NUCLEAR SCIENCE AND TECHNIQUES **26**, S10311 (2015)

Numerical analysis on element creation by nuclear transmutation of fission products

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(Received May 8, 2014; accepted in revised form July 19, 2014; published online December 20, 2014)

A burnup calculation was performed to analyze the Après ORIENT process, which aims to create highly-valuable elements from fission products separated from spent nuclear fuels. The basic idea is to use nuclear transmutation induced by a neutron capture reaction followed by a β^- decay, thus changing the atomic number Z of a target element in fission products by 1 unit. LWR (PWR) and FBR (MONJU) were considered as the transmutation devices. High rates of creation were obtained in some cases of platinum group metals ($_{44}$Ru by FBR, $_{46}$Pd by LWR) and rare earth ($_{64}$Gd by LWR, $_{66}$Dy by FBR). Therefore, systems based on LWR and FBR have their own advantages depending on target elements. Furthermore, it was found that creation rates of even Z' $(= Z + 1)$ elements from odd Z ones were higher than the opposite cases. This creation rate of an element was interpreted in terms of "average 1-group neutron capture cross section of the corresponding target element $\langle\sigma_{cZ}\rangle$" defined in this work. General trends of the creation rate of an even (odd) Z' element from the corresponding odd (even) Z one were found to be proportional to the 0.78th (0.63th) power of $\langle\sigma_{cZ}\rangle$, however with noticeable dispersion. The difference in the powers in the above analysis was explained by the difference in the number of stable isotopes caused by the even-odd effect of Z.

Keywords: Nuclear transmutation, Fission products, Element creation, Neutron capture cross section, Après ORIENT

DOI: 10.13538/j.1001-8042/nst.26.S10311

I. INTRODUCTION

Recently, demand for Pd, Nd, Dy, and other rare metals is increased because they are used for many rapidly-progressing technologies such as clean energy devices. As a result, in Japan, which relies on the import of them from other countries, supply risks of them have increased. Thus, a stable supply of these rare elements is strongly desired.

It is known that platinum group metals (PGMs), rare earth (RE), and other useful elements are contained in spent nuclear fuels (SNF) of nuclear reactors as fission products (FPs). They are defined nuclear rare metals (NRMs) in the concept of the Advanced ORIENT cycle [1], shown in the upper part of Fig. 1, in which these materials are actively retrieved from SNF by chemical partitioning to use as precious resources. Most of these elements, however, have very high or long-life radioactivity which prevents them to be considered as resources.

The nuclear transmutation process has a possibility of decreasing the amount of radioactive nuclides, as well as to change an element to another one, eventually leading to a stable isotope by such a process:

$$^{99}_{43}\mathrm{Tc}(\mathrm{n},\gamma)\,^{100}_{43}\mathrm{Tc}\xrightarrow{\beta^-}\,^{100}_{44}\mathrm{Ru(stable)}. \qquad (1)$$

The Après ORIENT research program [2] shown in the lower part of Fig. 1 was initiated in FY2011 aiming at creating stable, highly-valuable elements (i.e. secondary NRMs) by nuclear transmutation from FPs contained in SNF of nuclear reactors. The present investigation was carried out as a fundamental study to embody the Après ORIENT research program to clarify characteristics of creation of elements by nuclear transmutation.

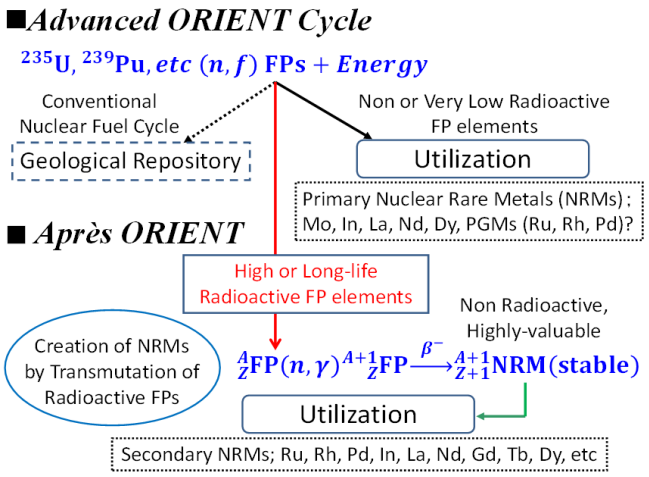

Fig. 1. (Color online) Scheme of Advanced ORIENT cycle concept and the Après ORIENT research program.

