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A novel method for the activity measurement of large-area beta reference sources

D. Stanga [a,*], P. De Felice [b], J. Keightley [c], M. Capogni [b], M.R. Ioan [a]

[a] National Institute of R&D for Physics and Nuclear Engineering-Horia Hulubei, IFIN-HH, P.O. Box MG-6, Bucharest-Magurele R-077125, Romania
[b] Instituto Nazionale di Metrologia delle Radiazioni Ionizzante, ENEA, C.R. Casaccia, P.O. Box 2400, I-00100 Rome, Italy
[c] National Physical Laboratory, Hampton Road, Teddington TW11 0LW, UK

HIGHLIGHTS

- A novel method for determining the activity of large-area sources is described.
- The method makes use of two emission rate measurements.
- It has uncertainties smaller than the limit of 10% required by ISO 8769.

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ABSTRACT

A novel method has been developed for the activity measurement of large-area beta reference sources. It makes use of two emission rate measurements and is based on the weak dependence between the source activity and the activity distribution for a given value of transmission coefficient. The method was checked experimentally by measuring the activity of two (^{60}Co and ^{137}Cs) large-area reference sources constructed from anodized aluminum foils. Measurement results were compared with the activity values measured by gamma spectrometry. For each source, they agree within one standard uncertainty and also agree within the same limits with the certified values of the source activity.

1. Introduction

Large-area reference sources are used for the calibration of contamination monitors and their characteristics are specified by ISO 8769 (ISO, 2010) standard. According to this standard, the surface emission rate of Class 1 large area beta reference sources (end-point energy greater than 150 keV) shall be measured by the national metrology institute with a standard uncertainty not exceeding 3% and the activity shall be derived by the manufacturer and stated with a standard uncertainty smaller than 10%.

There is a general opinion that the calibration of contamination monitors must be performed in terms of surface emission rate but the aim of contamination measurements is the measurement of the activity per unit area (ISO, 2010, 1988). It is possible to obtain a better accuracy of contamination measurements by calibrating the contamination monitors in terms of activity using appropriate large-area reference sources. For this purpose, it is desirable for the national metrology institute to have the capability of independently determining the activity of large-area reference sources. Methods for determining their activity have already been reported by several authors (Janssen and Klein, 1996; Berger, 1998; Stanga, 2014; Javornik and Svec, 2014).

This paper describes a novel method for the activity measurement of large-area beta emitting reference sources constructed according to the standard ISO 8769 requirements (active layer of the source is not too thick). The method was checked experimentally by measuring two certified large-area reference sources (^{60}Co and ^{137}Cs) constructed from anodized aluminum foils. Measurement results were compared with the activity values determined by gamma spectrometry and with the certified values of the activity. For each source, they agree within one standard uncertainty. Uncertainties smaller than the limit of 10% required by ISO 8769 were also obtained.

2. Theoretical basis of the method

We consider the large-area beta source shown in Fig. 1. The surface emission rate, E_S, of the source can be calculated using the plane source concept (Stanga, 2014). Thus, we have

* Corresponding author.
E-mail address: doru@nipne.ro (D. Stanga).

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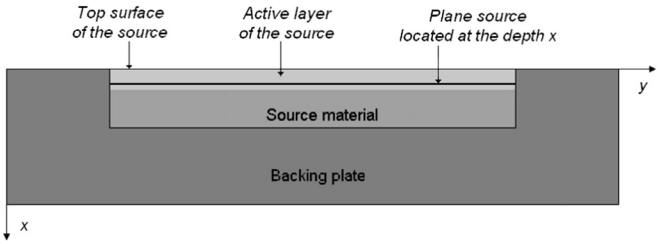

Fig. 1. Schematic view of a large-area source.

$$E_S = f_T \Lambda \int_0^{x_{\max}} \varepsilon_p(x) f(x) \mathrm{d}x \tag{1}$$

where $\varepsilon_p(x)$ is the plane source efficiency, f_T is the total emission probability for beta particles and conversion electrons, Λ is the activity of the source, $x_{\max}$ represents the thickness of the active layer of the source and $f(x) = \Lambda_p(x)/\Lambda$ is the activity depth distribution ($\Lambda_p(x)\mathrm{d}x$ is the activity of the plane source with thickness dx from the depth x).

