Paper

DECAY CHAIN DEDUCTION OF URANIUM FISSION PRODUCTS

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Abstract—**Delayed gamma spectrum is the fingerprint of uranium materials in arms control verification technology. The decay chain is simplified into basic state linear chain and excitation state linear chain to calculate and analyze the delayed gamma spectra of fission products. Formulas of the changing rule for nuclide number before and after zero-time are deduced. The C program for calculating the delayed gamma ray spectra data is constructed, and related experiments are conducted to verify this theory. Through analysis of the delayed gamma counts of several nuclides, the calculated results are found to be consistent with experimental values. Health Phys. 111(1):17–21; 2016**

Key words: decay chain; fission; spectroscopy, gamma; uranium

INTRODUCTION

In arms control verification, detection of the existence of enriched uranium in unknown materials and the amount of ^{235}U atoms in these materials (Slaughter 2007) is technically difficult to solve. For obtaining ^{235}U content in uranium materials, the delayed gamma spectra (Willman 2005; Liew 1991) of uranium fission products should be analyzed. Research results on delayed gamma spectra can be applied in numerous technical fields, such as verification of nuclear weapons, safe management of nuclear materials, monitoring of customs goods, nuclear reactors projects, and retirement of nuclear facilities (Gmar 1999; Slaughter 2005; Terremoto 1999).

As uranium fission products radiate prompt neutrons and gamma rays, the products will decay, undergo a series of beta decays because of the high neutron-proton ratio (Marrs 2007), and then become stable nuclides. Through the beta decay process, one nuclide changes into another in an excitation state. When this state jumps to the basic state, certain gamma rays, called delayed gamma (Wahab 1981), are radiated. Through β^- decay and electron capture (EC) decay, the atomic number of the nuclide increases or decreases, but the mass number remains the same (Pruet 2004). These nuclides with mass number equal to each other make up a decay chain. Fission products form into numerous decay chains with mass numbers from 66 to 107. Fig. 1a shows one simple decay chain, and Fig. 1b shows one complicated decay chain. In the figure, ^{115}Ag shows certain parental and complicated branching decays.

In the work, the slow neutron that does not render ^{238}U fissile is used to research delayed gamma rays. Therefore, several key gamma rays can be found to represent uranium fission products. If these key rays are detected in application, it is possible to judge whether ^{235}U nuclides exist in experimental samples. If the counts of key gamma rays achieve a certain value, the ^{235}U mass fraction of the uranium materials can be obtained.

DECAY CHAIN DEDUCTION OF URANIUM FISSION PRODUCTS

Physical model building

In the use of slow neutron irradiating materials, the ending time of irradiation is called zero-time (Hosni 2005). For obtaining the delayed gamma spectra, the change mechanism of a nuclide number before and after zero-time should be determined.

According to literature (Pruet 2004), one complex decay chain is divided into several linear chains, in which every nuclide is only related with one mother nuclide; these linear chains are devoid of branch chains. The total number of one nuclide is obtained by adding the branch number of every linear chain. For simplifying the physical model, the decay chain is classified into the basic-state chain and excitation-state chain. The former contains a mother nuclide at the basic state, whereas the latter possesses a mother nuclide at the excitation state.

In Fig. 1a, the basic-state chain and excitation-state chain are as follows:

basic-state chain: $^{90}\mathrm{Rb} \rightarrow^{\beta\cdot\ 90}\mathrm{Sr} \rightarrow^{\beta\cdot\ 90} Y \rightarrow^{\beta\cdot\ 90} Zr$
excitation-state chain: $^{90m}\mathrm{Y} \rightarrow^{IT\ 90} Y \rightarrow^{\beta\cdot\ 90} Zr$.