II. CALCULATION METHOD

A burnup calculation was performed to analyze the nuclear transmutation rate from FPs. In this study, nuclear reactors were assumed as sources of intense neutron flux and neutron capture reactions by reactor neutrons were considered as a method of creation of NRMs by nuclear transmutation. After a nucleus captures a neutron, it may eventually undergo

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β^- decay and, therefore, is transformed into another element having one proton more than before. Hence, stable highly-valuable rare metal elements (i.e. secondary NRMs) will be created by (n, γ) reaction, succeeded by β^- decay of FPs which are generally characterized by high radioactivity. Such a process can be written as

$$ {}^{A}_{Z}\mathrm{FP}(\mathrm{n},\gamma)\,{}^{A+1}_{Z}\mathrm{FP} \xrightarrow{\beta^-} {}^{A+1}_{Z+1}\mathrm{NRM(stable)}. \qquad (2) $$

The burnup calculation was carried out with the ORLIBJ40 package [4], which is a combination of ORIGEN2.2 code [5] as burnup calculation code and 1-group cross section library based on JENDL-4.0 [6]. In ORLIBJ40, the irradiation geometry is assumed to be homogeneous, which may not be realistic. This point will be improved in our future work by employing 3-dimensional Monte-Carlo simulators.

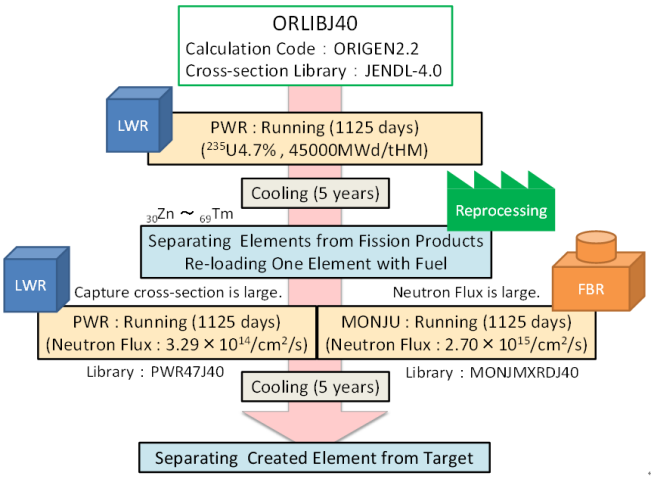

Fig. 2. (Color online) Flowchart of computation for the creation of secondary NRMs in Après ORIENT scheme.

The flowchart of computation adopted in this study is shown in Fig. 2. It is assumed to irradiate a target element by neutrons in LWR such as PWR, FBR such as MONJU (a sodium cooled prototype fast breeder reactor in Japan). The computation scheme consists of the following 7 steps:

STEP 1: A burnup calculation is performed with the condition that a PWR is operated for 1125 days to reach 45 000 MWd/tHM. The fuel is assumed to be fresh with an enrichment of ^{235}U by 4.7%. A 1-group cross section library PWR47J40.LIB [7] is used in this calculation.

STEP 2: A decay calculation of the spent nuclear fuels during a cooling period of 5 years is carried out. Yields of elements contained in FPs after the cooling are shown in Fig. 3.

STEP 3: Amount and isotopic composition of each element are analyzed as an initial condition of the target $_{Z}$FP element for neutron irradiation in the following steps.

TABLE 1. Isotope composition of the nuclear fuel

	Mass number	LWR (PWR) (%)	FBR (MONJU) (%)
U	235	4.7	0.24
	238	95.3	80.76
Pu	238	–	0.57
	239	–	10.07
	240	–	4.75
	241	–	2.28
	242	–	1.33

TABLE 2. Isotope composition of reloaded Ru and Rh as examples of reloaded a $_{Z}$FP (*metastable)

$_{44}$Ru			$_{45}$Rh		
Mass number	Weight (g)	Half-life	Mass number	Weight (g)	Half-life
98	1.20E-07	Stable	101	3.54E-10	3.3 years
99	2.35E-02	Stable	102	3.22E-06	207 days
100	1.42E+02	Stable	*102m	5.51E-08	3.742 years
101	1.05E+03	Stable	103	6.13E+02	Stable
102	1.07E+03	Stable	106	6.17E-06	30.07 sec
103	5.41E-13	39.26 days			
104	7.21E+02	Stable			
106	6.59E+00	371.8 days			

STEP 4: Input data of each target $_{Z}$FP element is prepared for a burnup calculation of a $_{Z}$FP assumed to have been obtained by perfect mutual element separation (amount and isotopic composition of which is determined in STEP 3). Then, the $_{Z}$FP is also assumed to have been reloaded in a core region of a PWR or a blanket region of MONJU at the time of the mutual element separation. Isotopic composition of the nuclear fuel of each reactor core is shown in Table 1. Isotopic composition of reloaded Ru and Rh are shown in Table 2 as examples of reloaded a $_{Z}$FP.

STEP 5: A burnup calculation is performed corresponding to operation of each reactor for 1125 days with the following conditions:

PWR; neutron flux: 3.29×10^{14}/(cm^2 s), with a 1-group cross section library as PWR47J40.LIB.

MONJU; neutron flux: 2.70×10^{15}/(cm^2 s), with a 1-group cross section library as MONJMXRDJ40.LIB [7].

STEP 6: A decay calculation is carried out corresponding to a cooling period of 5 years.

STEP 7: Analysis of the amount and isotopic composition of the $_{Z+1}$NRM element created from the $_{Z}$FP is performed.

