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# Methodical aspects and measuring set-up for investigation the ability to selective detection of different ionizing radiations by organic scintillators

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In this paper, we have discussed the technique for selective detection of different types of ionizing radiations. This technique is based on the discrimination procedure of particles by their scintillation pulse shape. We have studied the ability of organic crystalline and liquid scintillators to selective detection of neutron and γ-radiation. The results we have been obtained in this work give initial information for the quantitative analysis of the processes proceeding in side a particle track in contrast to earlier works when such a study, in fact, had qualitative character.

В работе рассматривается метод селективной регистрации различных типов ионизирующих излучений. Этот метод основывается на дискриминации частиц по форме сцинтилляционного импульса. Исследуется способность органических кристаллических и жидких сцинтилляторов к раздельной регистрации нейтронов и фотонов гамма-излучения. В отличие от результатов ранних исследований, носящих качественный характер, полученные в работе результаты дают исходную информацию для количественного анализа процессов, протекающих в треке ионизирующей частицы.

In real nuclear physics experiments, the detection of ionizing radiation always takes place in the presence of some background because the ionizing sources of only certain type of radiation, practically, do not exist. High-accuracy detection systems that mainly use scintillators are very sensitive to different types of ionizing radiations. As an example, in neutron measurements a scintillator detects both neutrons and accompanying background γ-radiation because the neutron detectors on the base of solid and liquid scintillators have a high efficiency of γ-radiation photon detection. Therefore, a highly accurate experiment is impossible without a technique of identification of ionizing radiations aimed at selective detection of different types of radiations.

The problem of selective detection of ionizing radiation, especially the problem of separation of the neutron spectrum and gamma background is very important in nuclear experiments. The most effective method for detection of fast neutrons is based on production in material the recoil protons. Therefore, the organic detectors with high containment of hydrogen are the most useful for fast neutrons spectroscopy. A selective ability of the scintillator used for detection of ionizing radiations is one of its major characteristics. Under conditions of mass production of scintillation crystals and detectors on their base, the method allowing during the short time to determine a selective ability of scintillation materials is required. This work is devoted to discussion of the methodical aspects of certification of organic scintillation detectors for applications in measurements with scintillation pulse-shape discrimination of radiations of different types.

Ionizing radiations generate in an organic scintillator two types of a luminescence referred to the prompt radioluminescence and the delayed radioluminescence. A formation of the fast component of the radioluminescence pulse essentially takes place outside the regions of particle tracks. It has an exponential decay with time constant about $10^{-9}$–$10^{-8}$ s. The process of scintillation pulse fast component formation proceeds within "the optical approximation" and practically does not depend on specific energy losses $dE/dx$ of a particle. The slow component of the scintillation pulse arises due to of triplet-triplet annihilation process that results in the delayed radioluminescence of a scintillator. These processes take place only in the regions of high activation density (e.g. a track of a particle) where the concentration of ionized and exited molecules is high and therefore the influence of $dE/dx$ on the slow component of scintillation pulse formation is of primary importance. Due to annihilation nature of such a process, hyperbolical functions describe the shape of slow component and it intensity depends on $dE/dx$. It is the physical base of the technique of particle identification by the radioluminescence pulse shape [1–5].

There are two ways to study the ability of organic scintillators to discriminate the ionizing radiation with different $dE/dx$. Firstly, it is the direct analysis of the radioluminescence pulse shape of organic scintillators, and secondly, it is the method of selective detection of events in real time [3–6].

The single-photon technique, proposed by L.M.Bollinger and G.E.Thomas, allows obtaining the precise shape of scintillation pulse and therefore to compare pulse shape parameters obtained in the measurements with radiations with different $dE/dx$. According to this technique, within some time interval the probability of a photon appearance in a scintillation pulse after excitation is measured. This technique possesses a series of unambiguous advantages. Among them an optimal the signal-to-noise ratio, a possibility to carry out the measurements both the fast component of a scintillation pulse, and the slow one, etc. Figure 1 presents the block diagram of the set-up for studying the time parameters of the radioluminescence pulse by the single-photon technique [3, 5, 6].