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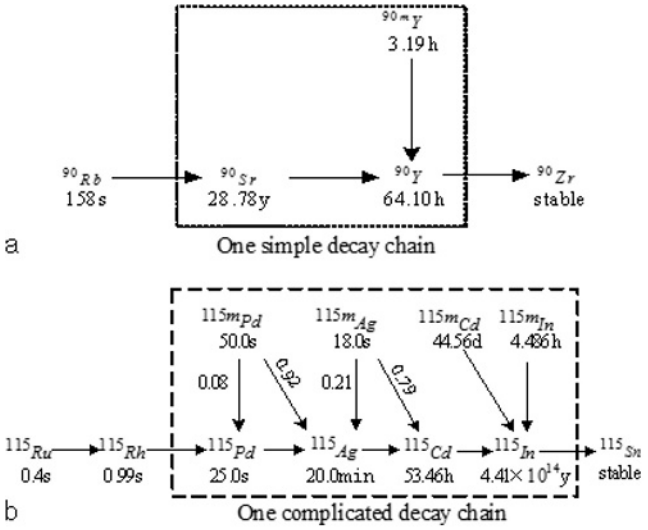

Fig. 1. Examples for two types of decay chains from uranium fission products. (a) Simple decay chain; (b) Complicated decay chain.

The mass fractions for ^{234}U, ^{235}U, ^{236}U, and ^{238}U in sample material are assumed to be p_1, p_2, p_3, and p_4, respectively, and the sample shape is disregarded. The original number of one uranium nuclide is N_{i0}:

$$N_{i0} = \frac{mp_i}{M} N_A \quad (i = 1, 2, 3, 4)\,. \tag{1}$$

where m is the sample mass, and M and N_A are the atomic weight of the target nucleus and constant, respectively. Now, take ^{235}U as an example. To find how the target nucleus number changes with time when $0 < t < t_0$, one assumes the following parameters: ϕ = flux density of neutron; σ = absorption cross section for target nucleus; σ_f = fission cross section for target nucleus; λ_a = alpha decay constant; λ_f = spontaneous fission constant; N_0 = original number of target nucleus; N = number of target nucleus at time t; λ_j = decay constant of fission product j in a decay chain; N_j = number of fission product j in a decay chain; Y_j = yield of fission product j in a decay chain; u = number of target nucleus that participates in nuclear reaction per second; and $u = \phi\sigma + \lambda_a + \lambda_f$. b_{jk} ($j > 0$, $0 \le k < j$)-parameter in formula before zero-time, which is related with decay constant, branch ratio, and yield of nuclide. c_{jk} ($j > 0$, $0 \le k < j$) = the parameter in the formula after zero-time, which is related to the nuclide number at zero-time.

By assuming that the incoming neutron possesses a single energy, the decay rate for the target nucleus is as follows:

$$\frac{dN}{dt} = -\left(\phi\sigma N + \lambda_a N + \lambda_f N\right) \quad N = N_0 e^{-ut}\,. \tag{2}$$

Change rule for the number of nuclides before zero-time

Number of nuclide on the basic-state chain. The nuclide number of the basic-state chain before zero-time is deduced. According to the above hypothesis, the rate of mutation for the mother nuclide is as follows:

$$\frac{dN_1}{dt} = \phi\sigma_f N_0 e^{-ut} Y_1 - \lambda_1 N_1 \qquad N_1 = \phi\sigma_f N_0 b_{10}\left(e^{-ut} - e^{-\lambda_1 t}\right). \tag{3}$$

where $b_{10} = \frac{Y_1}{\lambda_1 - u}$.

The number of the first progeny is contributed by ^{235}U fission, preceding level decay, and decay by itself:

$$N_2 = \phi\sigma_f N_0\left[b_{20}\left(e^{-ut} - e^{-\lambda_2 t}\right) + b_{21}\left(e^{-\lambda_2 t} - e^{-\lambda_1 t}\right)\right]. \tag{4}$$

where $b_{20}\,\frac{Y_2 + r_1\lambda_1 b_{10}}{\lambda_2 - u}$, $b_{21} = \frac{r_1\lambda_1}{\lambda_2 - \lambda_1} b_{10}$.