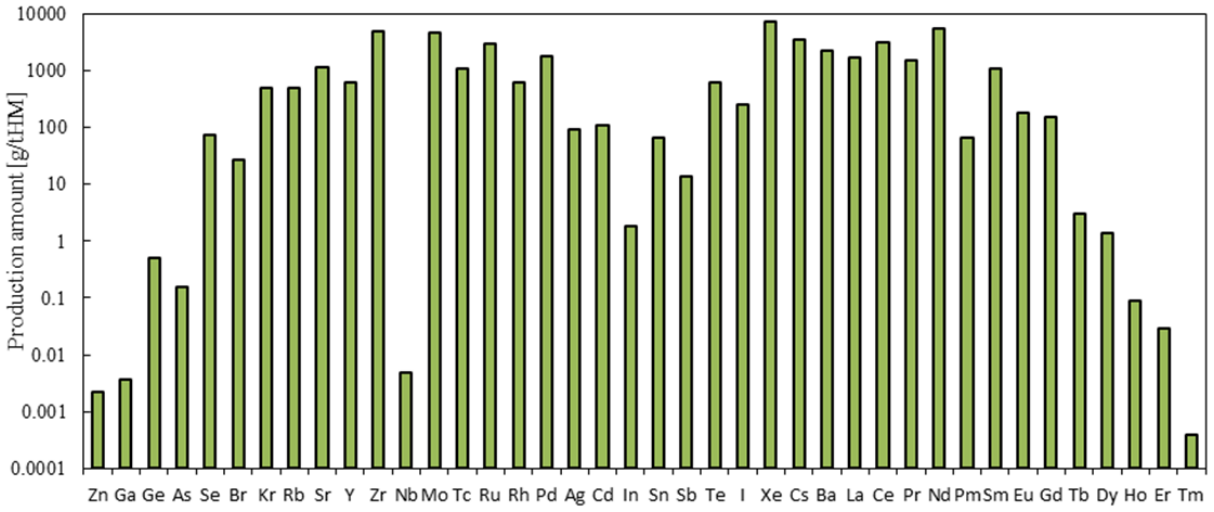

Fig. 3. (Color online) A bar chart showing calculated results on the amounts of FP elements in a PWR spent fuel from 1 t of fresh nuclear fuel (^{235}U: 4.7%) after an operation time of 1125 days (which leads to 45 000 MWd/tHM) followed by a cooling of 5 years.

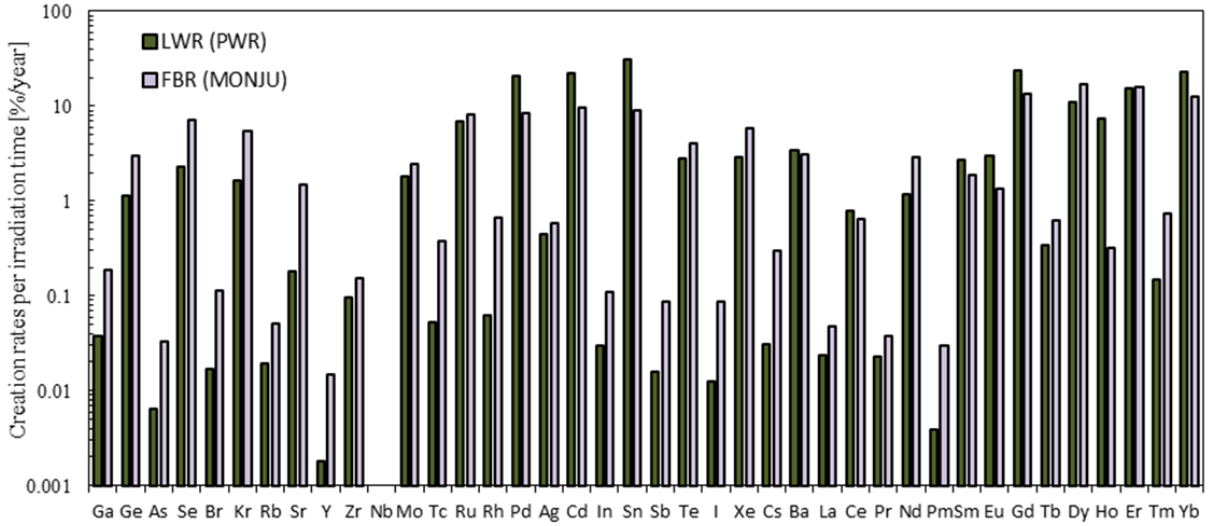

Fig. 4. (Color online) Comparison of creation rates of each $_{Z+1}$NRM element.

