Application of the MKGB-01 Spectrometer-Radiometer in the KX-Gamma Coincidences Setup at the D.I. Mendeleyev Institute for Metrology

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Abstract

The prototype of setup that implements of KX-gamma coincidences technique developed at the ionizing radiation department of the D.I. Mendeleyev Institute for Metrology is described. The basic element of setup is MKGB-01 spectrometer-radiometer (STC "Radek"). MKGB-01 units - NaI(Tl) crystals of 100 µm x 40 mm and 80 x 80 mm are used for photon radiation detection. The standard set of “CAMAC” units is used as a secondary electronic part. Original design multi-channel counting system is used for counting rate measurements. The main metrological characteristics of the setup model were determined: a long-term stability, background, dead time, resolution time.

Using the described setup the electron-capture radionuclides ^{54}Mn, ^{57}Co, ^{65}Zn, ^{88}Y, ^{139}Ce activity was measured. The combined uncertainty (K=2) of activity was estimated in the range 0,5-1,0 %. Using KX-KX coincidences technique ^{125}I activity was measured with combined uncertainty of 1,0 % (K=2). The results obtained are in good coincidence with international comparisons results.

The main result of investigations is a conception of a new KX-gamma coincidence setup based of the MKGB-01 serial spectrometer-radiometer. A number of requirements for the secondary electronic units have been declared, a device for source positioning has been designed and an achievable uncertainty of the KX-gamma coincidence method has been evaluated. The supplementary investigation for determination of possibility to use the MKGB-01 spectrometer-radiometer for realization in beta-gamma coincidence technique.

Introduction

The photon sources based on ^{54}Mn, ^{57}Co, ^{65}Zn, ^{88}Y, ^{139}Ce and ^{125}I radionuclides are used for spectrometers calibration, applied in radio-ecological measurements. Moreover, ^{125}I based sources are applying in nuclear medicine. These radionuclide disintegrates via electron capture (1) which is accompanied by characteristic radiation from the K, L shells, etc., and the excited state of daughter atom without delay becomes stable with gamma radiation emission (accompanied by the conversion characteristic radiation). The positron emission can also be present.

The main problem is the activity measurement of radionuclides in covered sources, when it is impossible to use beta-gamma coincidence methods (2). X-ray and γ-radiation coincidence method is practically the only method to measure radionuclides activity directly in the covered source.

In our opinion, the most appropriate, is the coincidences of KX and γ-radiation. The detector with thin crystal and Be entrance window was used for KX-radiation, and detector with large crystal and Al entrance window for γ-radiation. The crystals design provides registration mainly only one type of radiation in each detector, and the entrance windows construction excludes registration of KX-KX and KX-LX coincidences. The ^{125}I activity was measured by KX-KX coincidences method using two thin crystals NaI(Tl) (3).

Materials and methods

STC «Radek» produces spectrometer-radiometer MKGB-01, equiped by several detecting units of different type. In the ionizing radiation department at the D. I. Mendeleyev Institute for Metrology a prototype of setup for realisation of KX–γ и KX-KX coincidences method based on this commercially available device has been constructed.

The detecting units based on NaI(Tl) crystals 100 mkm*40 mm and 80*80 mm respectively were used for registration of KX and γ-radiation. For realization of KX-KX coincidences method two NaI(Tl) crystals, 40 mm diameter and 100 mkm thickness were used. Background radiation was shield by using 10 mm thickness lead screen. The prototype block-diagram is shown on Fig.1.