Let us describe the selective ability of a scintillator measured by the single-photon technique in as following [4, 5]:

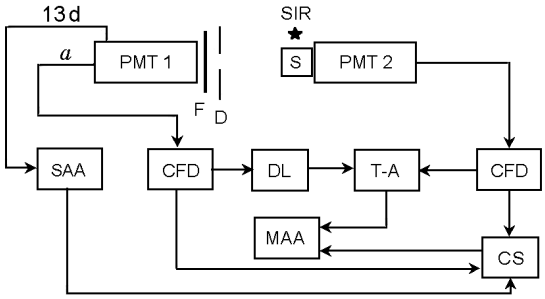


Fig. 1. Block diagrams of the set-up for measuring of time parameters of radioluminescence pulse by single-photon technique. Blocks of optical part of the set-up are following: S is a sample; PMT 1 and PMT 2 are photomultipliers; SIR is a source of ionizing radiation; D is a diaphragm, F is an optical filter. The units of electronics part of the set-up are the following: SAA is a single-channel amplitude analyzer; CFD — constant fraction discriminator; DL is a delay line; T-A is a time-to-amplitude converter; MAA is multichannel amplitude analyzer; CS is a coincidence circuit; $a$ is the anode output of photomultiplier tubes; 13d is the output from the 13th dynode of PMT 1.

$$\chi = \frac{\sigma_n}{\sigma_\gamma}, \qquad (1)$$

where $\sigma_n$ and $\sigma_\gamma$ are the ratios between the number of events in slow and fast component of a radioluminescence pulse for neutron and $\gamma$-excitation, respectively. The analysis of a scintillation pulse shape, obtained by the single-photon technique, provides the determination of the contribution of the radioluminescence pulse slow component with high degree of accuracy. However, the accumulation of the necessary statistics in single-photon measurements of a scintillation pulse shape, and especially its slow component may be a long procedure. It may continue from a few hours to a few days. It is the reason why the single-photon technique is not quite acceptable for a certification of scintillators. Besides that, it does not allow to obtain the information concerning the energy spectrum of scintillations [4].

Another way for studying the selective ability of scintillators is an identification of particles by their pulse shape. We used the method of selective detection of particles based on an immediate comparison of

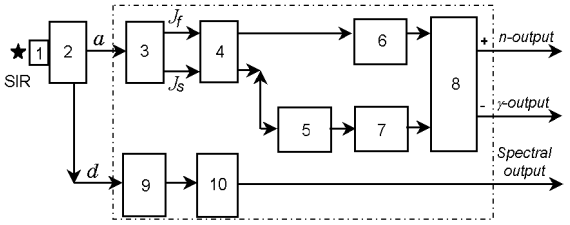


Fig. 2. Block-diagram of set-up for selective detection the ionizing radiations with different $dE/dx$. SIR is a source of ionizing radiation; *1* is a scintillator; *2* is a photomultiplier; *3* — the discrimination circuit that separates "fast" and "slow" signals; *4* is an integrator; 5 is an inverter; *6*, *7* are amplifiers with adjusted gain; *8* is a summer; *9* is a charge sensitive amplifier; *10* is a pulse former amplifier; *a* is a photomultiplier anode output; *d* is the output of the next to the last dynode of a photomultiplier; *n*-output is the neutron spectrum control output; γ-output is γ spectrum control output; spectral output is the spectrometric output.

