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Evaluation of the specific radioactivity of 40 elements created by nuclear transmutation of fission products

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ABSTRACT

A burnup calculation was performed to analyze Après ORIENT process which aims at creating highly-valuable elements by nuclear transmutation of fission products (FPs) separated from LWR spent nuclear fuels. In this paper, numerical evaluation of the specific radioactivity of 40 created elements, from reloaded each FP element, with atomic number from 31 to 70 at the end of the time of 5-year-cooling after the irradiation for 1125 days in each LWR and FBR was carried out. These 40 created elements were classified in 6 categories according to levels of the specific radioactivity and the length of additional cooling period, which was needed for the specific radioactivity to decrease below the exemption level defined by International Atomic Energy Agency. As a result, created $_{31}$Ga, $_{32}$Ge, $_{33}$As, $_{35}$Br, $_{68}$Er, and $_{70}$Yb did not contain any radioisotopes at the end of the 5-year-cooling. It should be noted that created $_{37}$Rb, $_{57}$La, and $_{60}$Nd had much lower specific radioactivities than natural composition of them. Moreover, specific radioactivities of created $_{40}$Zr, $_{42}$Mo, $_{44}$Ru, $_{46}$Pd, $_{49}$In, and $_{54}$Xe were sufficiently lower than their exemption levels at the end of the 5-year-cooling. On the other hand, created $_{39}$Y, $_{45}$Rh, $_{50}$Sn, $_{52}$Te, $_{58}$Ce, $_{59}$Pr, $_{65}$Tb, and $_{66}$Dy needed additional cooling period less than 10 years until their specific radioactivities decreased below their exemption levels. Then, each additional cooling period required for created $_{48}$Cd, $_{51}$Sb, $_{64}$Gd, and $_{69}$Tm was estimated at 10–100 years. Additionally, specific radioactivities of other 13 created elements would not decrease below their exemption levels even if they had been stored for 100 years. There could be significance to create important elements as resources classified in first 4 of the 6 categories defined in this paper, by nuclear transmutation of fission products. In consideration of the efficiency of creation, the radioactivity, and the importance as resources of each product, $_{44}$Ru, $_{46}$Pd, $_{52}$Te, $_{60}$Nd, and $_{66}$Dy were specially selected as the most important created elements to be more researched in the future Après ORIENT program.

1. Introduction

Recently, demand for many rare elements is rapidly increased especially in the field of clean energy technologies. For example, Pd belonging to platinum group metals (PGMs) is included in a catalyst for purifying exhaust gas of vehicles. Then, world demand of Pd is steadily increasing (Thomson Reuters, 2015). In addition, Nd and Dy classified as rare earth (RE) elements are used as a part of a magnetic material in a high-performance motor for electric automobiles. The availability of a number of these materials is at risk due to their location, vulnerability to supply disruptions and lack of suitable substitutes (USDOE, 2010). Actually, 76% of the mine production of Pd was supplied from only South Africa and Russia in 2014 (Thomson Reuters, 2015). Moreover, 86% of the mine production of rare earth elements was supplied from only China (USGS, 2015). Thus, a new method for stable supply of these rare elements is strongly needed.

In Japan, Advanced ORIENT cycle (Ozawa et al., 2008) and then Après ORIENT program (Han et al., 2012; Ozawa et al., 2013; Terashima and Ozawa, 2015) are proposed, which are concepts of advanced nuclear fuel cycle. It is well known that fission products

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(FPs) in spent nuclear fuels contain PGMs, RE, and other useful elements, which are defined nuclear rare metals (NRMs) in these concepts. Then, Advanced ORIENT cycle and Après ORIENT program are aiming at effective utilization of NRMs by combination of partitioning and transmutation processes. Especially, the Après ORIENT program aims at creating stable, highly-valuable elements (i.e. secondary NRMs) from FPs by nuclear transmutation in nuclear reactors, following such a reaction;

$$ {}^{A}_{Z}\mathrm{FP}(n,\gamma)^{A+1}_{Z}\mathrm{FP}\xrightarrow{\beta^-}{}^{A+1}_{Z+1}\mathrm{NRM(stable)} \quad (1) $$

The previous investigation had been carried out as a fundamental study to embody the Après ORIENT program to clarify characteristics of creation of elements by nuclear transmutation of FPs (Terashima and Ozawa, 2015). As a result, creation rates of each element with atomic number from 31 to 70 from FPs had been obtained. However, even though the creation rate of a $_{Z+1}$NRM element is high, the $_{Z+1}$NRM cannot be considered as resources if it have high radioactivity. Thus, in the present research, the specific radioactivity of $_{Z+1}$NRM created from $_{Z}$FP by nuclear transmutation in reactors was evaluated.

2. Calculation method

A burnup calculation was performed to analyze the specific radioactivity of each $_{Z+1}$NRM element created from $_{Z}$FP element by nuclear transmutation. In the same way to the previous study (Terashima and Ozawa, 2015), nuclear reactors were assumed as sources of intense neutron flux, and neutron capture reactions were considered as a method of creation of NRMs by nuclear transmutation. It is assumed to irradiate a $_{Z}$FP element target by neutrons in each LWR such as PWR and FBR such as MONJU (a sodium cooled prototype fast breeder reactor in Japan).