III. RESULTS AND DISCUSSION

A. Comparison of the "creation rate" of a $_{Z+1}$NRM element

In order to discuss transmutation efficiency quantitatively, "creation rate" of a $_{Z+1}$NRM element was defined as

$$\left(\begin{array}{c}\text{Creation rate}\\ \text{of}\ {}_{Z+1}\text{NRM}\end{array}\right)[\%/\text{year}] = \frac{\left(\begin{array}{c}\text{Amount of}\\ \text{created}\ {}_{Z+1}\text{NRM}\end{array}\right)[\text{g}]}{\left(\begin{array}{c}\text{Amount of}\\ \text{re-roaded}\ {}_{Z}\text{FP}\end{array}\right)[\text{g}]} \times \frac{100}{\left(\begin{array}{c}\text{Irradiation}\\ \text{time}\end{array}\right)[\text{year}]}. \quad (3)$$

Actually, this quantity indicates a yearly-average of the transmutation rate for an irradiation period of 1125 days (3.08 years). Then, comparison of creation rates of each $_{Z+1}$NRM element calculated by Eq. (3), as shown in Fig. 4. The creation rate of $_{45}$Rh represents the calculated result for $_{45}$Rh created from transmutation of reloaded $_{44}$Ru contained in FPs. It turned out that high creation rates appeared in valuable elements such as PGMs ($_{44}$Ru: 8.03%/year (FBR), $_{46}$Pd: 20.6%/year (LWR)) and middle or heavy RE elements (for example, $_{64}$Gd: 23.4%/year (LWR), $_{66}$Dy: 17.1%/year (FBR)). Overall, there is a tendency in many elements having small creation rates (less than 1%/year) such as $_{39}$Y, that the creation rate by FBR is higher than that by LWR, depending on the difference in the amount of neutron flux. On the contrary, some elements with high creation rates (more than 10%/year) such as $_{50}$Sn, show the opposite results regardless

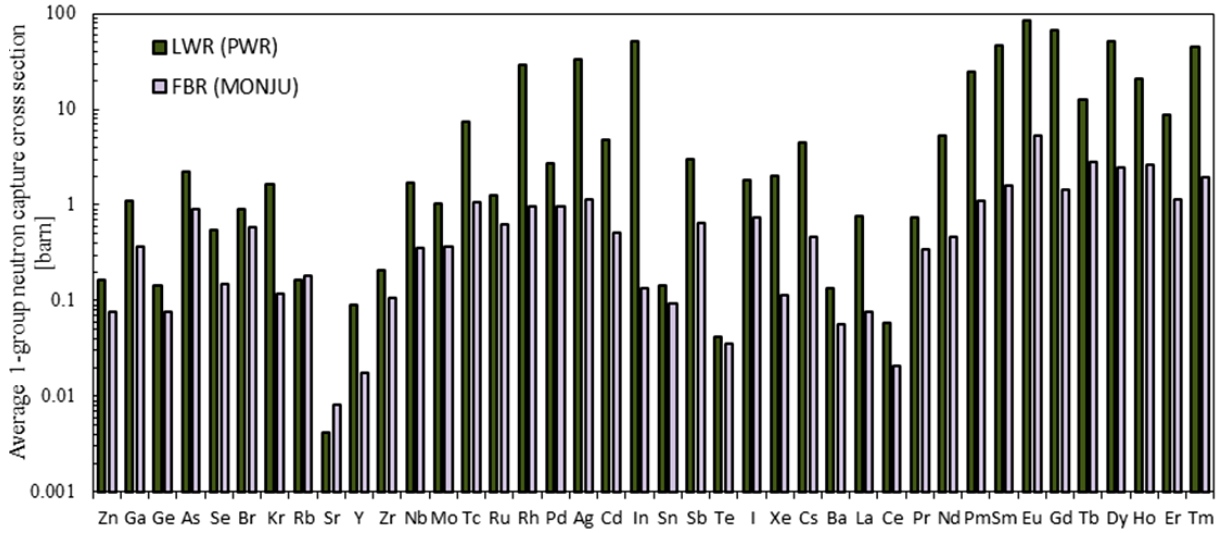

Fig. 5. (Color online) Comparison of average 1-group neutron capture cross sections of each target $_{Z}$FP element.

of the difference in the neutron flux. Therefore, it can be concluded that both transmutation systems have certain advantages and disadvantages and it should be carefully chosen which system to use to create a certain $_{Z+1}$NRM. Incidentally, if proper moderators were applied in the transmutation region of FBR, higher creation rates could be obtained for any $_{Z+1}$NRM element.

It is also clear in Fig. 4 that there is a difference in these creation rates according to even-oddness of atomic numbers Z' $(= Z + 1)$. In addition, it is shown that these elements with even Z' have higher creation rates than those with odd Z'. For example, the creation rate of $_{46}$Pd from $_{45}$Rh is higher than those of both sides, that is, $_{45}$Rh from $_{44}$Ru and $_{47}$Ag from $_{46}$Pd. It is generally known that nuclei with an even number of protons are more stable than those with an odd number of protons. Therefore, the difference in each creation rate of $_{Z+1}$NRM is considered to be based on the difference in the stability of the nucleus due to even-odd effects.