charges in slow and fast component of the scintillation pulse taken with different statistical weights [5]. Figure 2 demonstrates the set-up that realizes this method. The signal from anode output of the photomultiplier is transmitted to the electronic scheme that separates a charge in time intervals from 0 to 50 ns after an excitation and from 50 to 2 μs, i.e. the time of fast and the slow signals, respectively. Electronics integrates the charge accumulated in the interval from 0 to 50 ns and obtains the $J_f$ value, as well as integrates the charge accumulated in the interval from 50 ns to 2 μs and inverts to obtain $J_s$ value. The signals proportioned to $J_f$ and $J_s$ are summed in such a way that a detection of scintillation pulses initiated by neutrons results in a formation of the positive pulse on the summer output, whereas a detection of photons of gamma radiation results in a formation of the negative pulse. The digital signal appears on separate "neutron" or "gamma" output according to polarity of the previous signal. The charge sensitive amplifier and then the pulse former amplifier form the signal from the next to the last dynode of a photomultiplier for amplitude analysis. This instrument may works in measurements with separation of combined spectra of α- and β-scintillations, α- and γ-scintillations, etc. using the same principle of discrimination analysis.

According to [3], a quality of the discrimination procedure based on the abovementioned technique depends on time interval $t_f$ of "complete" decay of the fast component of the radioluminescence pulse. Single-photon measurements of the radioluminescence pulse shape give the $t_f$-value [4].

To describe the selective ability of scintillators measured by the discrimination procedure introduce the coefficient:

$$K_n = \theta_n / \theta_\gamma, \qquad (2)$$

where $\theta_{n(\gamma)}$ is the number of pulses formed on neutron (gamma) control output of the set-up (excluding noise and false operation pulses) normalized by the neutron (gamma) flux for certain radiative source [3].

The digital frequency meter of Ch3-33 type measures the number of pulses formed on neutron (gamma) control output of the set-up. The spectrometric signal goes to analogue-to-digital converter (ADC), and loads in a memory file if the control signal (neutron or gamma) allows writing.

Let us introduce, according to [4] parameter $\xi_{i,j}$ which characterizes integral effect of a quenching as a function of $dE/dx$ of ionizing radiation, i.e. the total energy losses for both the fast component and for the slow component of the radioluminescence pulse:

$$\xi_{i,j} = \left(\frac{N_i}{E_i}\right) \cdot \left(\frac{N_j}{E_j}\right)^{(-1)}, \qquad (3)$$

where $N_i$ and $N_j$ are the number of scintillation photons for $i$- and $j$-types of the excitation with energy $E_i$ and $E_j$, respectively, i.e. the relation between the light signals intensities normalized to the correspondent energy of the excitation. The calculation of the parameters $\xi_{n,\gamma}$ and $\xi_{\alpha,\gamma}$ provides the information about the processes of scintillation pulse formation under irradiations with different $dE/dx$.

We have used the radionuclide sources of $^{252}$Cf and $^{239}$Pu–Be in our measurements. They are the sources both of photons of γ-radiation and of fast neutrons. Therefore, the relation between the γ-radiation photon flux and the neutron flux is equal to the relation between the corresponding activities of γ-radiation and neutron of these sources. The ratio between the number of γ-radiation photons and the number of fast neutrons is equal to 8.46 for the act of spontaneous fission $^{252}$Cf radionuclide

Table 1. Comparative values of coefficients $\chi$ (1) and $K_n$ (2) for organic scintillators irradiated by $^{239}$Pu–Be source.

| | Scintillator | $\chi$, % | $K_n$, % |
|---|---|---|---|
| 1 | Liquid scintillator on the base of a vaseline/α-methylnaphthalene mixture | 14.7 | 14.4 |
| 2 | Stilbene single crystal | 24.0 | 24.0 |

Table 2. Results of the measurements of parameter $K_n$ (2) for organic single crystals

