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One candidate method based on short-lived indicator for burnup analysis

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HIGHLIGHTS

- One burnup measurement method is proposed based on short-lived fission products.
- The key of this method is to make the nuclides reach an equilibrium concentration.
- New short-lived nuclides are screened out as indicators for burnup analysis.
- this method has a wider range of applications for burnup measurement.

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ABSTRACT

One novel gamma spectrometry method based on short-lived fission products was developed that can infer the level of burnup without complex computations or knowing irradiation history. The relationship between the burnup information and equilibrium concentration of short-lived indicators was derived. In addition, new candidate indicators for gamma spectrum analysis, such as ^{88}Kr, ^{91}Sr, ^{92}Sr, ^{135}I and ^{142}La were screened out. The results of three cases simulated with ORIGEN and MCNP were in good agreement with the theoretical values.

1. Introduction

For the purpose of improving safety and economy, burnup credit (International Atomic Energy Agency, 2001; Lebrun et al., 2001; Parks et al., 1999) has been applied in spent fuel management systems in recent decades. However, to apply burnup credit, it is important to first obtain the burnup information. Many burnup measurement methods have been proposed based on destructive assays or non-destructive assays (NDA) in the past few decades. Destructive analysis method (INOUE et al., 1969) is to dissolve the spent fuel and perform chemical analysis or mass spectral analysis. Though destructive analysis methods are intuitive, the measurement period is long, and operation is complicated, it is often used as an auxiliary method. NDA methods a well-established technique is to obtain the burnup information based on the delayed neutrons or fission product (FP) gamma-ray, which can be subdivided into neutron measurement methods (Akyurek and Usman, 2015; Jordan and Perret, 2011; Yokoyama et al., 1981) and gamma measurement methods (Bevard et al., 2009; Knowles et al., 2016).

Gamma spectrometry which enables to determine the activity of FPs is one well-known NDA technique for burnup analysis based on characteristic gamma intensity because of its high accuracy and simple operation. The FPs chosen for gamma spectrum analysis are called indicators. In the conventional nondestructive assay, long-lived FPs, such as ^{137}Cs, ^{134}Cs, ^{154}Eu, ^{106}Ru and ^{144}Ce (Akyurek et al., 2014; Caruso et al., 2008, 2007; Favalli et al., 2016; Harp et al., 2014; Iqbal and Mehmood, 2001; Navarro et al., 2015; Willman et al., 2006), are commonly used as indicators for gamma spectrum analysis. However, the characteristic gamma peaks intensity of these isotopes is relatively lower compared with some short-lived isotopes unless the spent fuel has been cooled for several months or years. In addition, the accuracy of this burnup analysis method, when using these isotopes as indicators, strongly depends on the fuel irradiation history, which is often unavailable or unreliable (Peir et al., 1999).

To solve the above difficulties, Wang et al. (Wang et al., 1990a, 1990b; Peir et al., 1999, Wang and Peir, 2000) have proposed a method that determines burnup through iterative calculations based on the activities of short-lived FPs, such as ^{97}Zr/^{97}Nb, ^{132}I and ^{140}La. An essential feature of their method is to activate the short-lived isotopes by re-irradiating the spent fuel for a few hours. Although their method is consistent with conventional methods which based on long-lived FPs, the spent fuel need to be cooled for adequate time before re-irradiation to ensure the short-lived FPs decay out, besides the data processing is

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complicated.

In this paper, we made some modifications based on the method of Wang et al. and proposed a new approach to determine burnup, via some new indicators, but without complex computations or the limitation of cooling time before re-irradiation. To evaluate the proposed method, one gamma spectrum measured in the zero-power light-water research LR-0 reactor (Košťál et al., 2017, 2016; Švadlenková et al., 2014) of VVER-type reactor cores, whose fuel assemblies enriched by ^{235}U range from 1.6 to 4.4 wt%, is selected as the contrast experiment. The numerical simulations were performed with ORIGEN and MCNP, and new indicators that conform to this method were analyzed.