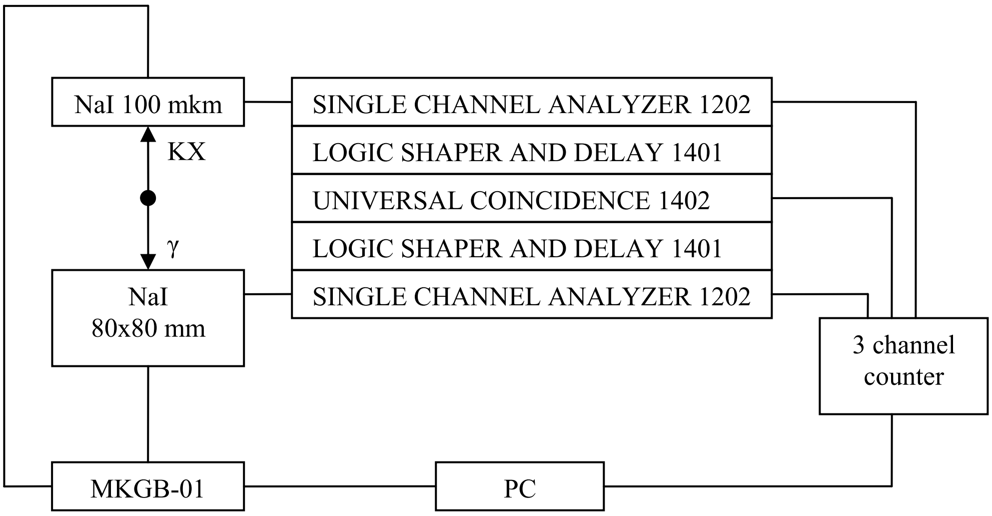

Fig.1. Prototype of setup block-diagram

The control of spectrometer operating modes (PMT high voltage, preamplifier gain factors and etc.) was carried out using the MKGB-01 firmware.

The registration system is created using set of CAMAC module and consist of single-channel analyzer, pulse shaper and coincidence unit. X-rays, gamma-rays and coincidence counting rates are measured using original design multichannel counting system.

Before making the measurements the main characteristics of the measurement system were determined. These characteristics are following:

Warming time is the time interval, after which the device readings satisfy the requirement:

$$\left| N_i - \bar{N} \right| \le 3 \cdot S(\bar{N})$$

Where N_i is the device reading of the i-th measurement; $\bar{N}$ is the mean value of the device readings calculated as a result of m measurements performed two hours after the device has been turned on; $S(\bar{N})$ is the standard deviation of the mean value calculated by equation:

$$S(\bar{N}) = \sqrt{\frac{\sum (Ni - \bar{N})}{\sum (m-1)}}$$

Reproducibility after turning off is the relative deviation of the device reading after it has been turned off and turned on again at the unchanging (invariable, constant, permanent) irradiation field:

$$R = \frac{\left| N_i - N_j \right|}{N_i} \cdot 100\% \,,$$

where N_i and N_j are the device readings before and after turning off.

Instability during the working day is the maximum deviation of detector readings from the mean value during the working day:

$$D = \frac{\max \left| N_i - \bar{N} \right|}{\bar{N}} \cdot 100\%$$

Temperature dependence of sensitivity is the variation of device readings when the environment temperature is changing :

$$k(t) = \frac{N(t) - N(t_o)}{t - t_o} \cdot 100\% \,,$$

where $N(t)$ is the device reading when the environment temperature is t°C; $N(t_o)$ is the device reading when the environment temperature is t_o°C.

"Dead"time of the devices was determined using the "two sources technique":

- gamma or X-ray source was placed at a fixed point to provide the device counting rate of about 10^4 s^{-1}, the counting rate N_1 was measured;

- device and source remained immovable, the other source was placed in a position providing to make the counting rate $N_{12} \sim 2N_1$, then the counting rate N_{12} was measured;

- counting rate N_2 with the first source moved out was measured.

The "dead" time value τ was calculated by equation :

$$\tau = \frac{1}{N_{12}} \{1 - \sqrt{\frac{(N_{12} - N_1)(N_{12} - N2)}{N_1 N_2}}\}$$

The following results were obtained for the described prototype.

For X-ray channel:

- warming time is less than 30 min;
- reproducibility after turning off does not exceed 0.1 %;
- instability during the working day does not exceed 0.1%;
- temperature dependence of the sensitivity is 0.05%/°C;
- "dead" time $\tau = 1.21 \pm 0.01$ μs for first KX-detector
- "dead" time $\tau = 1.44 \pm 0.02$ μs for second KX-detector

For gamma-ray channel:

- warming time is less than 30 min;
- reproducibility after turning off does not exceed 0.1 %;
- instability during the working day does not exceed 0.1%;
- temperature dependence of the sensitivity is 0.05%/°C;
- "dead" time $\tau = 2.42 \pm 0.02$ μs.

The coincidence system resolving time τ_p was measured using a pulse generator and is equal 0,514, 1,004 and 2,018 μs for 1402 unit modes «½», «1» and «2» respectively. These τ_p values provided the random coincidence contribution less than 10% of coincidence counting rate.