| | Scintillator | Dimensions, mm | $K_n$, % | |
|---|---|---|---|---|
| | | | $^{252}$Cf | $^{239}$Pu–Be |
| 1 | Stilbene single crystal | 80×50 | 34.8 | 42.6 |
| 2 | -//- | 80×50 | 38.0 | 42.9 |
| 3 | -//- | 50×50 | 48.6 | 48.9 |
| 4 | -//- | 50×50 | 25.4 | 32.4 |
| 5 | -//- | 50×40 | 20.8 | 31.8 |
| 6 | -//- | 50×40 | 15.2 | 27.0 |
| 7 | -//- | 40×40 | 23.0 | 34.8 |
| 8 | -//- | 30×30 | 16.1 | 22.5 |
| 9 | -//- | 25×20 | 15.2 | 22.2 |
| 10 | -//- | 25×20 | 16.4 | 21.6 |
| 11 | -//- | 25×18 | 9.6 | 22.8 |
| 12 | -//- | 40×14 | 41.1 | 46.8 |
| 13 | -//- | 40×14 | 25.9 | 35.4 |
| 14 | -//- | 40×14 | 19.6 | 32.4 |
| 15 | -//- | 40×14 | 46.7 | 42.3 |
| 16 | *p*-Terphenyl single crystal | 50×50 | 39.9 | 34.5 |
| 17 | -//- | 50×40 | 19.6 | 21.3 |
| 18 | -//- | 40×14 | 15.9 | 12.0 |
| 19 | *p*-Terphenyl polycrystal | 40×3 | 4.7 | 6.3 |

source. For the Pu–Be source the proportion between the number of γ-radiation photons and the number of fast neutrons is equal to 3 [7, 8]. These characteristics of the sources have been considered in calculations by Eq.(2).

The results of the measurements with the radionuclide source of $^{241}$Am allow to calculate the $\xi_{\alpha,\gamma}$-value (3). It is the source of alpha ($E_\alpha = 5.4$ MeV) and gamma ($E_\gamma = 59.5$ keV) radiations. To separate α- and γ-scintillations the following measurements were run. Firstly, we have measured the scintillation spectrum when the source of $^{241}$Am placed directly on the sample. Then, we run the same measurements but with a sheet of paper between the sample and the source. The high-energy peak disappeared. At last, we have irradiated the sample by the source of γ-radiation on the base of $^{241}$Am (it had the special film that absorbs α-radiation). In this case, we observed the peak in the region of amplitude spectrum that corresponded to the previous measurement with a sheet of paper. It has allowed us to identify the high-energy peak as α-scintillations, and the low-energy peak as γ-scintillations.

Table 3. Results of the measurements of parameter $K_n$ (2) for a series of organic liquid scintillators irradiated by $^{239}$Pu–Be source

| | Scintillator base | $K_n$, % |
|---|---|---|
| 1 | Diisopropylnaphthalene | 6.0 |
| 2 | Benzyl benzoate/naphthalene mixture | 13.0 |
| 3 | Vaseline/α-methylnaphthalene mixture | 11.2 |

We calculated the values of $\chi$ (1) and $K_n$ (2) for stilbene single crystal and for liquid scintillator on the base of the mixture of vaseline and α-methylnaphthalene. Table 1 gives the results of these calculations and illustrates the correlation between the values $\chi$ and $K_n$. It shows that the discussed technique is quite correct for selective spectral measurements of an ionizing radiation in the presence of background.

We studied the ability of organic scintillators to selective detection of ionizing radiations. Table 2 presents the values of coefficient $K_N$ for stilbene and *p*-terpenyl organic scintillators and detectors on their base. Scintillators were of different diameters (from 25 to 80 mm) and thickness (from 3 to 50 mm). Table 3 shows the results of a studying of the selective ability for the series of liquid scintillators. These

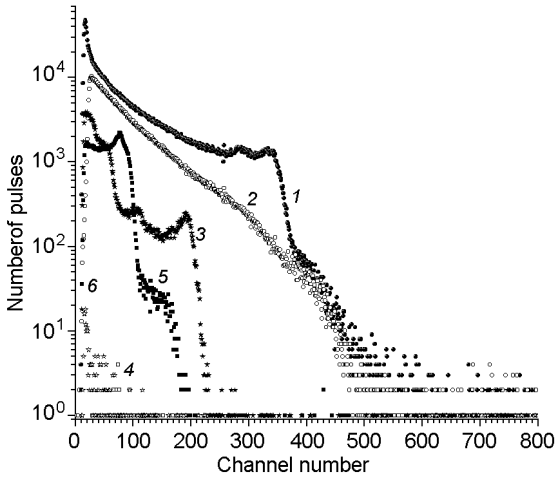


Fig. 3. Amplitude spectra of 50×50 mm stilbene detector irradiated by the sources of ionizing radiation: $^{239}$Pu–Be (curves *1* and *2*), $^{232}$Th (curves *3* and *4*) and $^{60}$Co (curves *5* and *6*). Curves *1*, *3* and *5* have been measured without the neutron control signal, curves *2*, *4* and *6* with the neutron control signal.