According to the mathematical method of induction, the number of nuclides at different levels is obtained as follows:

$$\begin{aligned} N_j = \phi\sigma_f N_0\big[&b_{j0}\left(e^{-ut} - e^{-\lambda_j t}\right) \\ &+ b_{j1}\left(e^{-\lambda_j t} - e^{-\lambda_1 t}\right) + \cdots \\ &+ b_{jk}\left(e^{-\lambda_j t} - e^{-\lambda_k t}\right) + \cdots \\ &+ b_{j,j-1}\left(e^{-\lambda_j t} - e^{-\lambda_{j-1} t}\right)\big] (j > 1) \end{aligned} \tag{5}$$

where $b_{j0} = \frac{Y_j}{\lambda_j - u} + \sum_{m=1}^{j-1} \frac{Y_m \left(\prod_{i=m}^{j-1} (r_i\lambda_i)\right)}{\prod_{i=m}^{j} (\lambda_i - u)}$, $b_{j1} = \frac{Y_1 \prod_{i=1}^{j-1} r_i\lambda_i}{(\lambda_1 - u)\prod_{i=2}^{j} (\lambda_i - \lambda_1)}$.

$$b_{jk} = \frac{\prod_{i=k}^{j-1} r_i\lambda_i}{\prod_{i=k+1}^{j} (\lambda_i - \lambda_k)} \left\{ \frac{Y_k}{\lambda_k - u} + \sum_{m=1}^{k-1} \left[Y_m \prod_{i=m}^{k-1} (r_i\lambda_i) \left(\frac{1}{\prod_{i=m}^{k} (\lambda_i - u)} - \sum_{i=m}^{k-1} \frac{1}{(\lambda_i - u) \prod_{\substack{n=m \\ n \ne i}}^{k} (\lambda_n - \lambda_i)} \right) \right] \right\} (1 < k \le j-1).$$

In eqn (5), $t = t_0$, and the zero-time number of the nuclide at different levels for the basic-state chain is obtained.

Number of nuclides on the excitation-state chain. When calculating the number of a nuclide on the excitation-state chain, ^{235}U fission contribution is considered only for the mother nuclide to prevent recycled counting. One may take the β^- excitation-state chain as an example: the number of every nuclide on the chain is N'_j (j=1, 2, …):

$$\frac{dN_1'}{dt} = \phi\sigma_f N_0 Y_1 e^{-ut} - \lambda_1 N_1'. \quad N_1' = \phi\sigma_f N_0 b_{10}'(e^{-ut} - e^{-\lambda_1 t}). \tag{6}$$

In eqn (6), $b_{10}' = \frac{Y_1}{\lambda_1 - u}$.

$$N_2' = \phi\sigma_f N_0\left[b_{20}'\left(e^{-ut} - e^{-\lambda_2 t}\right) + b_{21}'\left(e^{-\lambda_2 t} - e^{-\lambda_1 t}\right)\right]. \tag{7}$$

where $b_{20}' = \frac{r_1\lambda_1}{\lambda_2 - u} b_{10}'$, $b_{21}' = \frac{r_1\lambda_1}{\lambda_2 - \lambda_1} b_{10}'$.

With the same method:

$$\begin{aligned} N_j' = \phi\sigma_f N_0\big[&b_{j0}'\left(e^{-ut} - e^{-\lambda_j t}\right) \\ &+ b_{j1}'\left(e^{-\lambda_j t} - e^{-\lambda_1 t}\right) + \cdots \\ &+ b_{jk}'\left(e^{-\lambda_j t} - e^{-\lambda_k t}\right) + \cdots \\ &+ b_{j,j-1}'\left(e^{-\lambda_j t} - e^{-\lambda_{j-1} t}\right)\big] (j > 1). \end{aligned} \tag{8}$$

where $b_{j0}' = \frac{Y_1}{\lambda_1 - u}\prod_{i=2}^{j}\frac{r_{i-1}\lambda_{i-1}}{\lambda_i - u}$, $b_{j1}' = \frac{Y_1}{\lambda_1 - u}\prod_{i=2}^{j}\frac{r_{i-1}\lambda_{j-1}}{\lambda_i - \lambda_1}$.