B. Introduction of "average 1-group neutron capture cross section"

The transmutation rate of a nuclide can be approximately represented to be directly proportional to the neutron capture cross section of the nuclide and neutron flux, such as

$$\left(\begin{array}{c}\text{transmutation rate}\\ \text{of a nuclide } Z_i\end{array}\right) \propto \left(\begin{array}{c}\text{capture cross section}\\ \text{of the nuclide } \sigma_{\mathrm{c}Z_i}\end{array}\right) \times (\text{neutron flux } \phi). \quad (4)$$

Naively, the creation rate of a $_{Z+1}$NRM element might be represented in the same way. However, the cross section of a $_{Z}$FP element does not exist because a cross section is a physical quantity associated with a nucleus but not an element. In the present study, the "average 1-group neutron capture cross section" of a $_{Z}$FP element was introduced, and was defined as

$$\langle \sigma_{\mathrm{c}Z} \rangle = \frac{\sum_i \sigma_{\mathrm{c}Z_i} m_{Z_i}}{\sum_i m_{Z_i}} \quad (5)$$

$\langle \sigma_{\mathrm{c}Z} \rangle$: average 1-group neutron capture cross section of element Z;

$\sigma_{\mathrm{c}Z_i}$: 1-group neutron capture cross section of isotope i of element Z;

m_{Z_i}: weight of isotope i of element Z in FP.

Figure 5 shows average 1-group neutron capture cross sections of each target $_{Z}$FP element. It turns out that the structure of these average 1-group neutron capture cross sections of each target $_{Z}$FP element (Fig. 5) has a clear correlation with that of those creation rates of each $_{Z+1}$NRM element (Fig. 4). In addition, it is noticed that there are two valleys minimizing cross sections (around Sr and Ce). This structure is considered to be formed by the presence of $N = 50$ and 82 "neutron magic numbers" in this mass region, as indicated in Table 3.

TABLE 3. Nuclides with neutron magic numbers in target $_{Z}$FP elements reloaded (*radioactive nuclide)

Neutron magic number	$N = 50$	$N = 82$
The nuclide which exists in target $_{Z}$FP elements reloaded	Kr 86	Xe 136
	Rb 87	Cs 137*
	Sr 88	Ba 138
	Y 89	La 139
	Zr 90	Ce 140
	Nb 91*	Pr 141
	Mo 92	Nd 142

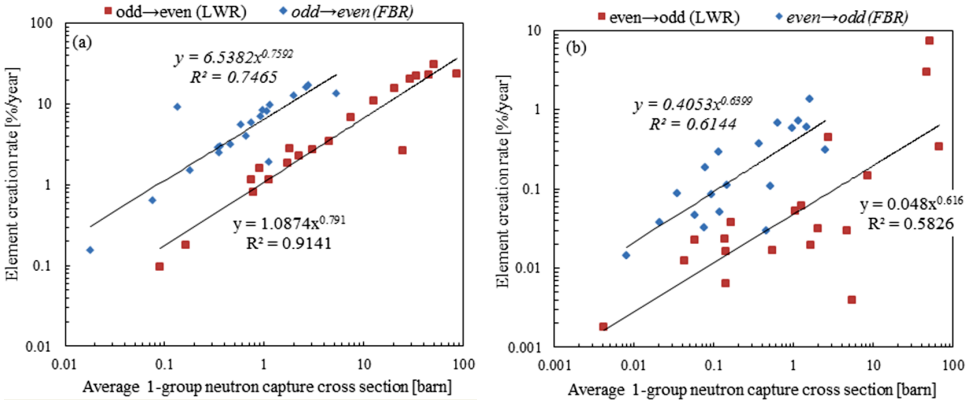

Fig. 6. (Color online) Correlation between creation rates of each $_{Z+1}$NRM element and average 1-group neutron capture cross sections of these corresponding target $_{Z}$FP elements. (a) exhibits results of transmutation from odd Z target FPs to even Z' NRMs. On the other hand, (b) exhibits from even Z target FPs to odd Z' NRMs.

C. Relation of creation rates of $_{Z+1}$NRM elements with average 1-group neutron capture cross sections of target $_{Z}$FP elements

Figure 6(a) shows creation rates of even Z' $(= Z + 1)$ NRMs as a function of average 1-group neutron capture cross sections of target FPs having an odd Z. Fig. 6(b) shows similar data for even Z target FPs leading to odd Z' NMRs. Because FBR (MONJU) had a larger neutron flux than LWR (PWR), creation rates by FBR were higher than those by LWR. In addition, the larger the average 1-group neutron capture cross section of each target $_{Z}$FP element was, the higher the creation rate of each $_{Z+1}$NRM element was. Thus, a clear correlation was established between creation rates and average 1-group neutron capture cross sections. In the case of the creation of NRMs with even Z', the creation rate of a $_{Z+1}$NRM element was approximately proportional to the 0.78th power of the average 1-group neutron capture cross section of the target $_{Z}$FP element, as

$$\left(\begin{array}{c}\text{creation rate of an element}\\ \text{with even atomic number}\end{array}\right) \propto \langle\sigma_{cZ}\rangle^{0.78}. \quad (6)$$

In addition, deviation from this trend is rather small. On the other hand, in the creation of NRMs with an odd Z', the creation rate of a $_{Z+1}$NRM element was roughly proportional to the 0.63th power of the average 1-group neutron capture cross section of the corresponding target $_{Z}$FP element, as

$$\left(\begin{array}{c}\text{creation rate of an element}\\ \text{with odd atomic number}\end{array}\right) \propto \langle\sigma_{cZ}\rangle^{0.63}. \quad (7)$$

However, this case is characterized with a large dispersion from the average trend. Reasons for the difference in the powers and dispersion will be discussed in the following section.