The efficiency, $\varepsilon_p(x)$, of the plane sources is given by

$$\varepsilon_p(x) = \frac{E_p(x)}{f_T \Lambda_p(x)} = \frac{E_b(x) + E_{ce}(x)}{f_T \Lambda_p(x)} = \frac{f_b \varepsilon_{pb}(x) + f_{ce} \varepsilon_{ce}(x)}{f_T} \tag{2}$$

where $E_b(x)$, f_b and $\varepsilon_{pb}(x)$ are, respectively, the emission rate in 2π, the emission probability and the plane source efficiency corresponding to beta particles, $E_{ce}(x)$, f_{ce} and $\varepsilon_{pce}(x)$ are, respectively, the emission rate in 2π, the total emission probability and the plane source efficiency corresponding to conversion electrons and $f_T = f_b + f_{ce}$.

When the source is covered by an inactive foil of thickness s, the emission rate in 2π of both beta particles and conversion electrons that emerge from the top surface of the covering foil, $E(s)$, is given by

$$E(s) = f_T \Lambda \int_0^{x_{\max}} \varepsilon_p(x, s) f(x) dx \tag{3}$$

where $E(0) = E_S$ and $\varepsilon_p(x, 0) = \varepsilon_p(x)$.

In case that the source and the covering foil are made from the same material, Eq. (3) becomes

$$E(s) = f_T \Lambda \int_0^{x_{\max}} \varepsilon_p(x + s) f(x) dx \tag{4}$$

The transmission coefficient, t, is defined as the fraction of beta particles and conversion electrons emitted by the source and transmitted through a foil of thickness s. As a result, we have

$$t = \frac{E(s)}{E_S} = \frac{\int_0^{x_{\max}} \varepsilon_p(x, s) f(x) dx}{\int_0^{x_{\max}} \varepsilon_p(x) f(x) dx}, \tag{5}$$

where $\varepsilon_p(x, s) = \varepsilon_p(x + s)$ when the source and the covering foil have identical effective atomic numbers.

The efficiency of ^{60}Co and ^{137}Cs plane sources, located at the depth x in aluminum (backing plate is also made from aluminum), was calculated by Monte Carlo method using the PENCYL code from the simulation package PENELOPE (Baro et al., 1995; Salvat et al., 2003). In the case of ^{137}Cs sources, we took into account both beta particles ($f_b = 1$) and conversion electrons ($f_{ce} = 0.094$). Monte Carlo results were fitted by the function $\varepsilon_p(x) = a_0 + a_1 x^{0.5} + a_2 x + a_3 x^{1.5} + a_4 x^2 + a_5 x^{2.5}$ with residuals smaller than 0.5%. In Table 1 the fitting parameters a_0, a_1, a_2, a_3, a_4 and a_5 for ^{60}Co and ^{137}Cs sources are shown.

3. Description of the method

3.1. Dependence of the source activity on the transmission coefficient

From Eq. (4), it follows that

$$\Lambda = E(s)/K, \tag{6}$$

$$\text{where } K = \int_0^{x_{\max}} \varepsilon_p(x + s) f(x) dx. \tag{7}$$

Eq. (6) shows that Λ depends on both x_{max} and $f(x)$ which are usually unknown. Eq. (5) shows that t depends also on x_{max} and $f(x)$. It follows that Λ can be expressed as a function of t and $f(x)$ where t can be determined by measuring $E(s)$ and E_S. To investigate the dependence of Λ on $f(x)$, we used two opposite distributions $f_1(x) = \frac{1+\alpha}{x_{max}}(1 - \frac{x}{x_{max}})^{\alpha}$ and $f_2(x) = \frac{1+\alpha}{x_{max}}(\frac{x}{x_{max}})^{\alpha}$ together with the uniform distribution $f_3(x) = 1/x_{max}$. In the case of $f_1(x)$, the activity is concentrated close to the top surface of the source ($\alpha \geq 1$) while for $f_2(x)$ the activity is concentrated close to x_{max} ($\alpha \geq 1$). Fig. 2 shows K as a function of t for ^{60}Co and ^{137}Cs sources constructed from anodized aluminum foils (covered with an aluminum foils of thickness $s = 2.4$ mg/cm^2) and for the distributions mentioned above ($\alpha = 1$ and $\alpha = 5$). As one can see from Fig. 2, K depends weakly on $f(x)$ for a given value of t. It follows that Λ depends weakly on $f(x)$ and this dependence is also weak for other beta emitters such as ^{14}C, ^{147}Pm, ^{36}Cl and ^{90}Sr-^{90}Y. One can also see from Fig. 1 that K takes the maximum value $K_{\max}$ and the minimum value $K_{\min}$ for $f_2(x)$ and $f_1(x)$ for $\alpha = 5$, respectively.

