Measurement of the neutron source strength of a spontaneous fission source based on ^{244}Cm and ^{252}Cf by the calorimetric method

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The sources of spontaneous fission neutrons based on ^{252}Cf, ^{244}Cm, ^{248}Cm, ^{238}Pu, ^{240}Pu etc. are used in the metrological practice along with (α,n) sources.
Since neutrons in these sources are produced as a result of the acts of spontaneous fission only, in order to determine the neutron source strength we need to know with a reasonable accuracy the spontaneous fission activity (number of fission's per unit of time) and the average number of neutrons per act of spontaneous fission.

The number of neutrons irradiated by such sources depends, actually, on the quantity of fissile substance only , and their flux can be determined by precise weighing.

The most suitable materials for neutron sources are ^{244}Cm and ^{252}Cf, which are not fissioned by thermal neutrons, and can be purified well by modern radiochemical methods. They have a convenient half-life time, and a low intensity of associated gamma - radiation.

The neutron source strength can be calculated by the formula:

$$\Phi = A_f \, \nu \, (1+k_1)(1+k_2)\, k_3 \, , \qquad /1/$$

where
A_f is the number of acts of spontaneous fission of the basic radionuclide (hereinafter referred to as the activity of a spontaneous fission);
ν is the average number of neutrons per act of spontaneous fission;
k_1 is the relative part of neutrons born at (α,n)-reactions in construction materials;
k_2 is the relative part of neutrons born in the fission process of the other spontaneously fissineg isotopes;
k_3 is the correction for capture of neutrons by construction materials;
The activity value can be determined from measurements of the heat flux of a source W by a calorimetric method.
The calorimetric method is very convenient, due to the following properties of the source:
- the energy release of the source comes mostly from the basic isotope (^{244}Cm or ^{252}Cf). The remaining part of it, coming from isotopes and "daughters" elements, can be taken into account by some corrections. The uncertainty of the nuclear data does not influence the final output;
- material of the source does not practically contain any light elements (except for oxygen), which may contribute to the full neutron source strength due to (α,n) reactions;
- according to the manufacturer data, uncertainty of isotopic composition at the time of packaging the source does not exceed 0.3 %. This fact minimizes the uncertainty due to the influence of other isotopes.

The full thermal flux is

$$W = \sum_{i=1}^{n}(W_{\alpha i} + W_{f.i}) = \sum_{i=1}^{n}(A_{\alpha i}E_{\alpha i} + A_{f.i}E_{f.i}) \qquad /2/$$

where "*f*" corresponds to a spontaneous fission, "α" corresponds to the other fission, that , with a small exeption, is an α-fusion. For ^{244}Cu the total energy release of α-decay and spontaneous fission is about 99.82% of the full energy.

A is the number of the corresponding nuclear reactions per a unit of time.

E is the energy, released at the nuclear reaction.

n is the number of fissile radionuclide in the source material

The relation between α-decay and spontaneous fission is given by the equation:

$$A_f = A_\alpha \frac{T_\alpha}{T_f}, \qquad /3/$$

where T is the corresponding half-life time.

The activity of a radionuclide is connected with its mass by the well known formula:

$$A = m\frac{N_o \ln 2}{M_z T}, \qquad /4/$$

where N_o is the Avagadro number;

M_z is the atomic mass,

T is the half-life time.

The mass ratio of radionuclides in the mixture (***m***) are equal to the ratio of their concentrations (***μ***).

Therefore equation (2) for the full thermal flux can be written the following way:

$$W = A_{f0}\{\frac{T_{f0}}{T_{\alpha 0}}E_{\alpha 0} + E_{f0} + \sum_{j=1}^{n-1}\frac{A_{\alpha j}E_{\alpha j} + A_{fj}E_{f.j}}{A_{f0}}\}, \qquad /5/$$

where $A_{f0}E_{f0}$ is the energy release from the spontaneous fission of the main radionuclide (^{244}Cm or ^{252}Cf).

or

$$W = A_{f0}E_{f0}\{1 + \frac{T_{f0}}{T_{\alpha 0}}\frac{E_{\alpha 0}}{E_{f0}} + \frac{T_{f0}M_0}{E_{f0}\mu_0} + \sum_{j=1}^{n-1}\frac{\mu_j}{M_j}\left(\frac{E_{\alpha j}}{T_j} + \frac{E_{fj}}{T_{fj}}\right)\} = A_{f0}E_{f0}(1+K), \qquad /6/$$

where K is the contribution into the full energy release from all the channels except spontaneous fissioned of the main radionuclide.

Thus, according to the equation (1):

$$\Phi = W\frac{W}{E_f}\frac{1}{(1+K)}(1+k_1)(1+k_2)(1-k_3) \qquad /7/$$

The coefficient k_2 for the contribution into the full neutron flux from the other spontaneously fissioned isotopes can be calculated from the following equation

$$k_2 = \frac{M_0 T_{f0}}{\mu_0 \nu_0}\sum_{j=1}^{n-1}\frac{\mu_i \nu_i}{M_i T_{fi}} \qquad /8/$$

For calculation of the correcting factors k_1, k_2, k_3, K, the manufacturer's data on the isotopic composition and the shell material were used together with the nuclear data from publications (1, 2, 3, 4).

Table presents the results of the neutron source strength measurements by a nonconventional calorimetric method in comparison with the results obtained using a traditional "***Mn***-bath" method. The overall uncertainty of both methods is less than 1%.

Source	Results of neutron source strength measurements	
	The calorimetric method	***Mn***-bath
Cm-244 № 46.8/01	$2.60 \cdot 10^6$ c^{-1}	$2.75 \cdot 10^6$ c^{-1}
Cm-244 № 46.8/02	$2.53 \cdot 10^6$ c^{-1}	$2.67 \cdot 10^6$ c^{-1}
Cf-252 № 10-9/01	$2.46 \cdot 10^7$ c^{-1}	$2.44 \cdot 10^7$ c^{-1}
Cf-252 № 7-9/12	$1.12 \cdot 10^6$ c^{-1}	$1.12 \cdot 10^6$ c^{-1}

The coincidence of the results from ^{252}Cf -source confirms the correctness of the measurement procedure.

At present attempts are being made to explain this difference for the ^{244}Cm -source.

References

1. V.V.Malinovsky. Semi-empirical calculation of the average number of instantaneous fission neutrons. – Problems of Nuclear Science and Technology. Series: Nuclear Constants. № 2, 1987, p.25 (In Russian).
2. B.I.Starostov at al. Analysis and estimation of the experimental data on spectra of instantaneous fission neutrons. – Problems of Nuclear Science and Technology. Series: Nuclear Constants. № 2, 1980, p.3 (In Russian).
3. V.V.Frolov. Nuclear physical methods for control of fissers. – M.: Energoatomisdan, 1989. (In Russian)
4. L.G.Manevich et al. Calculation of the integral characteristics of delayed neutrons. – Problems of Nuclear Science and Technology. Series: Nuclear Constants. № 2, 1988, p.3 (In Russian).
5. Decay Data of Transactinium Nuclides. Technical Report № 261, IAEA, Vienna, 1986.