TABLE 4. The difference in the created $_{Z+1}$NRM element by the even-odd effect of atomic number

	Atomic number of created $_{Z+1}$NRM	
	Even [ex. Pd]	Odd [ex. Rh]
The number of stable isotopes of Target $_{Z}$FP	1 or 2 (It is easy to change elements.) [Rh:1]	Many (It is hard to change elements.) [Ru:7]
The number of stable isotopes of Created $_{Z+1}$NRM	Many (Created elements are stable.) [Pd:6]	1 or 2 (Created elements are unstable.) [Rh:1]
The dispersion from the average trend	Small	Large

D. Classification of the difference in the even-odd effect of atomic number

Table 4 summarizes the difference the even-odd effect has an atomic number Z' $(= Z + 1)$ from created NRMs. In the case of creation of NRMs with even atomic number Z' (ex. $_{46}$Pd), target FPs ($_{45}$Rh) have an odd atomic number Z, and they have only 1 or 2 stable isotopes ($_{45}$Rh: 1). That is, if an isotope ($_{45}$Rh) captures a neutron, it can decay easily, as in Fig. 7. Additionally, the $_{Z+1}$NRM element ($_{46}$Pd), created by β^- decay of an isotope of the target $_{Z}$FP element capturing a neutron, has so many stable isotopes ($_{46}$Pd: 6) that it hardly causes additional transmutation to another element (ex. $_{46}$Pd→ $_{47}$Ag). Fig. 8 shows the time variation of the weight of isotopes of target $_{45}$Rh in the case of irradiation by LWR. It turns out that because a majority of isotopes of target $_{45}$Rh are almost all ^{103}Rh, ^{103}Rh is mainly transmuted and decreased, as

$$^{103}_{45}\mathrm{Rh}(\mathrm{n},\gamma)^{104}_{45}\mathrm{Rh} \xrightarrow{\beta^-} {}^{104}_{46}\mathrm{Pd}(\text{stable}). \quad (8)$$

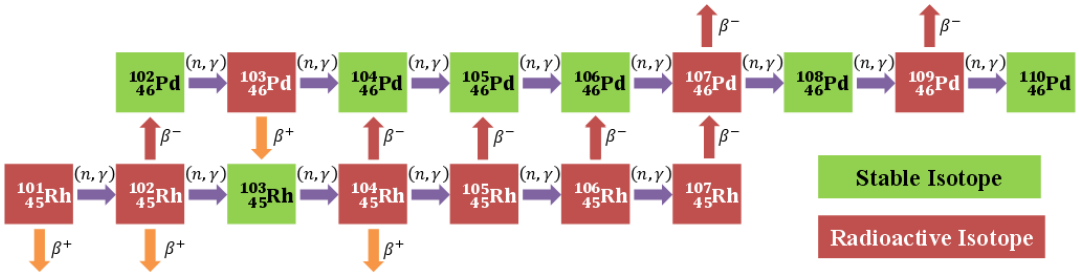

Fig. 7. (Color online) Main burnup chain from target $_{45}$Rh to created $_{46}$Pd.

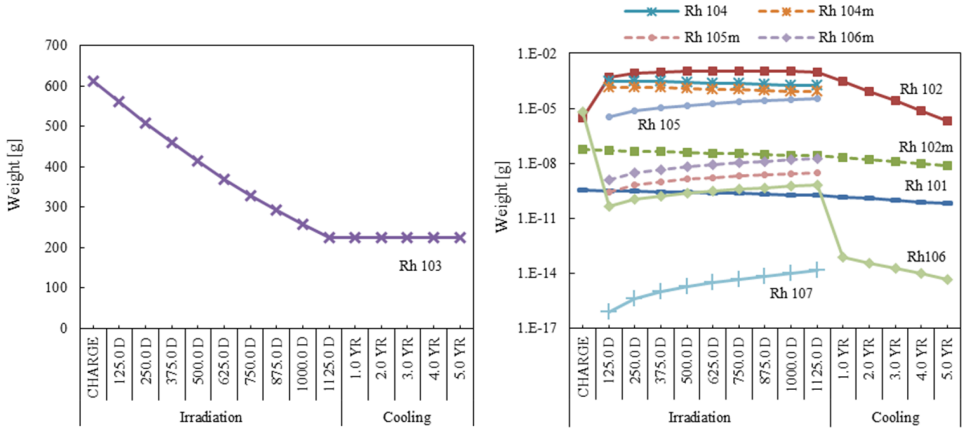

Fig. 8. (Color online) Time variation of the weight of isotopes of target $_{45}$Rh in the case of irradiation by LWR.