The final result - activity value was calculated according to Campion (4) and Brian (5) equations.

Measurement modes selection.

The setup operating modes were the follow:

- The source-detector distance in any cases should minimize of summary events probability.
- The KX-channel energy «window» is configured to register the KX-radiation only. In γ-channel the integral operating mode with lower threshold higher than KX-radiation energy is used. It is accepted, that the KX-radiation registration efficiency by γ-detector is close to zero.
- The γ-channel lower threshold for ^{54}Mn and ^{88}Y activity measuring is adjusted above 255 keV to avoid coincidence registration with back scattered radiation.
- The γ-channel lower threshold for ^{65}Zn activity measuring is adjusted above 511 keV to avoid coincidence registration with annihilation radiation.
- The γ-channel lower threshold for ^{57}Co and ^{139}Ce activity measuring is adjusted in energy 80 keV and 100 keV respectively.
- For measuring ^{125}I activity in both KX-channels, the integral operating mode with lower threshold higher than the radiation emission peak (≥15 keV) is used.

Derivation of measurement equations.

In general the measurement equations system for electron-capture radionuclide accompanied by internal conversion of γ–transition (^{125}I, for example), taking into account the summary events in each channel, will be follows:

$$\begin{cases} N_1 = N_0 \cdot (\varepsilon_{\gamma 1} I_\gamma + \varepsilon_{kx1}(I_{ec} + I_{ic}) - \varepsilon_{\gamma 1} I_\gamma \varepsilon_{kx1} I_{ec} - \varepsilon_{kx1}^2 I_{ec} I_{ic}) \\ N_2 = N_0 \cdot (\varepsilon_{\gamma 2} I_\gamma + \varepsilon_{kx2}(I_{ec} + I_{ic}) - \varepsilon_{\gamma 2} I_\gamma \varepsilon_{kx2} I_{ec} - \varepsilon_{kx2}^2 I_{ec} I_{ic}) \\ N_c = N_0 \cdot (\varepsilon_{\gamma 1} I_\gamma \varepsilon_{kx2} I_{ec} + \varepsilon_{\gamma 2} I_\gamma \varepsilon_{kx1} I_{ec} + 2\varepsilon_{kx1} I_{ec} \varepsilon_{kx2} I_{ic}) \end{cases} \quad (1)$$

Where: $N_1; N_2; N_c$ - pulse counting rates in γ-channel, KX channel and in coincidence channel, $\varepsilon_{\gamma 1}; \varepsilon_{\gamma 2}; \varepsilon_{kx1}; \varepsilon_{kx2}$ - efficiencies of γ and KX-detector to γ and KX-radiation; I_γ - emission probability of γ-radiation, $I_{ec} = P_k \omega_k$ - emission probability of KX-radiation due to electron capture, $I_{ic} = (\alpha_k \omega_k)/(1+\alpha_t)$ - emission probability of KX-radiation due to internal conversion. The coincidences with LX – radiation are not considered for reasons described above.

In conditions of summary events low probability ($\varepsilon_{\gamma 1}; \varepsilon_{\gamma 2}; \varepsilon_{kx1}; \varepsilon_{kx2} < 5\%$) the equations (1) are simplified to (2).

$$\begin{cases} N_1 = N_0 \cdot (\varepsilon_{\gamma 1} I_\gamma + \varepsilon_{kx1}(I_{ec} + I_{ic})) \\ N_2 = N_0 \cdot (\varepsilon_{\gamma 2} I_\gamma + \varepsilon_{kx2}(I_{ec} + I_{ic})) \\ N_c = N_0 \cdot (\varepsilon_{\gamma 1} I_\gamma \varepsilon_{kx2} I_{ec} + \varepsilon_{\gamma 2} I_\gamma \varepsilon_{kx1} I_{ec} + 2\varepsilon_{kx1} I_{ec} \varepsilon_{kx2} I_{ic}) \end{cases} \quad (2)$$

The contribution of γ-radiation to KX-detector $N_2^\gamma = N_0 \cdot \varepsilon_{\gamma 2} I_\gamma$ is defined using aluminium filters (1mm for ^{54}Mn, ^{57}Co and ^{65}Zn and 5mm for ^{88}Y) and copper filter (1 mm for ^{139}Ce).

