THE EFFECTIVENESS OF REGISTRATION OF FISSILE MATERIALS BY VARIOUS METHODS

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Fission process of nuclear materials is accompanied by the yield of fission fragments and corresponding emission of γ-quanta and fission neutrons. In this work comparative analysis of effectiveness of registration of radioactive elements by two various methods: double γ-quanta coincidence and neutron detection methods. In the frame of developed approach the analysis of comparative effectiveness of registration methods for some fissile materials was done.

1. Introduction

Presently there are various methods of detection of fissile materials [1 - 4]. Our experience of using radiation portal monitors at Uzbekistan borders shows rather well their effectiveness [5]. But the modern trends of society development demand increasing of registration effectiveness of radioactive materials. First of all it is related to strengthening non-proliferation regime and prevention of illicit trafficking of nuclear and other radioactive materials. Besides, modern nuclear technologies are oriented on use of low-enrichment nuclear materials thus increasing the demands on nuclear security and radiation safety control. The methods of control should be more effective and fast. Earlier we studied the possibility of detection of fissile nuclear materials by double γ-γ coincidence method [4]. In present work we continue to study registration effectiveness with inclusion into consideration multi-particle correlations of registered γ-quanta and prompt fission neutrons.

2. Basic approach

Let's consider any radioactive element with atomic number A. Its mass is m, molar mass is μ_A half-life is $T_{1/2}$. Then the radioactivity of this material – number of nuclear decays per time unit is defined as: $A_{Rad} = \frac{\ln 2}{T_{1/2}} \frac{m}{\mu_A} N_A$, where N_A - Avogadro's number. As registered particle we'll consider prompt neutrons and multiple γ-quanta being accompanying fission products. Let's designate their number in each fission act as ν_n and ν_γ. Supposing the isotropic character of fission products distribution we consider effectiveness of registration of one neutron n and one γ - quantum in each fission act. Let probability of registration by detector of one neutron and one γ-quantum be q_n and q_γ correspondingly. Then using binomial distribution we get:

$$P_\gamma = C^1_{\nu_\gamma} \cdot q_\gamma (1-q_\gamma)^{\nu_\gamma - 1} . \tag{1}$$

Note that in case of γ-quanta registration radiation background is the factor to be excluded. Neutron background is much lower and in this case problem of background exclusion is nor so crucial. For solving the background problem the method of double γ - γ coincidence demanding two detectors is used. Then we have:

$$P_{\gamma\gamma} = \nu_\gamma (\nu_\gamma - 1) q_\gamma^2 (1-q_\gamma)^{2\nu_\gamma - 3} \equiv \frac{\nu_\gamma - 1}{\nu_\gamma (1-q_\gamma)} \cdot P_\gamma^2 . \tag{2}$$

We supposed here that two registering detectors are equivalent. For detection of one neutron, as it follows from (1) we have:

$$P_n = C^1_{\nu_n} \cdot q_n (1-q_n)^{\nu_n - 1} . \tag{3}$$

So, detection effectiveness of on neutrons and double γ - γ coincidence is given from comparison of relations (2) and (3). Here it's necessary to note that analysis of detection problem should include peculiarities knowledge of which is necessary for elaboration of effective registration methods. As an example we'll consider actinides, namely $^{235}_{92}U$, $^{238}_{92}U$, $^{239}_{94}Pu$ and $^{252}_{98}Cf$. The main thing representing fundamental difficulty is the following: during the fission of studied nuclei through various fission channels the main channel is α-decay. By this, as it is easy to show using half-lives, their specific activities are several orders higher than those of spontaneous fission (excluding $^{252}_{98}Cf$ nucleus). This fact nevertheless does not make task of detection of heavy radioactive elements easier because even in the presence of minimal lead shield (with thickness ~ 2 mm), the total absorption of fission products in lead is provided. It is due to the fact that characteristic radiation originating during α-particle movement is close by its energy parameters to lead absorption band. Therefore it's impossible to register fission products by external detectors with shields. In case of products of spontaneous fission channel, though fission products pass through lead shield (thickness ~ 30mm), but their specific activity is low on background level. So, in case of registration of fissile elements on their spontaneous fission products, the problem of separation of fission products registered by detectors from background appears. Note that in this case spontaneous fission is characterized by emission of accompanying prompt neutrons and γ -quanta. One of effective methods to solve this problem is the method of double γ - γ coincidence. The problem of separation from background in case of neutrons became urgent at low activities of registered radioactive elements. Let the effectiveness of detection of one neutron and one γ -quantum be written as:

$$q_n = \frac{S_D}{4\pi R^2}(1-\exp[-\bar{\sigma}_n n_D L]) , \quad q_\gamma = \frac{S_D}{4\pi R^2}(1-\exp[-\bar{\sigma}_\gamma n_D L]) , \tag{4}$$

where L - thickness of detector; n_D - concentration of atoms of working substance of detector; $\bar{\sigma}_n$ and $\bar{\sigma}_\gamma$ - averaged over energy cross sections of interaction with atoms of working substance of detector; S_D - working square of detector and R -characteristic distance from radioactive source to detector. Supposing that detector thickness $L >> \lambda$ mean free path in detector, one may assume neglecting edge effects that:

$$q_n = q_\gamma = q = \frac{S_D}{4\pi R^2} . \tag{5}$$

Then registration effectiveness of double γ - γ coincidence detection or neutron detection one can define as:

$$\eta = \frac{P_{\gamma\gamma}}{P_n} = \frac{\nu_\gamma(\nu_\gamma - 1)}{\nu_n} q(1-q)^{2(\nu_\gamma - 1)-\nu_n} . \tag{6}$$

Let us consider the further generalization of method of double γ - γ coincidence to increase the reliability of detection of fissile material. If one considers triple correlations of type γ - γ - γ or γ - γ - n coincidence, it is easy to obtain the following useful relations:

$$P_{\gamma\gamma\gamma} = \nu_\gamma \cdot (\nu_\gamma - 1) \cdot (\nu_\gamma - 2) q_\gamma^3 (1-q_\gamma)^{3\nu_\gamma - 6} , \tag{7}$$

$$P_{\gamma\gamma n} = \nu_\gamma \cdot \nu_n \cdot (\nu_\gamma - 1) q_n q_\gamma^2 (1-q_\gamma)^{2\nu_\gamma - 3} (1-q_n)^{\nu_n - 1} . \tag{8}$$

These relations may be generalized on case of arbitrary number of detectors, for example let them be N , then for coincidence scheme $\underbrace{\gamma-\gamma-\gamma....-\gamma-\gamma}_{N}$ we'll get the following relation:

$$P_{N\gamma} = C_{\nu_\gamma}^N q_\gamma^N (1-q_\gamma)^{N\cdot\nu_\gamma - N(N-1)/2} , \tag{9}$$

where $N \le \nu_\gamma$.

3. Numerical calculations

Let's consider registration effectiveness of nuclear materials by various methods on some examples. We'll calculate specific activities of: ^{235}U , ^{238}U , ^{239}Pu и ^{252}Cf . Registration effectiveness of fissile material depends on intensity of emitted radiation of fission product and the amount of radioactive substance. We'll make calculations of specific activity per 1 g of radioactive substance. If we consider the half-lives of fissile material in case of SF are known then specific activity of source may be calculated as follows:

$$A_R = \frac{N_A \ln 2}{A \cdot T_{1/2}} = \frac{const}{A \cdot T_{1/2}} , \tag{10}$$

where A - atomic number; N_A - Avogadro's number; $T_{1/2}$ - half-life in decay channel. Calculation results are shown in Table 1.

Table 1. **Specific activities of fissile materials at SF**

Nucleus	$T_{1/2}(SF)$, years	A_R , Bq
^{235}U	$(1 \pm 0.3)\cdot 10^{19}$	$4.3 \cdot 10^{-6}$
^{238}U	$(8.2 \pm 0.1)\cdot 10^{15}$	$6.7 \cdot 10^{-3}$
^{239}Pu	$(1 \pm 0.3)\cdot 10^{19}$	$5.54 \cdot 10^{-3}$
^{252}Cf	86 ± 1	$6.04 \cdot 10^{11}$

The obtained absolute activity of source needs measuring during detection taking into account its shielding (as a rule lead shield). Accounting shield the main fission products emitted outside are prompt neutrons and γ -quanta. For them we'll take number of neutrons and γ-quanta per one SF act at an average as 2 - 3 and 8 - 12, correspondingly. If we consider probability of detection of prompt neutrons and γ-quanta, then we take for estimation the following parameters:

$$S_D = 100 \ \ cm^2 ; \ r = 100 \ cm. ; \ \varepsilon_{eff(n)} = 0.2 ; \ \varepsilon_{eff(\gamma)} = 0.4 \tag{11}$$