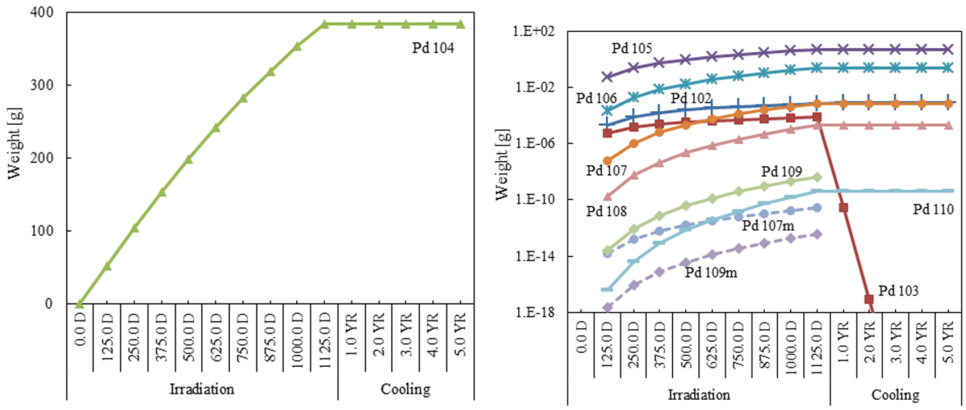

Fig. 9. (Color online) Time variation of the weight of isotopes of $_{46}$Pd created from target $_{45}$Rh in the case of irradiation by LWR.

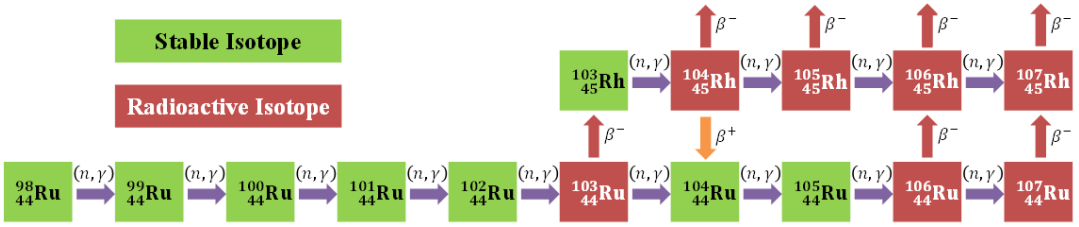

Fig. 10. (Color online) Main burnup chain from target $_{44}$Ru to created $_{45}$Rh.

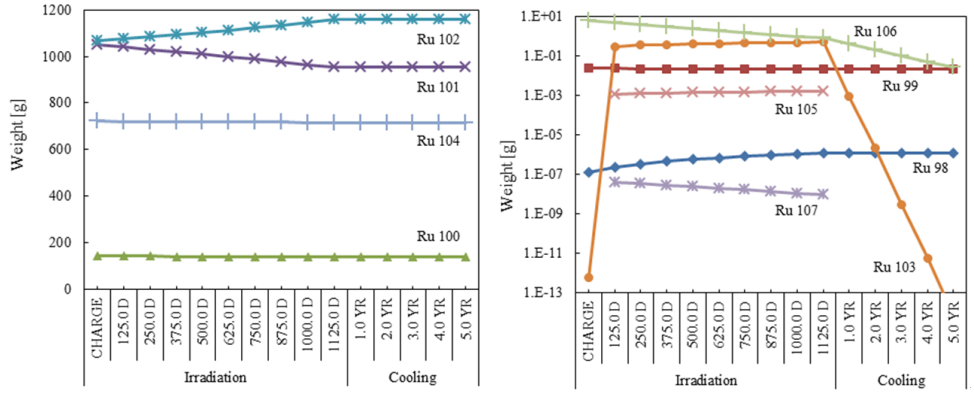

Fig. 11. (Color online) Time variation of the weight of isotopes of target $_{44}$Ru in the case of irradiation by LWR.

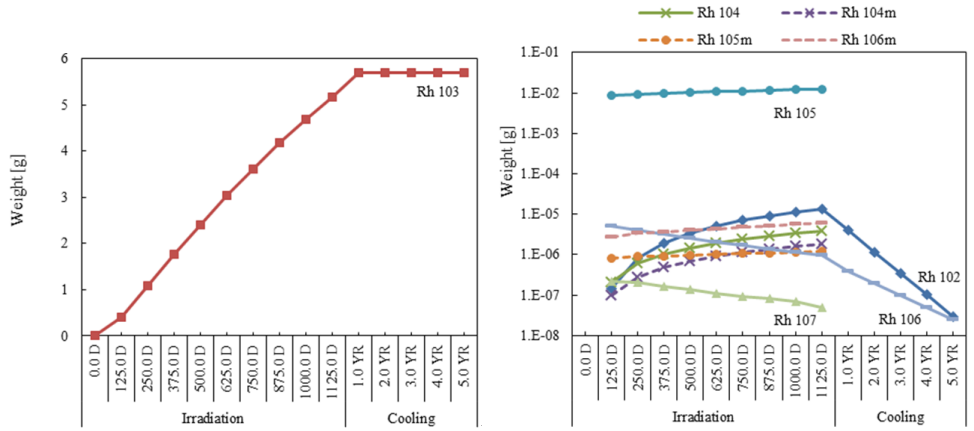

Fig. 12. (Color online) Time variation of the weight of isotopes of $_{45}$Rh created from target $_{44}$Ru in the case of irradiation by LWR.