$$b_{jk}' = \frac{Y_1\prod_{j=1}^{j-1}(r_i\lambda_i)}{\prod_{i=k+1}^{j}(\lambda_i-\lambda_k)}\left(\frac{1}{\prod_{i=1}^{k}(\lambda_i-u)} - \sum_{i=1}^{k-1}\frac{1}{(\lambda_i-u)\prod_{\substack{m=m\\ m\neq i}}^{k}(\lambda_m-\lambda_i)}\right) (1 < k \leq j-1).$$

When $t = t_0$, the zero-time number of every nuclide for the excitation-state chain is obtained using eqn (8).

Change rule for the number of nuclides after zero-time

In the decay chain, the growth and decay process of each nuclide can be described by the Bateman equation (Bateman 1843) as follows:

$$N_n(t) = \sum_{i=1}^{n}\sum_{k=1}^{n} {}_nC_{ki}N_i^0 e^{-\lambda_k t}. \quad (9)$$

$${}_nC_{ki} = \frac{\prod_{j=i}^{n-1}\lambda_j}{\prod_{j=i,\, j\neq k}^{n}(\lambda_j-\lambda_k)}. \quad (10)$$

where $N_n(t)$ is the number of nuclides n at time t, N_i^0 is the number of nuclide i ($i \leq n$) at zero-time, and λ_k is the decay constant of nuclide k ($k \leq n$).

According to the Bateman equation, one assumes that the first three nuclides, A, B, and C, possess decay constants of λ_1, λ_2, and λ_3, nuclide numbers at t_0 of $N_1(t_0)$, $N_2(t_0)$, and $N_3(t_0)$, and nuclide numbers at t ($t > t_0$) of $N_1(t)$, $N_2(t)$, and $N_3(t)$, respectively.

Number change of nuclide A, B, and C. As time goes on, the number of nuclide A undergoes an exponential decay:

$$N_1(t) = c_{10}e^{-\lambda_1(t-t_0)} \quad c_{10} = N_1(t_0). \quad (11)$$

Nuclide B is produced from A at the rate of $r_1\lambda_1 N_1$ and decays to C at the rate of $\lambda_2 N_2$:

$$\frac{dN_2}{dt} = r_1\lambda_1 N_1 - \lambda_2 N_2.$$

$$N_2(t) = c_{20}e^{-\lambda_1(t-t_0)} + c_{21}e^{-\lambda_2(t-t_0)}. \quad (12)$$

while $c_{20} = \frac{r_1\lambda_1}{\lambda_2-\lambda_1}c_{10}$, $c_{21} = N_2(t_0) - c_{20}$.

Therefore, after zero-time, the number of nuclide j in the linear chain is $N_j(t)$:

$$N_j(t) = c_{j0}e^{-\lambda(t-t_0)} + \cdots + c_{jk}e^{-\lambda_{k+1}(t-t_0)} \cdots + c_{j,j-1}e^{-\lambda_j(t-t_0)}. \quad (13)$$

when $c_{j0} = \frac{\prod_{i=1}^{j-1} r_i\lambda_i}{\prod_{i=2}^{j}(\lambda_i-\lambda_1)}N_1(t_0)$,

$$c_{jk} = \sum_{m=1}^{k}\left(\frac{\prod_{i=m}^{j-1} r_i\lambda_i}{\prod_{\substack{i=m\\ i\neq k+1}}^{j}(\lambda_i-\lambda_{k+1})}N_m(t_0)\right) + \frac{\prod_{i=k+1}^{j-1} r_i\lambda_i}{\prod_{i=k+2}^{j}(\lambda_i-\lambda_{k+1})}N_{k+1}(t_0) \quad (0 < k < j-1).$$

$$c_{j,j-1} = \sum_{m=1}^{j-1}\left[\left(\prod_{i=m}^{j-1}\frac{r_i\lambda_i}{\lambda_i-\lambda_j}\right)N_m(t_0)\right] + N_j(t_0).$$