The burnup calculation was carried out with the ORLIBJ40 package (Okumura et al., 2013), which is a combination of burnup calculation code based on ORIGEN2 (Croff, 1983) and 1-group cross section library based on JENDL-4.0 (Shibata et al., 2011). It should be noted that, in the ORLIBJ40 package, the range of nuclear mass number A of $_{Z}$FP are defined from 66 to 172. Then, elements with at least one stable isotope in the range are from $_{30}$Zn to $_{70}$Yb, except for $_{43}$Tc and $_{61}$Pm which have only radioactive isotopes. Therefore, in the previous and present studies, 40 elements from $_{30}$Zn ($_{31}$Ga) to $_{69}$Tm ($_{70}$Yb) were defined as $_{Z}$FP ($_{Z+1}$NRM).

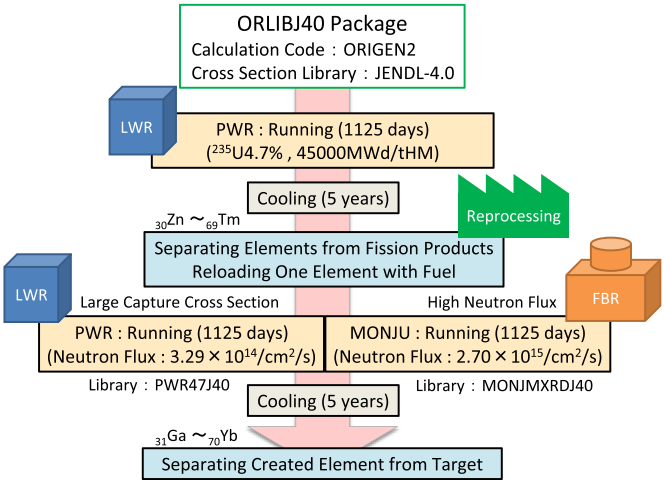

Fig. 1. Flowchart of computation for the creation of secondary NRMs in Après ORIENT scheme, which is reproduced from the previous paper (Terashima and Ozawa, 2015).

A flowchart of computation adopted in this study is shown in Fig. 1, which was reproduced from the previous paper (Terashima and Ozawa, 2015). Thus, the basic procedure of calculation in the present study is same to one in the previous paper (Terashima and Ozawa, 2015). After the procedure of computational calculation, evaluation of the specific radioactivity of each created element and estimation of "additional cooling period" were carried out. In this paper, "additional cooling period" was defined as a time required for the specific radioactivity to decrease below the exemption level defined by International Atomic Energy Agency (IAEA, 2014). A created element with lower specific radioactivity than the exemption level can be considered as not radioactive material but available resources.

Referring to the calculation method of the previous study (Terashima and Ozawa, 2015), the computation scheme consisted of the following 7 steps;

STEP 1: A burnup calculation was performed in the condition that a PWR (1100 MW type) was operated for 1125 days to reach 45,000 MWd/tHM. The fuel was assumed to be fresh with an enrichment of ^{235}U by 4.7%. A 1-group cross section library PWR47J40.LIB was used in this calculation (cf. Appendix A).
STEP 2: A decay calculation of the spent nuclear fuels during a cooling period of 5 years was carried out.
STEP 3: Amount and isotopic composition of each element were analyzed as the initial condition of a $_{Z}$FP element target for neutron irradiation in the next steps.
STEP 4: Input data of each target $_{Z}$FP element was prepared for a burnup calculation of a $_{Z}$FP assumed to have been obtained by perfectly mutual element separation. Production amount and specific radioactivity of each $_{Z}$FP element after such separation are shown in Fig. 2. Then, the $_{Z}$FP element was also assumed to have been reloaded in each a core region of a PWR and in a radial blanket region of MONJU at the same time of the mutual element separation. Thus, amount and isotopic composition of a reloaded $_{Z}$FP were same as ones determined in STEP 3. It should be noted that, in the case of MONJU, the $_{Z}$FP target was supposed to be placed in the position of a blanket region adjacent an outer core region in order to irradiate it by the neutron flux as high as possible within the limits of the region.
STEP 5: A burnup calculation was performed corresponding to operation of each reactor for 1125 days in each condition shown in Table 1 (cf. Appendix A). In order to suppose such position of $_{Z}$FP target in MONJU, comparatively high neutron flux (namely burnup in the input data) was given in this calculation. Thus, although it was higher than the average neutron flux in the radial blanket region of MONJU, it would be lower than the maximum neutron flux in the region ($\sim 3.0 \times 10^{15}$/cm^2/s).
STEP 6: A decay calculation was carried out corresponding to a cooling period of 5 years.
STEP 7: Analysis of the amount, isotopic composition, and specific radioactivity of the $_{Z+1}$NRM element created from the $_{Z}$FP element was performed.

In this study, the same cross section was used with and without $_{Z}$FP target. Thus, the self-shielding effect of reloaded $_{Z}$FP had not been taken into account. Then, the actual effective cross section could depend on the position of target in the reactor, on the number density of target, and on other parameters which had not been able to be considered in this simple calculation for as many as 40 F P elements. Therefore, much strictly simulation for some specific elements by using 3-dimensional Monte Carlo code will be done in the future works based on the present paper.