In addition, Fig. 9 shows the time variation of the weight of isotopes of $_{46}$Pd created from target $_{45}$Rh in the case of irradiation by LWR. It turns out that many isotopes of $_{46}$Pd, which are heavier than ^{104}Pd (that is, the nuclide had captured more neutron after reaction as Eq. (8)), remain after cooling without decay because they are stable (Fig. 7).

On the other hand, in the case of the creation of NRMs with odd atomic number Z' (ex. $_{45}$Rh), target FPs ($_{44}$Ru) have an even atomic number Z and many stable isotopes ($_{44}$Ru: 7). That is, even if an isotope ($_{44}$Ru) captures a neutron, it cannot decay easily because it probably changes another stable isotope, such as in Fig. 10. Fig. 11 shows the time variation of the weight of isotopes of target $_{44}$Ru in the case of irradiation by LWR. It turns out that ^{102}Ru, which has a possibility of creation of ^{103}Rh, is the only stable isotope of $_{45}$Rh by (n, γ) reaction and β^- decay, as

$$^{102}_{44}\mathrm{Ru}(\mathrm{n},\gamma)^{103}_{44}\mathrm{Ru} \xrightarrow{\beta^-} {}^{103}_{45}\mathrm{Rh}(\mathrm{stable}), \qquad (9)$$

had not decreased, but increased. It suggests that not ^{102}Ru, but ^{101}Ru had mainly captured many neutrons and more nuclei of ^{102}Rh had been created by (n, γ) reaction of ^{101}Ru, then decreased them such as

$$^{101}_{44}\mathrm{Ru}(\mathrm{n},\gamma)^{102}_{44}\mathrm{Ru}(\mathrm{stable}). \qquad (10)$$

Thus, it turns out that the creation rate of $_{45}$Rh was not high (Fig. 4) because target $_{44}$Ru had been wasting many neutrons by other reactions not involved in the creation of $_{45}$Rh. In addition, Fig. 12 shows the time variation of the weight of isotopes of $_{45}$Rh created from target $_{44}$Ru in the case of irradiation by LWR. It turns out that all created isotopes of $_{45}$Rh except ^{103}Rh had decayed, although they had been created (Fig. 10). Therefore, many neutrons must be captured before they reach a β^--unstable isotope which finally undergoes β^- decay. In this case, the creation rate of a $_{Z+1}$NRM element is not directly affected by a single neutron capture process, but is affected by the number of neutron capture reactions to reach to the β^--unstable isotope of the target $_{Z}$FP element. Since the number of stable isotopes varies from element to element, the dispersion from the average trend is expected to be larger in the latter case.

IV. CONCLUSION

A burnup calculation has been performed in order to analyze the process which aims at creating highly-valuable stable elements (i.e. secondary nuclear rare metals; NRMs) from fission products (FPs) by using a neutron capture reaction,

followed by β^- decay. As a result, high rates of creation of $_{Z+1}$NRM elements were obtained for platinum group metals ($_{44}$Ru: 8.03%/year (FBR), $_{46}$Pd: 20.6%/year (LWR)) and middle or heavy rare earth elements (ex. $_{64}$Gd: 23.4%/year (LWR), $_{66}$Dy: 17.1%/year (FBR)). It was found that creation rates of NRMs with even atomic number Z' $(= Z + 1)$ were generally high, but those of NRMs with odd Z' were comparatively low. Having introduced average 1-group neutron capture cross sections of target $_{Z}$FP elements, correlation with creation rates of $_{Z+1}$NRM elements was analyzed and the following rules were established: the creation rate of a NRM element with even (odd) atomic number Z' was approximately proportional to the 0.78th (0.63th) power of the average 1-group neutron capture cross section of the corresponding target FP element with odd (even) atomic number Z. It was also found that the dispersion from the average trend was much larger in the case of creation of NRMs with odd Z' than in the case of creation of ones with even Z'. These differences could be attributed to the even-odd effect of average 1-group neutron capture cross sections and the number of stable isotopes. Based on the methodologies established and results obtained in this work, a design optimization of a reactor core and condition of neutron irradiation is going to be performed as a future work in order to increase the creation rate of NRMs.

ACKNOWLEDGEMENT

The authors would like to thank for the productive discussions with Prof. Satoshi Chiba, Tokyo Institute of Technology, Japan.

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