Number of delayed gamma for arbitrary nuclides in the decay chain: Decay correction. When the measure time

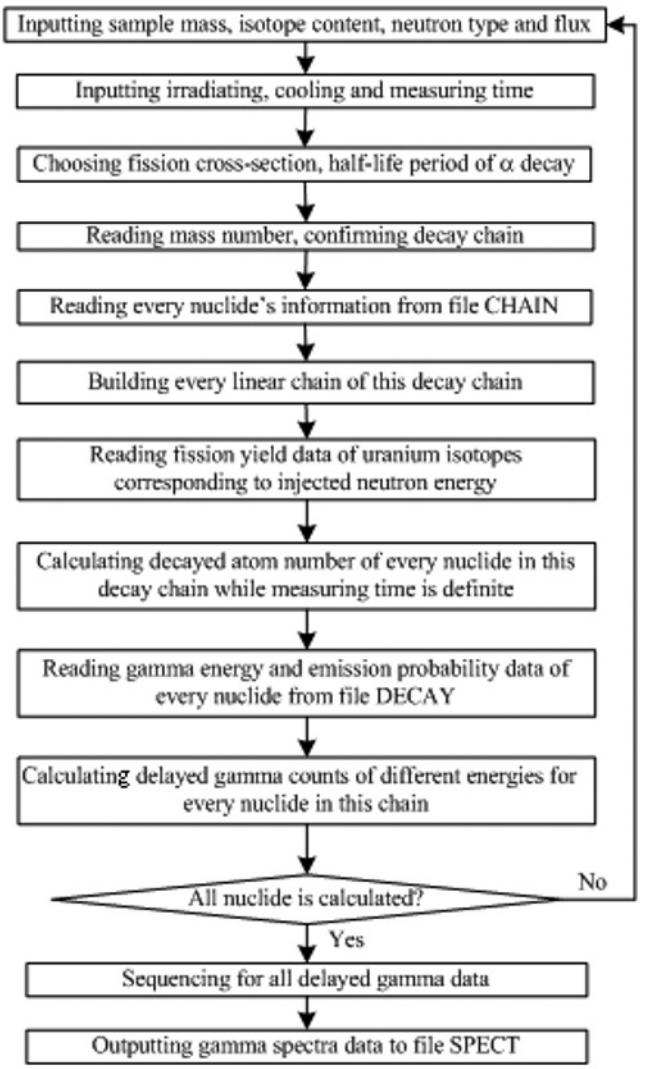

Fig. 2. Structure of main program.

is t_2–t_1, the decayed atom number of an arbitrary nuclide j is n_j (j = 1, 2, 3…). For the mother nuclide,

$$n_1 = \int_{t_1}^{t_2} \lambda_1 N_1(t_0) e^{-\lambda_1 t} dt = N_1(t_0)\left(e^{-\lambda_1 t_1} - e^{-\lambda_1 t_2}\right). \quad (14)$$

For progeny nuclides,

$$\begin{aligned} n_2 &= \int_{t_1}^{t_2} r_2 \lambda_2 \left(c_{20} e^{-\lambda_1 t} + c_{21} e^{-\lambda_2 t}\right) dt \\ &= r_2 \left[\frac{\lambda_2}{\lambda_1}\, c_{20}\left(e^{-\lambda_1 t_1} - e^{-\lambda_1 t_2}\right) + c_{21}\left(e^{-\lambda_2 t_1} - e^{-\lambda_2 t_2}\right)\right]. \end{aligned}$$