liquid scintillators filled the 30×40 mm$^2$ glass cells. They were bubbled by argon during 10 minutes. After that, the cells were sealed. The side surface of cells had light-reflective enamel VL-548.

We measured the amplitude spectra of 50×50 mm$^2$ stilbene detector, and used the data obtained to calculate the parameter $\xi_{n,\gamma}$. A $^{239}$Pu–Be fast neutron source gave gamma radiation as well. Radionuclides of $^{60}$Co and $^{232}$Th gave set of gamma lines in referent experiments. Fig. 3 shows these spectra obtained on the set up (see Fig. 2). Curve *1* presents the total neutron and gamma spectrum of $^{239}$Pu–Be (discrimination is not used). Curve 2 presents the neutron spectrum of $^{239}$Pu–Be (control from neutron output). Curves *3* and *5* show the amplitude gamma-spectra of the sources $^{60}$Co and $^{232}$Th measured without the neutron control signal, and curves *4* and *6* show amplitude gamma-spectra of the sources $^{60}$Co and $^{232}$Th with the neutron control signal, respectively. A negligible number of pulses for curves *4* and *6* confirm that discussed discrimination procedure is enough effective. Using the number of channel in which generated by the neutron of energy $E_n$ the recoil protons form the "step" [2, 4, 6], we calibrated the measured spectrum in the scale of neutron energies. Fig. 4 demonstrates the relation between the number of such a channel of the multichannel analyzer and the neutron energy for measured stilbene single crystal irradiated by $^{239}$Pu–Be source [2, 7, 8]. To obtained the neutron and gamma energies those correspond to the same amplitude in scintillation spectra we selected the channel number of Compton edge in the amplitude spectra obtained for $^{60}$Co and $^{232}$Th sources of gamma-radiation (see Fig. 3), and the neutron energy, which according to calibration curve for neutron energies (see Fig. 4), corresponds to the same channel. After that, we calculated parameter $\xi_{n,\gamma}$ in accord with Equation (3). Compton edge energies $E_{comp}$ for sources with energies $E_\gamma$ obtained according to well-known relations (see [2]). Table 4 shows these results.

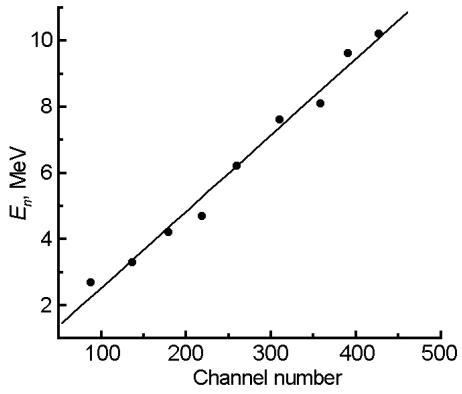


Fig. 4. The number of a channel of the analyzer versus the neutron energy for measured amplitude spectrum of 50×50 mm stilbene single crystal irradiated by $^{239}$Pu–Be. The solid circles are experimental data, the solid line is a linear approximation $Y = A + B \cdot X$, where $A$ = 0.2 MeV; $B$ = 0.023 MeV/channel.