Modifying the equations (2) and computing I_γ, I_{ec} and I_{ic} from the data (1), we obtain measurements equation for each radionuclide. The numerical value of corrections standard uncertainty is indicated in brackets.

For ^{54}Mn

$$\frac{N_1 \cdot (N_2 - N_2^\gamma)}{N_c} = N_0 \quad (3)$$

For ^{65}Zn 1115 keV γ-transition only is taken into account.

$$\frac{N_1 \cdot (N_2 - N_2^\gamma)}{N_c} = N_0 \cdot \frac{(a \cdot I_{ec}^a + b \cdot I_{ec}^b)}{I_{ec}^a} = N_0 \cdot 0.989(9) \quad (4)$$

Where: $a; b$ - probability of electron capture to the 2 and 0 level for ^{65}Zn,

$I_{ec}^a; I_{ec}^b$ - emission probability of KX-radiation due to of electron capture to the 2 and 0 level.

For ^{139}Ce

$$\frac{N_1 \cdot (N_2 - N_2^\gamma)}{N_c} = N_0 \cdot \frac{(I_{ec} + I_{ic})}{I_{ec}} = N_0 \cdot 1.239(5) \quad (5)$$

For ^{88}Y 898 keV and 1836 keV γ-transitions are taken into account.

$$\frac{N_1 \cdot (N_2 - N_2^{\gamma})}{N_c} = N_0 \cdot \frac{(a \cdot I_{\gamma}^{898} + X \cdot I_{\gamma}^{1836}) \cdot (a \cdot I_{ec}^{a} + b \cdot I_{ec}^{b})}{(a \cdot I_{ec}^{a} \cdot (I_{\gamma}^{898} + X \cdot I_{\gamma}^{1836}) + b \cdot I_{ec}^{b} \cdot X \cdot I_{\gamma}^{1836})} = N_0 \cdot 0{,}989(10) \quad (6)$$

Where: $a;b$ - electron capture probability to the 2 and 1 level for ^{88}Y, $I_{ec}^{a};I_{ec}^{b}$ - emission probability of KX-radiation due to electron capture to the 2 and 1 level, $I_{\gamma}^{898};I_{\gamma}^{1836}$ - γ-radiation emission probability with energies 898 keV and 1836 keV, $X = \varepsilon_{\gamma 1}^{898} / \varepsilon_{\gamma 1}^{1836}$ - ratio of γ-radiation registration efficiency with energy 898 keV to the γ-radiation registration efficiency with energy 1836 keV in the selected «window» (≥400 keV).

The value of X =1,00(1) was calculated by Monte-Carlo method (6) for selected distances source-detector. The maximum contribution of X in correction standard uncertainty in the right part of the equation (6) was equal 0,1%.

For ^{125}I

$$\frac{N_1 \cdot N_2}{N_c} = N_0 \cdot \frac{(K_1 \cdot I_{\gamma} + I_{ec} + I_{ic}) \cdot (K_2 \cdot I_{\gamma} + I_{ec} + I_{ic})}{(I_{\gamma} \cdot I_{ec} \cdot (K_1 + K_2) + 2 \cdot I_{ec} \cdot I_{ic})} = N_0 \cdot 2.0064(42) \qquad (7)$$

Where: $K_1 = \varepsilon_{\gamma 1} / \varepsilon_{kx1}; K_2 = \varepsilon_{\gamma 2} / \varepsilon_{kx2}$ - ratio of γ-radiation registration efficiency to the KX-radiation registration efficiency in each KX-detector in the selected energy «window» (≥15 keV).

The value of K_1=K_2 = 1,215(45) were calculated by Monte-Carlo method (6) for selected distances source-detector. The maximum contribution of K_1 and K_2 in correction standard uncertainty in the right part of the equation (7) was equal 0,25%.