$$\begin{aligned} n_j &= \int_{t_1}^{t_2} r_j \lambda_j \left(c_{j0} e^{-\lambda_1 t} + \cdots + c_{jk} e^{-\lambda_k t} + \cdots + c_{j,j-1} e^{-\lambda_j t}\right) dt \\ &= r_j \left[\frac{\lambda_j}{\lambda_1} c_{j0}\left(e^{-\lambda_1 t_1} - e^{-\lambda_1 t_2}\right) + \cdots + \frac{\lambda_j}{\lambda_k} c_{jk}\left(e^{-\lambda_k t_1} - e^{-\lambda_k t_2}\right)\right. \\ &\quad \left. + \cdots + c_{j,j-1}\left(e^{-\lambda_j t_1} - e^{-\lambda_j t_2}\right)\right]. \end{aligned} \quad (15)$$

One nuclide may belong to numerous linear chains; therefore, the decayed atom number of every linear chain is added together.

Number of delayed gamma for arbitrary nuclides in the decay chain: Gamma emission probability correction. One assumes that nuclide j emits m gamma rays of different energy. For the k gamma ray, whose energy is E_k (k = 1, 2, …, m), the emission probability is P_k. Then, the total gamma rays produced by one nuclide of the decay chain are

$$n_j \sum_{k=1}^{m} P_k .$$

MONTE CARLO CALCULATION OF DELAYED GAMMA SPECTRA

Structure of the main program

The Monte Carlo program MCNP is used to build a physical model for delayed gamma spectra. Decay data of Firestone (1996) and fission yield data of James et al. (1991) are applied in the calculation.

The structure of the main program is as follows:

1. Inputting calculation parameter, reading the mass number of one fission product, and reading information on every nuclide from file CHAIN.txt, including proton number, decay mode, branch ratio, and half-life period;
2. In one decay chain, if one nuclide's proton number is at minimum, the nuclide is considered as a mother nuclide to build a β^- basic chain. If the proton number of one nuclide is at maximum, the nuclide is considered as a mother nuclide to build an EC basic chain. If nuclides exist in the excitation state, the nuclides are considered as mother nuclides to build an excitation-state chain;
3. According to the type of incoming neutron (thermal neutron, fissile neutron, or high-energy neutron), the fission yield data of every uranium isotope (^{234}U, ^{235}U, ^{236}U, and ^{238}U) is read;
4. Reading gamma energy and emission probability data from file DECAY.txt. Using formulas deduced by stage 3, the authors calculated the decayed atom number of every nuclide in the corresponding chain and then obtained the delayed gamma counts of different energies; and
5. Recycle calculation from mass number A = 66 to A = 172 was conducted. According to a certain algorithm, the delayed gamma data of all fission products is sequenced, and gamma spectra data were output to file SPECT.txt.

The structure of the main program is shown in Fig. 2.

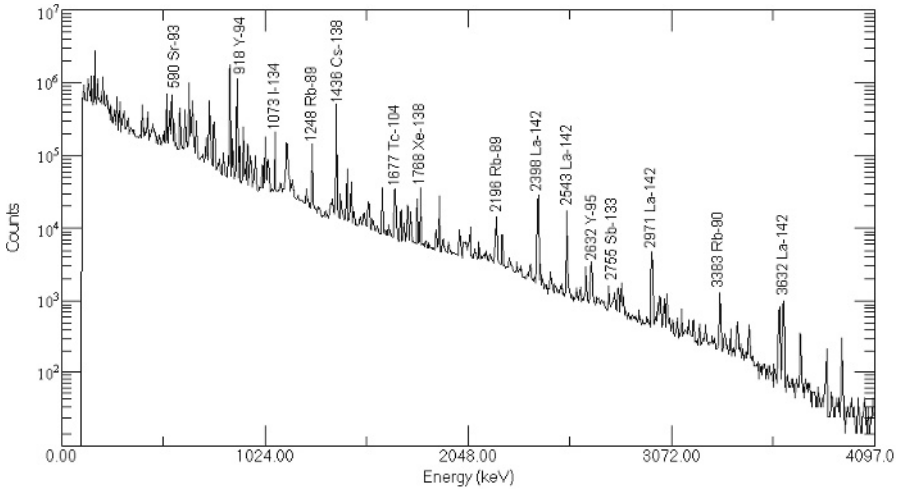

Fig. 3. Delayed gamma spectra.

