivity calculation based on the generalized depletion perturbation theory. As an application of the methodology, we clarify the physical mechanisms of burnup actinide composition change in MOX spent fuels of conventional light water reactors (LWRs) in this paper. First, the physical mechanisms for burnup actinide composition are discussed using the reaction-rate flow chart on the burnup chain. Next, we particularly analyze the complicated physical mechanisms for the actinide composition changes in LWRs using a combination of a reaction-rate flow chart and the burnup sensitivity coefficients with respect to capture and fission cross sections and the half-lives of the actinides. Here we focus on the production mechanisms of ^{244}Cm and ^{238}Pu, which are important actinides in the nuclear fuel cycle field. Moreover, we describe a method to determine the priorities for the uncertainty reduction of specific nuclear data as the application of the burnup sensitivity coefficients. In addition, we categorize the actinides into patterns according to a sensitivity trend.

Section 2 describes the conditions of the numerical analysis of actinide burnup. Section 3 shows the reaction-rate flows for actinide production. In Section 4, the burnup sensitivity coefficients are described. Section 5 discusses the results of the analysis of burnup sensitivity and the physical mechanism. Section 6 shows the uncertainty evaluation to improve the accuracy of nuclear data using the burnup sensitivity coefficients and nuclear data covariance. Finally, Section 7 presents conclusions to the present study.

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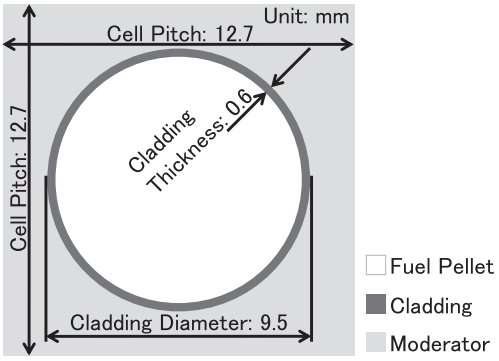

Fig. 1. Cell model.

Table 1
Calculation condition.

Parameter	Description
^{235}U enrichment (wt%)	0.23
Pu total enrichment (wt%)	6.3
Pu fissile fraction (wt%)	68
Linear heat rate (W/cm)	176 (constant)
Burnup (GWd/t)	50.0
Cladding material	Zircaloy
Moderator material	H_2O
Fuel temperature (K)	973.15
Cladding temperature (K)	603.15
Moderator temperature (K)	573.15

Appendix A shows the overall classification list of burnup sensitivity trends for LWR MOX fuel.

2. Conditions of the numerical analysis of actinide burnup

The fuel treated here is MOX of a 17 × 17 pressurized water reactor (PWR) fuel assembly (Ando and Takano, 1999). The cell model is shown in Fig. 1 where the white boundary condition is assigned.

Table 1 shows ^{235}U enrichment, Pu total enrichment, Pu fissile fraction of fresh fuel, the linear heat rate of the fuel pin and its final burnup value, materials of cladding and moderator, and temperatures in each region.

The burnup chain includes actinides from ^{234}U through ^{246}Cm, as shown in Fig. 2.

Neutron transport calculation and burnup chain analysis are based on the collision probability method and the matrix exponential method, respectively. The multi-purpose reactor analysis code system, MARBLE, (Yokoyama et al., 2008) and a 107 energy-group constant set based on the latest version of the Japanese Evaluated Nuclear Data Library (JENDL-4.0) (Shibata et al., 2011) are utilized here. The eigenvalues are calculated at 25 depletion steps each of which is constructed of 22 sub-steps where each sub-step is divided into 10 sub-sub-steps near the end of life (EOL) since the adjoint flux changes intensively near EOL.

3. Result of calculations for actinide production

Table 2 shows important physics data of nuclides related to the MOX burnup-chain analysis, i.e., half-life, number density at the beginning of life (BOL) and at the end of life (EOL), and one-group cross sections of capture and fission reactions at EOL.

Fig. 3 summarizes the macroscopic reaction-rate flows at EOL.

The macroscopic capture and fission reaction rates, R_C and R_F, are calculated using the number density, one-group capture and fission cross sections, and neutron flux. The decay rate, R_D, is defined as the product of number density and decay constant. The capture reaction rates of ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, and ^{244}Cm and the decay rates of ^{239}Np, ^{241}Pu, ^{242g}Am, and ^{242}Cm, which are shown by solid arrows, are larger than the others shown by dotted arrows. We will scrutinize the mechanisms of ^{244}Cm and ^{238}Pu production with Fig. 3, since they are important in the nuclear fuel cycle field such as reprocessing and high-level radioactive waste disposal.

First, we focus on the ^{244}Cm production mechanism. As shown in Fig. 3, ^{244}Cm is possible to be produced from the capture reaction of ^{243}Am and ^{243}Cm. Since the number density and the capture cross section of ^{243}Am are larger than those of ^{243}Cm as shown in Table 2, ^{243}Am has larger R_C than ^{243}Cm. Thus, the main production path of ^{244}Cm is from the capture reaction of Pu isotopes via that of ^{243}Am. However, it is difficult to judge which Pu isotope is most important for the ^{244}Cm production only from Fig. 3 and Table 2, therefore, we need the sensitivity analysis described in Section 5.

Second, the production of ^{238}Pu is considered. As shown in Fig. 3, ^{238}Pu is possible to be produced via the capture reaction of ^{237}Np and the decay of ^{242}Cm. Since the R_D of ^{242}Cm is larger than R_C of ^{237}Np, the main production path of ^{238}Pu is from the capture reaction of ^{241}Am via the decay of ^{242g}Am and ^{242}Cm if we can consider the mechanism of ^{238}Pu production similar to that of ^{244}Cm. However, ^{241}Am, which is the nearest source of ^{238}Pu production, is a little far from ^{238}Pu on the burnup chain. In addition, it is possible that a lot of ^{238}Pu at BOL does not transmute into the other nuclides and remains itself during the burnup, since plenty of ^{238}Pu is included in the MOX fuel at BOL and R_C, R_F, and R_D of ^{238}Pu are relatively-small. Thus, we are not able to conclude whether the main source of ^{238}Pu amount at EOL is ^{241}Am or ^{238}Pu itself only from Fig. 3 and Table 2.

As seen, we roughly understand the basic physical mechanisms for burnup actinide compositions using the reaction-rate flow chart and the physical data related to the burnup chain in Fig. 3 and Table 2. However, to evaluate and improve the accuracy of

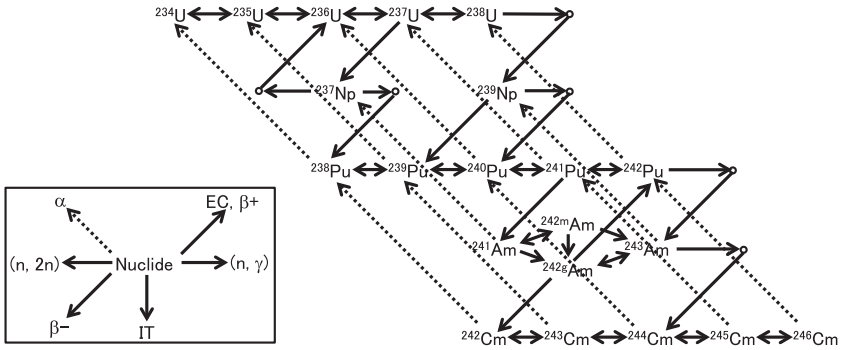

Fig. 2. Burnup chain for actinides.