3.2. Basis of the method

The method described here can be applied to different types of large-area beta reference sources constructed according to ISO 8769, namely, the active layer of the source is not too thick. It makes use of two emission rate measurements. Thus, the surface emission rate, E_S, is firstly measured according to ISO 8769 using a windowless gas-flow proportional detector. Secondly, the source is covered with a foil (it is assumed that both the foil and source material have identical effective numbers) of thickness s and the emission, $E(s)$, is measured with the same detector. In this way, the transmission coefficient can be calculated using $t = E(s)/E_S$.

The method makes use of Eq. (6) and is based on the weak dependence of Λ on $f(x)$ for a given value of t. As a result, the source activity Λ can be expressed as

$$\Lambda = f_m \frac{E(s)}{K}, \tag{8}$$

where f_m is a factor which is equal to one and takes into account the uncertainties of the model used for obtaining $\varepsilon_p(x + s)$ and computing K. If the interval $[K_{min}, K_{max}]$ is the only available information regarding K, a rectangular distribution over this interval can be assumed. It follows that

Table 1
Fitting parameters for ^{60}Co and ^{137}Cs plane sources (backing and source are made from aluminum).

	a_0	a_1	a_2	a_3	a_4	a_5
^{60}Co	0.717789557	−0.340266940	0.084607399	−0.020256230	0.003832716	−0.000318940
^{137}Cs	0.714323109	−0.176106937	0.023339380	−0.003506495	0.000442019	−0.000023705

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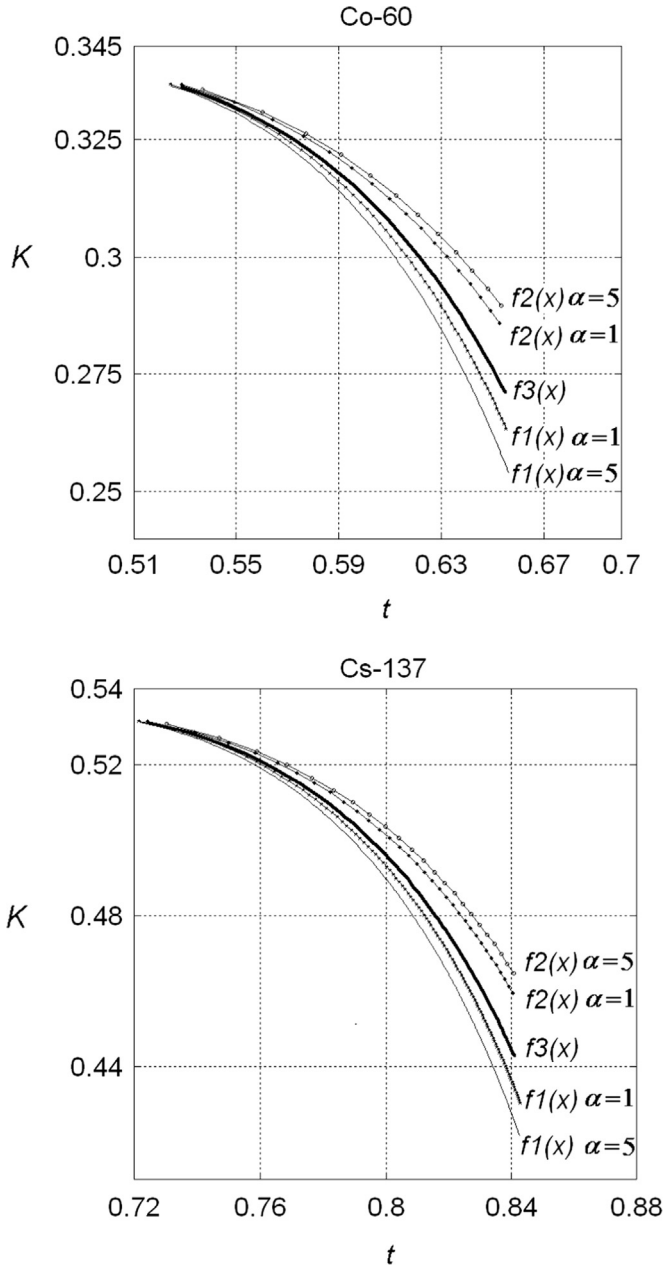