2. Methods

The main feature of this method is to make some short-lived FPs establish an equilibrium concentration, rather than just activating them by reinserting the spent fuel into the reactor for re-irradiation compared with the previous method (Wang et al., 1990a, 1990b; Peir et al., 1999, Wang and Peir, 2000). To achieve this purpose, for the discharged spent fuel, it should be re-irradiated under stable neutron flux for few days. For the fuel to be unloaded, an additional constant neutron flux irradiation process is favorable before unloading. Furthermore, for the fuel in stable operating reactors such as commercial reactors, whose short-lived indicators have established an equilibrium concentration already, it can be carried out gamma measurement conveniently without an additional re-irradiation procedure after unloading. The corresponding theoretical analysis is shown below.

When the yield of nuclide i derived from β decay is much larger than the non-fission reaction yield such as (n, γ), (n, α) and (n, 2 n), and the half-life period of its parent nuclide is very short compared with nuclide i, nuclide i can be regarded as the cumulative FP from ^{235}U and other actinides such as ^{238}U, ^{239}Pu, ^{240}Pu, and ^{241}Pu. Because the reaction cross sections of ^{238}U (n, γ) ^{239}Pu, ^{239}Pu (n, γ) ^{240}Pu, ^{240}Pu (n, γ) ^{241}Pu are very small, and the fission rate of ^{238}U is negligible in the thermal reactor, the main source of nuclide i is from the fission of ^{235}U and ^{239}Pu (Wang et al., 2000). Then the content of nuclide i can be express by Eq. (1) (Duderstadt and Hamilton, 1976).

$$\frac{dN_i(t)}{dt} = \gamma'_i\sigma'_f\phi N_{U5}(t) + \gamma''_i\sigma''_f\phi N_{Pu9}(t) - (\lambda_i + \sigma_{\alpha,i}\phi)N_i(t) \quad (1)$$

Where γ'_i and γ''_i stand for the cumulative fission yields from ^{235}U and ^{239}Pu, respectively. σ'_f and σ''_f are the 1-group neutron fission cross section of nuclide ^{235}U and ^{239}Pu, $N_i(t)$, $N_{U5}(t)$ and $N_{Pu9}(t)$ are the atomic density of nuclide i, ^{235}U and ^{239}Pu, respectively. ϕ, λ_i and $\sigma_{\alpha,i}$ denote neutron flux, radioactive disintegration constant and the 1-group neutron absorption cross section of nuclide i, respectively.

When the spent fuel is irradiated under a relatively low neutron flux (such as 1.0×10^{13} cm^{-2} s^{-1}) for an additional few days to ensure the nuclide i establish an equilibrium concentration ($dN_i(t)/dt = 0$), it is reasonable to treat the residual ^{235}U content and ^{239}Pu content as constants during the re-irradiation. Then the equilibrium concentration $N_i(\infty)$ of short-lived nuclide i can be expressed by Eq. (2).

$$N_i(\infty) = \frac{\gamma'_i\sigma'_f\phi N_{U5}(t) + \gamma''_i\sigma''_f\phi N_{Pu9}(t)}{\lambda_i + \sigma_{\alpha,i}\phi} \quad (2)$$

Because the ^{239}Pu production content is related to the burnup level, while burnup level is expressed in terms of residual ^{235}U content. To obtain the relationship between the ^{235}U and ^{239}Pu, the fuel consumption process should be simulate according to the corresponding situation (Wang et al., 2000). Then the correlation between the ^{239}Pu production and ^{235}U depletion can be fitted based on the results of ORIGEN or other cell codes such as WIMS (Deen et al., 1995; Wang et al., 2000). Fig. 1. shows the correlation between the ^{239}Pu production and ^{235}U depletion in the UO_2 fuel with 20% enrichment, which irradiated under the neutron flux of 1.0×10^{14} cm^{-2}·s^{-1}, based on 4th order polynomial fitting.