Table 2. **Probabilities of neutron registration in one SF act**

Nucleus	ν_n	$P_n, \ \frac{n}{\sec}$
^{235}U	2.407	$3.83 \cdot 10^{-4}$
^{238}U	2.05	$3.26 \cdot 10^{-4}$
^{239}Pu	2.874·	$4.57 \cdot 10^{-4}$
^{252}Cf	3.724	$5.92 \cdot 10^{-4}$

In this case, taking into account obtained relations we may write: $q_\gamma \approx 3.176 \cdot 10^{-4}$ and $q_n \approx 1.592 \cdot 10^{-4}$. Results of probabilities of one neutron registration for spontaneous fission are shown in Table 2.

Here for ν_γ we took $\nu_\gamma = 8$. Having it in mind one can see that in accordance with relations (1) and (2) we get that registration probability $P_\gamma \approx 2.54 \cdot 10^{-3}$ and $P_{\gamma\gamma} \approx 1.5 \cdot 10^{-4}$. The values ν_n were taken from [6 - 8].

4. Conclusions

Table 3. **The registration effectiveness of neutrons and of events of γ-γcoincidence**

Nucleus	ε
^{235}U	1.3
^{238}U	1.8
^{239}Pu	0.76
^{252}Cf	0.41

The number of registered by detector particles is determined as product of registration probability of one particle multiplied by the number of particle emitted by fissile material. Let's suppose that $N\gamma = A_{Rad} \cdot \nu_\gamma$ and $Nn = A_{Rad} \cdot \nu_n$ correspondingly. Then the registration effectiveness of neutrons and of γ-γ coincidence events may be estimated by the following relation: $\varepsilon = \frac{\nu_\gamma}{\nu_n} \frac{P_{\gamma\gamma}}{P_n}$. Calculation results are presented in Table 3.

Here we took lower limit of value $\nu_\gamma = 8$. In case if higher value is taken, registration effectiveness of double γ-γ coincidence increases approximately1.5 times. So in this work the approach was developed and possibility of determining the registration effectiveness of prompt γ-quanta by double γ-γ coincidence method in comparison with neutron registration method was studied. This statement needs more detailed study using calculations made in spirit of work [9]. But from Table 3 it is possible already to make conclusion for elaboration of detection technique based on results obtained.

REFERENCES

1. *Gurevich I.I., Tarasov L.V.* Fizika nejtronov nizkikh ehnergij. - Moscow: Nauka, 1965. - 607 p.
2. *Kramer-Ageev E.A., Lavrenchik V.N., Samosadnyj V.T., Protasov V.P.* Ehksperimental'nye metody nejtronnykh issledovanij. - Moscow, 1990. - 272 p.
3. *Reilly D., Ensslin N., Smith H., Kreiner S.* Passive Nondestructive Assay of Nuclear Materials. LA-UR-90-732, Los Alamos National Laboratory, Los Alamos, New Mexico, 1991.
4. *Karimov Yu.N., Shipilov N.N., Fazilov M.I. et al.* // J. of Applied Radiation and Isotopes. - 2005. - Vol. 63. - P. 655 - 658.
5. *Petrenko V.D., Yuldashev B.S., Ismailov U.N. et al.* // AIP Proceedings. - 2009. - Vol. 1194. - P. 49 - 55.
6. *Gorbachev V.M., Zamyatin Yu.S., Lobov A.A.* Vzaimodejstvie izluchenij s yadrami tyazhelykh ehlementov i delenie yader. - Moscow: Atomizdat, 1976. - 461 p.
7. *Holden N. E., Hoffman D. C.* // Pure and Applied Chemistry. - 2000. - Vol. 72, No. 8. - P. - 1525.
8. *NuDAT 2.5*, http://www.nndc.bnl.gov/nudat2/, (National Nuclear Data Center, Brookhaven National Laboratory, Uoton, NY, 2009).
9. *Khokonov A.A., Savojskij Yu.V., Kamarzaev A.V.* // Yadernaya Fizika. - 2010. - Vol. 73, No. 9. - P. 1528 - 1532.