Fig. 2. K as a function of t for ^{60}Co and ^{137}Cs large-area sources constructed from anodized aluminum foils.

$$K = (K_{min} + K_{max})/2 \tag{9}$$

$$\delta_f(K) = (K_{\max} - K_{\min})/(\sqrt{3}\,(K_{\max} + K_{\min})) \tag{10}$$

where $\delta_f(K)$ is the relative standard uncertainty of K due to the fact that $f(x)$ is not exactly known.

To apply the method to the activity measurement of large-area beta reference sources constructed from anodized aluminum foils, K_{max} and K_{min} were computed as a function of t for different s values using Eqs. (5) and (7). The data were firstly fitted by least square method, for a given value of s, by means of polynomials of degree three. Thus, we have

$$K_{max} = \sum_{i=1}^{4} p_i(s)t^{4-i} \quad \text{and} \quad K_{min} = \sum_{i=1}^{4} q_i(s)t^{4-i} \tag{11}$$

The values of $_{pi}(s)$ and $q_i(s)$, obtained in the first step for different values of s, were also fitted by least square method using the following polynomials

$$p_i(s) = \sum_{j=1}^{4} P_{ij}s^{4-j} \quad \text{and} \quad q_i(s) = \sum_{j=1}^{4} Q_{ij}s^{4-j} \tag{12}$$

A Matlab script was written to compute P_{ij} and Q_{ij} for ^{60}Co and ^{137}Cs sources constructed from anodized aluminum foils. Expressed in matrix form, values of P_{ij} and Q_{ij} for these sources are

Pco-60 =	0.42313	−5.6149	26.1080	−47.5870
	−1.00500	13.1860	−60.2370	10.0890
	0.77050	−9.9850	44.4570	−69.9690
	−0.19161	2.4526	−10.7000	16.4460
Qco-60 =	0.133820	−1.84620	8.4152	−20.6970
	−0.307910	4.12450	−19.6250	40.1360
	0.223950	−2.97400	13.9370	−25.0930
	−0.052696	0.69329	−3.1939	5.6036
Pcs-137 =	0.22244	−4.9174	36.356	−97.578
	−0.56648	12.4640	−91.277	234.280
	0.47614	−10.4180	75.384	−186.560
	−0.13226	2.8771	−20.591	49.968
Qcs-137 =	0.071848	−1.63040	12.4510	−40.916
	−0.179060	4.03520	−30.8710	92.471
	0.146000	−3.26970	24.7280	−68.875
	−0.039104	0.87006	−6.4978	17.578

The values of P_{ij} and Q_{ij} are valid for x_{max} smaller than 5.4 mg/cm^2 (for large-area sources constructed from anodized aluminum foils, x_{max} is usually smaller than 3 mg/cm^2) and s chosen in the interval [2, 5] mg/cm^2 for ^{60}Co sources and the interval [2, 10] mg/cm^2 for ^{137}Cs sources.

The source activity Λ and its associated standard uncertainty were computed using the propagation of distributions according to GUM Supplement 1 (ISO, 2008). Consequently, a Matlab script was developed for computing the probability distribution of Λ using Monte Carlo simulation, Eq. (8) and normal probability distributions for the values of the input quantities s, t, f_m and $E(s)$. The standard uncertainty of f_m (due to the uncertainties of the model used for obtaining $\varepsilon_p(x+s)$ and computing K) was taken equal to 0.015. The Monte Carlo simulation generates n_d draws from the probability distributions of all input quantities and Λ is calculated for each of n_d draws resulting the values Λ_i where $i=1, 2,\ldots,n_d$. The average $\overline{\Lambda}$ and its associated standard uncertainty $u(\overline{\Lambda})$ are computed using

$$\overline{\Lambda} = \frac{1}{n_d}\sum_{i=1}^{n_d}\Lambda_i \quad u^2(\overline{\Lambda}) = \frac{1}{n_d - 1}\sum_{i=1}^{n_d}(\Lambda_i - \overline{\Lambda})^2 \tag{13}$$