Table 4. Results of the calculations of parameter $\xi_{n,\gamma}$ (3) for 50×50 mm stilbene single crystal

| $E_\gamma$, MeV | $E_{comp}$, MeV | Channel number | $E_n$, MeV | $\xi_{n,\gamma}$ |
|---|---|---|---|---|
| 1.173 | 0.963 | 83 | 2.125 | 0.453 |
| 1.332 | 1.117 | 97 | 2.447 | 0.456 |
| 2.614 | 2.381 | 202 | 4.846 | 0.491 |

The calculated value of $\xi_{\alpha,\gamma}$ for stilbene single crystal irradiated by $^{241}$Am is equal to 0.05. The $\xi_{n,\gamma}$ values (see Table 4) are about 0.4–0.5. So, the values obtained, i.e. $\xi_{n,\gamma}$ and $\xi_{\alpha,\gamma}$, differ by an order of magnitude. This result shows the difference in influence of particle energy $E$ and $dE/dx$ on the quenching processes in it track [4–6].

Thus, designed set-up and technique gives a possibility to obtain the energy spectra of recoil protons generated by fast neutrons excluding the influence of accompanied gamma-radiation. The maximum energy of recoil protons gives information about neutron energies, and the ratio between the number of pulses on the neutron and gamma control outputs allows estimating the ability of scintillators to selective determination of neutron and gamma-radiation. The $K_n$-value increases with growth of sample dimensions (Table 2). It means that for energy range discussed a dependence of scintillation efficiency from sample dimensions is stronger for neutrons than for photons of gamma radiation. The comparison of the results obtained for stilbene and *p*-terphenyl single crystals indicates that detectors on the base of stilbene scintillators more effective for the modern tasks of neutron spectrometry. Notwithstanding that, it is necessary to take into account for practical use, that scintillators on the base of *p*-terphenyl have better strength and operating characteristics. The values of parameter $\xi_{n,\gamma}$ have been obtained for different energies of ionizing radiation. It gives very important initial information for the quantitative analysis of the processes proceeding inside a particle track in contrast to earlier works when such a study, practically, had qualitative character [4–6].

### *References*


1. T.A.King, R.Voltz, *Proc. Royal Soc.*, **A286**, 424 (1966).
2. J.B.Birks, The Theory and Practice of Scintillation Counting, Pergamon Press, London (1964).
3. N.Z.Galunov, Yu.A.Tsirlin, Some Aspects of the Radioluminescence Pulse Formation Process in Organic Single Crystals and Chemically Pure Liquid Scintillation Solutions and the Ability thereof to Selective Detection of Ionizing Radiation, NIITECHIM, Moscow (1987) [in Russian].
4. N.Z.Galunov, V.P.Seminozhenko, Theory and Applications of the Radioluminescence of Organic Condensed Media, Naukova Dumka, Kiev (1997) [in Russian].
5. N.Z.Galunov, E.V.Martynenko, in: Abstr. of the Int. Conf. on Crystal Materials, Institute for Single Crystals, Kharkov (2005), p.89.
6. N.Z.Galunov, B.V.Grinyov, O.A.Tarasenko, E.V.Martynenko, *J. Korean Assoc. Radiat. Protect.*, **30**, 85 (2005).
7. I.V.Goryachev, Ya.I.Kovalevatov, L.A.Trykov, Integral Experiments in the Problem of the Ionizing Radiation Transfer, Energoatomizdat, Moscow (1985) [in Russian].
8. Certificate No.481 from July 23, 1999. Metrological Attestation of Closed Californium Source of Neutron Radiation, type INK-2, Passport No.074, National Research-and-Production Association "Metrologia" (1999) [in Russian].


## Методичні аспекти та апаратура для дослідження здатності органічних сцинтиляторів до селективної реєстрації різних типів іонізуючих випромінювань

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У роботі розглядається метод селективної реєстрації різних типів іонізуючих випромінювань. Метод грунтується на дискримінації частинок за формою сцинтиляційного імпульсу. Досліджується здатність органічних кристалічних і рідких сцинтиляторів до роздільної реєстрації нейтронів та фотонів гама-випромінювання. На відміну від результатів попередніх досліджень, які мають якісний характер, отримані у роботі результати надають вихідну інформацію для кількісного аналізу процесів, що відбуваються у треку іонізуючої частинки.