Table 1. Delayed gamma counts from experiment and calculation.

Nuclide name	Delayed gamma energy (keV)	Counts from experiment	Counts from calculation	Deviation (%)
^{93}Sr	590.00	6.7610^5	6.58×10^5	−2.68
^{94}Y	918.00	1.26×10^6	1.23×10^6	−2.25
^{134}I	1072.55	2.21×10^5	2.14×10^5	−3.14
^{89}Rb	1248.10	2.21×10^5	1.59×10^5	−3.56
^{89}Rb	2196.00	1.48×10^4	1.41×10^4	−4.25
^{138}Cs	1435.86	5.27×10^5	5.12×10^5	−2.81
^{138}Xe	1768.00	3.67×10^4	3.54×10^4	−3.75
^{142}La	2397.80	2.58×10^4	2.48×10^4	−3.97
^{142}La	2542.70	1.76×10^4	1.69×10^4	−4.13
^{142}La	2971.00	4.36×10^3	4.17×10^3	−4.69
^{95}Y	2632.00	3.72×10^3	3.53×10^3	−4.94
^{133}Sb	2755.00	1.70×10^3	1.60×10^3	−5.51
^{90}Rb	3383.00	1.38×10^3	1.30×10^3	−5.67

Comparison between experiment and calculation results

Uranium material (^{235}U content is 90%) was irradiated by impulse reactor with an irradiating time of 10 min. The target weighed 0.3 mg. The thermal neutron flux was 1×10^{12} cm^{-2} s^{-1}. The irradiated sample was transferred to a lead shielding device by the rabbit assembly. The thicknesses of the lead plate, copper lining plate, cadmium lining plate, and Plexiglass were 100, 1, 1, and 5 mm, respectively. Coaxial N-type HPGe detector (model: GMX10180) was used to measure 1 h. Detection distance was 35 cm, and the dead-time ratio was 35.95%. The delayed gamma spectra are shown in Fig. 3.

After measurement, several typical fission products were selected for obtaining the corresponding delayed gamma counts. Based on the same technical condition, a simulating calculation was conducted using the current program. Table 1 shows the comparison between experimental and calculated delayed gamma counts.

In Table 1, the absolute value of deviation is in 2.25% to 5.67%. The calculated value is smaller than the experimental value because of the following aspects:

1. For gamma rays of specific energy, the measured count means the net area of related energy peak, which includes gamma ray counts of different nuclides whose energy values are close;
2. When building a decay chain, linear chains with extremely small branch ratios are not considered in the theoretical calculation; thus, the corresponding nuclide types are reduced; and
3. When deducting formulas, other situations that may produce delayed gamma rays are disregarded. For example, the fission phenomenon is induced by the prompt neutron and the (neutron, gamma) reaction between fission products and slow neutrons.

CONCLUSION

The physical model of a complex decay chain is simplified based on fission products research, and formulas of the changing rule for nuclide number before and after zero-time are deduced. The C program to calculate the delayed gamma ray spectra data is established, and related experiments are conducted to verify the formulas. Results show that the calculation values conform to the experimental values.

Research shows that, under certain conditions, the number of any nuclide on a decay chain is not only related with the half-time period of itself, the branch ratio of the decay chain, fission yield, and others but also on the physical parameters of all mother nuclides. The total number for one nuclide is contributed by all linear decay chains to which the nuclide belongs.

In future work, the effect of different shielding, shape, and size of uranium materials on the delayed gamma spectra will be focused on and studied.

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