To compute Λ_i, the value of K was sampled from the rectangular distribution defined on the interval $[K_{min}, K_{max}]$. The values of K_{min} and K_{max} for each sample are computed by means of Eq. (11) using the matrices written above. The quantities $\overline{\Lambda}$ and $u(\overline{\Lambda})$ are taken, respectively, as an estimate of Λ and its associated standard uncertainty. It should be noted that the uncertainty of Λ can also be evaluated using the law of propagation of uncertainty (ISO, 1995).

4. Experimental results

4.1. Experimental results obtained by gamma spectrometry

The activity of two large-area reference sources (^{60}Co and ^{137}Cs) was firstly measured by gamma spectrometry. These sources, of

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rectangular shape, are constructed from 0.3 mm thick anodized aluminum foils of $(10\times10)\,cm^2$ mounted on backing plates of $(12\times12)\,cm^2$ and 3.0 mm thickness.

A high resolution gamma spectrometric system was used in the measurements. It consists of a HPGe detector type GEM60P4 having a relative efficiency of 65% (1.33 MeV) and a resolution (FWHM) of 1.92 keV at 1.33 MeV. It is placed in a shielding made of lead bricks 10 cm thick with 1 mm cadmium and 1 mm copper inner lining. The detector is linked to a digital spectrometer DSPEC jr. 2.0 which is connected to the computer through USB (Universal Serial Bus). GammaVision 32 software was used for data acquisition and spectrum analysis. In all measurements the average dead time loss was smaller than 5% and long counting times were chosen to achieve very good counting statistics.

The activity of the sources was measured by the method of efficiency transfer using point sources (^{60}Co and ^{137}Cs) for reference measurements. According to this method, the activity of large-area sources, Λ_g, was calculated by means of the following equation

$$\Lambda_g = \frac{R_g(E)}{T\cdot R_{ref}(E)}\Lambda_{ref} \tag{14}$$

where T is the efficiency transfer factor, $R_g(E)$ and $R_{ref}(E)$ are, respectively, the net counting rates (corrected for coincidence summing effect in the case of ^{60}Co sources) corresponding to the energy E of the peaks of large-area and reference point sources and Λ_{ref} is the activity of the reference point sources. The efficiency transfer factor was computed using GESPECOR code (Sima and Arnold, 2002). To determine $R_g(E)$, the large-area sources were placed at 20.6 cm from the end cap of the detector with the center located on the detector axis. The reference sources were placed on the detector axis at 20.7 cm from the end cap. Measurement results and their associated uncertainties together with the certified values of the source activity (Λ_{cert}) and its associated standard uncertainty are shown in Table 2. One can see from Table 2 that the measurement result of each source agrees within one standard uncertainty with the certified value of the activity.

4.2. Experimental results obtained by the novel method

The same sources measured by gamma spectrometry were used for testing the performance of the novel method. Thus, they were measured according to ISO 8769 using a windowless gas-flow proportional counter type DEXTRAY AB710 $(250\times400\times30)\,mm^3$ provided with a sliding tray to insert the source into the active volume of the detector. The counter was operated at +3050 V in the beta plateau using methane as counting gas. The gas-flow of about 1 L/h was controlled by a two-stage pressure regulator and needle valves. A charge-sensitive preamplifier and a spectroscopy amplifier, which feeds a discriminator, amplified the signals of the detector. The pulses from the discriminator were counted by conventional counting logic. A dead time of 10 μs was superimposed to allow precise corrections of dead time losses. The discrimination level in the counting system was set to 0.59 keV using a multi-channel analyzer and a ^{55}Fe source.