For ^{57}Co 14,4, 122 и 136 keV γ-transitions are taken into account.

$$\frac{N_1 \cdot (N_2 - N_2^{\gamma})}{N_c} = N_0 \cdot \frac{(K \cdot a \cdot I_{\gamma}^{136} + b \cdot I_{\gamma}^{122}) \cdot (I_{ec} + a \cdot I_{ic}^{136} + b \cdot I_{ic}^{122} + b \cdot I_{ic}^{14})}{(K \cdot a \cdot I_{ec} \cdot I_{\gamma}^{136} + b \cdot I_{ec} \cdot I_{\gamma}^{122} + b \cdot I_{\gamma}^{122} \cdot I_{ic}^{14})} = N_0 \cdot 1.016(9)$$

(7)

Where: $a;b$ - transition probability from 2 to 0 and from 2 to 1 level for ^{57}Co, $I_{\gamma}^{122};I_{\gamma}^{136}$ - γ-radiation emission probability with energies 122 keV and 136 keV, $I_{ic}^{14};I_{ic}^{122};I_{ic}^{136}$ - KX-radiation emission probability due to γ-radiation internal conversion with energies 14,4 keV, 122 keV and 136 keV, $K = \varepsilon_{\gamma 1}^{136} / \varepsilon_{\gamma 1}^{122}$ - ratio of γ-radiation registration efficiency with energy 136 keV to the γ-radiation registration efficiency with energy 122 keV in the selected energy «window» (≥80 keV).

The value of K =1,060(4) was calculated by Monte-Carlo method (6) for selected distances source-detector. The maximum contribution of K in correction standard uncertainty in the right part of the equation (9) was equal 0,2%

Results

The ^{54}Mn, ^{57}Co, ^{65}Zn, ^{88}Y and ^{139}Ce radionuclide activity in film covered sources OSGI type was measured using KX-γ coincidences method. The results obtained are presented in table 1. In the last column of table 1 the deflection of these results from the same obtained by national activity standard of Russia Federation. The ^{54}Mn №5123 and ^{57}Co №5171 sources are the witnesses of international comparisons (7,8). Other radionuclides activity measurements results was traced to the results of key comparisons (9,10,11).

Table 1. The results.

Nuclide	Source number	Date	KX-γ coincidences activity, Bq	Uc, k=2 %	Activity, measured on the standard, Bq	Uc, k=2 %	Deflection, %
^{54}Mn	5123	01.01.09	12 956	1.0	12 860	1.0	0.8
	41-06	01.01.09	36 363	1.0	36 462	2.0	-0.3
	384-08	01.01.09	72 186	1.1	72 451	2.0	-0.4
^{65}Zn	5201	01.01.09	5 593	2.1	5 524	2.0	1.2
	16-08	01.01.09	41 130	2.1	41 221	2.0	-0.2
	387-08	01.01.09	90 294	2.1	90 236	2.0	0.1
	350-07	01.01.09	171 258	2.2	171 755	2.0	-0.3
^{88}Y	15-08	01.01.09	14 004	2.3	14 172	2.0	-1.2
	24-06	01.01.09	38 236	2.3	38 549	2.0	-0.8
	469-08	01.01.10	97 031	2.3	98 369	2.0	-1.4
^{139}Ce	14-08	01.01.09	18 446	1.8	18 340	2.0	0.6
	47-06	01.01.09	67 000	1.8	66 007	2.0	1.5
	471-06	01.01.10	150 746	1.9	149 330	2.0	1.0
^{57}Co	5171	01.01.09	8 229	2.0	8 108	1.0	1.5
	01	01.01.09	205 224	2.1	206 731	2.0	-0.7

^{125}I activity was measured in 5 film covered sources, prepared from one solution with well known masses. A detailed description of sources preparation procedure is given in (3). The specific activity is calculated as the average value of 5 results. The result obtained is given in comparison with specific activity (table 2), measured on the national activity standard of Russia Federation with KX-γ coincidences method.

Table 2. ^{125}I Specific activity

Nuclide	Date	KX-γ coincidences specific activity, Bq/mg	Uc, k=2 %	Specific activity, measured on the standard, Bq/mg	Uc, k=2 %	Deflection, %
I-152	21.05.09	3084	2.0	3135	2.0	1.6

Discussion

Table 1 and 2 data analysis shows, results obtained using described prototype are in good agreement with national standard results. It means spectrometer-radiometer MKGB-01 can be used for activity measurements without preliminary calibration.

Moreover setup described makes it possible to measure radionuclides activity directly in mass-produced film covered sources (12).

On the basis of this investigations an original KX-γ coincidences setup construction is proposed (MKGB-01 units based primary converter, background shielding system, positioning device, secondary electronics).

Conclusions

A concept of creation of the setup, implements the KX-γ coincidences method, based on commercially available spectrometer-radiometer MKGB-01 is developed. The main requirements to the future KX-γ coincidences setup are formulated.

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