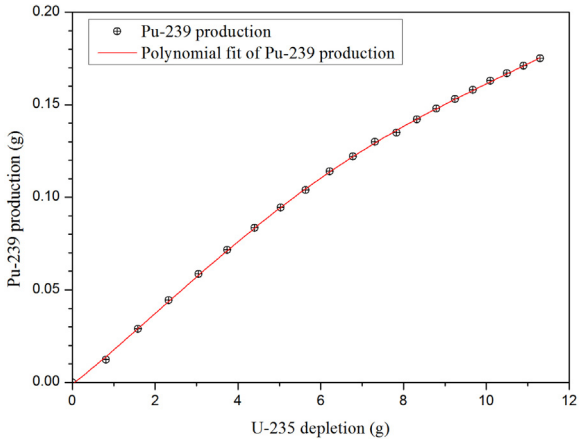

Fig. 1. The correlation between ^{239}Pu production and ^{235}U depletion in the fuel with 20% enrichment.

When an assumed $N_{U5}(t)$ value is given, a corresponding $N_{Pu9}(t)$ value can be obtained based on the fitted polynomial. Then the equilibrium concentration of indicator i is first calculated via Eq. (2) based on an assumed $N_{U5}(t)$ value and the corresponding $N_{Pu9}(t)$ value. The calculated indicator content $N_i(\infty)$ is then compared with the measured one, and this ratio is used to modify the $N_{U5}(t)$ value and the corresponding $N_{Pu9}(t)$ value for the next indicator content calculation. The iteration procedure continues until the calculated indicator content converges to the measured value. Finally, the residual ^{235}U content is obtained at the last iteration step.

The short-lived indicator content accumulated during constant neutron flux irradiation depends on the residual ^{235}U content and neutron flux. When the indicator content and the neutron flux value obtained by measurement, the residual ^{235}U content in spent fuel can be calculated. Based on the residual ^{235}U content and initial ^{235}U content, the burnup can be deduced by Eq. (3).

$$\alpha_F = \frac{N_{U5}(t_0) - N_{U5}(t_1)}{N_{U5}(t_0)} \times 100\% \quad (3)$$

Where α_F stands for the burnup level, $N_{U5}(t_0)$ denotes the initial atomic density of ^{235}U in the fresh fuel.

To ensure that the indicator reaches the equilibrium concentration and the consumption of ^{235}U is negligible during irradiation, it is necessary to determine some new indicators whose half-life periods are relatively short and can be easily discriminated from the gamma spectrum. The corresponding candidate isotopes are analyzed in the following content.

3. Candidate indicators analysis

Because the neutron absorption cross section $\sigma_{\alpha,i}$ of candidate indicator i is very small, it can be obtained that $\lambda_i + \sigma_{\alpha,i}\phi \approx \lambda_i$. Then the solution of partial differential Eq. (1) can be expressed as follow.

$$N_i(t) = \frac{\gamma'_i\sigma'_f\phi N_{U5}(t) + \gamma''_i\sigma''_f\phi N_{Pu9}(t)}{\lambda_i + \sigma_{\alpha,i}\phi}[1 - (\frac{1}{2})^{\frac{t}{T_{1/2}}}] \quad (4)$$

Where $T_{1/2}$ is the half life of nuclide i.

From the Eq. (4) it can be inferred that the spent fuel should be irradiated under stable and suitable neutron flux for about tenfold half-life period of the corresponding FPs to make the short-lived FPs reached equilibrium concentration. However, one requirement of this method is that the irradiation time should be relatively short to ensure that ^{235}U remains constant. To meet the above conditions, a ten-hour half-life period is set as the upper limit for indicators in this paper. In addition, considering the irradiation dose of the operators, a one-hour half-life period is treated as the lower limit in this paper. Furthermore, the