The counting rate of each source was measured in the first step by placing it inside the detector volume. In the second step each source was covered with an aluminum foil of thickness $s=2.4\pm0.05\,mg/cm^2$ and the counting rate measured by means of the same detector and counting system. The emission rates were calculated by using the following equation:

$$E(s) = f_R\frac{R(s)}{1-\tau\cdot R(s)} - B \tag{15}$$

where, $R(s)$ is the counting rate corresponding to a covering foil of thickness s, $\tau=10$ μs is the dead time of the counting system, B is the detector background and f_R is a correction factor which is equal to unity (Stanga and De Felice, 2009). The detector background, B, was measured by placing the sources inside the detector volume with the active layer just above the sliding tray so that beta particles emitted by them are completely absorbed by the backing plate and sliding tray. In this way, only gamma rays can be detected by the proportional counter. The standard uncertainty of $E(s)$ was derived from Eq. (15) using the law of propagation of uncertainty and taking $f_R=1$. Table 3 shows the measurement results and their associated uncertainties together with the calculated value of t and its associated standard uncertainty.

The Matlab script described above was employed for computing the activity, Λ, of sources and their associated standard uncertainties using the transmission coefficients from Table 3. A number of 10^5 Monte Carlo trials ($n_d=10^5$) were performed and the script was run several times to check the stability of the results. In Table 4 are shown the computed values of K, Λ and their associated standard uncertainty for both ^{60}Co and ^{137}Cs sources. One can see from Tables 4 and 2 that the measurement results of the novel method agree within one standard uncertainty with the measurement results obtained by gamma spectrometry. They also agree within one standard uncertainty with the certified values of the source activity.

5. Conclusions

A novel method has been developed for determining the activity of large-area beta reference sources which is based on the weak dependence between the source activity and its depth distribution $f(x)$ for a given value of the transmission coefficient. It can be applied to different types of large area beta reference sources constructed according to ISO 8769, namely, the active layer of the source is not too thick. Here, the method was applied to the measurement of two large-area sources (^{60}Co and ^{137}Cs) constructed from anodized aluminum foils. A Matlab script was developed for estimating the activity of sources and their associated standard uncertainties. The performance of the novel method was tested experimentally by comparing its measurement results with the results obtained by gamma spectrometry and with the certified values of the activity. For each source, they agree within one standard uncertainty. In addition, uncertainties smaller than the limit of 10% required by ISO 8769 were obtained.

Table 2
Measurement results and their associated uncertainties obtained by gamma spectrometry together with the certified values of the source activity and its associated standard uncertainty.

Nuclide	T	$u(T)$	R_{ref} in s^{-1}	$u(R_{ref})$ in s^{-1}	R_g in s^{-1}	$u(R_g)$ in s^{-1}	Λ_{ref}^* in Bq	$u(\Lambda_{ref})$ in Bq	Λ_g in Bq	$u(\Lambda_g)$ in Bq	Λ_{cert}^* in Bq	$u(\Lambda_{cert})$ in Bq
^{60}Co	0.974	0.015	3.02	0.015	1.59	0.010	3035	45	1641	37	1648	41
^{137}Cs	0.973	0.015	4.38	0.018	3.09	0.012	3220	48	2335	51	2337	58

* Values of Λ_{cert} and Λ_{ref} are given for the actual date of measurements.

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Table 3
Measured values of E_S, $E(2.35)$, t and their associated standard uncertainties.

Nuclide	B in s^{-1}	$s=0$ mg/cm^2			$s=2.35$ mg/cm^2			t	$u(t)$
		$R(0)$ in s^{-1}	$E_S=E(0)$ in s^{-1}	$u(E_S)$ in s^{-1}	$R(2.35)$ in s^{-1}	$E(2.35)$ in s^{-1}	$u(E(2.35))$ in s^{-1}		
^{60}Co	52.7	905.2	860.8	4.7	554.8	505.3	3.9	0.5870	0.006
^{137}Cs	33.8	1499.5	1488.5	12.7	1181.3	1161.6	8.1	0.7804	0.009

Table 4
Computed values of K, Λ and their associated standard uncertainties for ^{60}Co and ^{137}Cs large-area reference sources constructed from anodizing aluminum foils.

^{60}Co				^{137}Cs			
K	$u(K)$	Λ in Bq	$u(\Lambda)$ in Bq	K	$u(K)$	Λ in Bq	$u(\Lambda)$ in Bq
0.3195	0.0030	1581.8	30.6	0.5114	0.0059	2271.7	46.1

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