A TECHNIQUE FOR THE SELECTIVE DETECTION OF NEUTRONS IN GEOLOGICAL APPLICATIONS

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We present a new, combined technique for the detection of fast and thermal neutrons in the presence of accompanying gamma radiation, for geological and radioecological applications. To separate the slow neutron pulses and fast gamma pulses of organic scintillators, we used the method of particle identification by radioluminescence pulse shape. The device has been tested by the combined neutron and gamma radiation sources ^{252}Cf and ^{239}Pu-Be, and by a 14.2 MeV D-T source of fast neutrons both in the laboratory, and under real, field conditions as applied at selected sites of the European Union.

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1. INTRODUCTION

Environmental site surveys require accurate, in depth analysis of potential contaminants in the sub-surface. Currently this is done by sampling boreholes, sending samples for analysis, and then interpreting the data, which often are time consuming. There is a need for a logging tool, which can rapidly provide this data so that site assessment can be carried out quickly and efficiently.

Geological research and mineral investigations on site also involve time delays between sampling rocks and getting analytical data. Therefore, the analysis has to be made on site, thus guiding exploration and research in a more effective manner. Currently no such device is available commercially. A number of electric logging tools exist for water and soil analysis, but these are not multi-purpose, and the number of parameters, which can be measured is limited. Previous neutron logging devices required a radioactive source, which made them impracticable due to handling and storage safety considerations, and only gave qualitative analyses of parameters.

In neutron measurements, a scintillator detects both neutrons and accompanying background gamma radiation because the neutron detectors based on solid and liquid scintillators have a high efficiency of gamma radiation detection. Therefore, a highly accurate experiment is impossible without using a technique with the 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 the production in organic material of recoil protons. Therefore, organic detectors with a high content of hydrogen are the most useful for fast neutron spectroscopy.

This work is devoted to the new applied technique of geological research based on using a combined scintillation method in the detection of fast and thermal neutrons in the presence of accompanying, induced gamma radiation. We have mounted the circuitry and controlling software with photomultiplier and other components on a unit that allows operating the detector system in the research phase of this work.

2. THEORY

Ionizing radiation in an organic scintillator generates two types of luminescence referred to as the prompt radioluminescence and the delayed radioluminescence. The formation of the fast component of the radioluminescence pulse essentially takes place outside the region of particle tracks. It has an exponential decay with time constant of about $10^{-9} - 10^{-8}$ s [1]. The process of scintillation pulse fast component formation takes place within "the optical approximation" and practically does not depend on the specific energy losses dE/dx of a particle. The slow component of the scintillation pulse arises due to a triplet-triplet annihilation process that results in the delayed radioluminescence of the scintillator. These processes take place only in the regions of high activation density (e.g. a track of a particle) where

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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 its intensity depends on dE/dx [2]. It is the physical base of the technique of particle identification by radioluminescence pulse shape.

3. EXPERIMENT

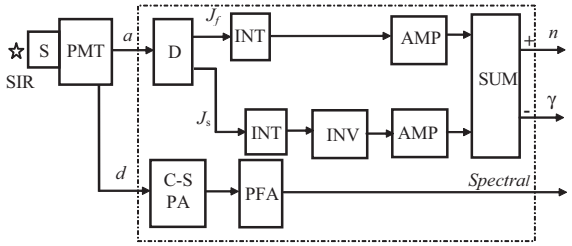

Fig.1. *Block diagram of the set-up for selective detection the ionizing radiation with different dE/dx: SIR is a source of ionizing radiation; S is a Ø40 mm×40 mm stilbene scintillator; PMT is a 9272B ETL photomultiplier tube (a is the anode output and d is the next to the last dynode output); D is a discriminator; INT is an integrator; INV is an inverter; AMP are amplifiers with adjusted gain; SUM is a summer; C-S PA is a charge-sensitive preamplifier; PFA is a pulse former amplifier; n is the neutron spectrum control output; γ is gamma spectrum control output; spectral output is the spectrometric output*

Fig.1 illustrates the measuring set-up that is used for above-mentioned method. We used Ø(40×40) mm stilbene detector optically coupled with a 52 mm type 9272B Electron Tubes Ltd. photomultiplier. The signal from the anode output of the photomultiplier is transmitted to the electronic part that separates a charge in time intervals from 0 to 50 ns after an excitation and from 50 to 2000 ns, i.e. the time of the fast and the slow signals, respectively. The 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 to 2000 ns and inverts to obtain J_s-value. The signals proportional to J_f and J_s are summed in such a way that the detection of scintillation pulses initiated by neutrons results in the formation of the positive pulse on the summation output, whereas the detection of photons of gamma radiation results in the formation of the negative pulse. The digital signal appears on a separate "neutron" or "gamma" output according to the polarity of the previous signal. The charge-sensitive preamplifier, followed by the pulse former amplifier, forms the signal from the next to the last dynode of a photomultiplier for amplitude analysis. This instrument can work for 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], the quality of the discrimination procedure based on the technique described above 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.

In our experiments, we used a high output neutron emitting tube that generates 14.2 MeV monoenergetic neutrons by deuterium-tritium (D-T) reaction. The frequency of bursts is 20 Hz and this source emits 10^8 neutrons per second in 4π-geometry. Accelerating potential for deuterons was equal to 120 keV that allows obtaining practically isotropic diagram of neutrons in a burst [4]. Additionally, for laboratory tests we used a Pu-Be radionuclide source with neutron flux equal to $\sim 10^5$ neutrons per second, and a radionuclide ^{252}Cf as fission type source with neutron flux $\sim 10^4$ neutrons per second.

The dominant type of radiation determines the choice of scintillation material. Our previous investigations indicate that detectors based on stilbene scintillators are the most effective for the required tasks of neutron spectrometry. Notwithstanding that, it is necessary to take into account, that scintillators based on *p*-terphenyl have better strength and operating characteristics [3].

For the detection of thermal neutrons we used a Ø(20×3) mm detector on the basis of a LiI(Eu) crystal. It contains an element with a high value of thermal neutron absorption cross-section, e.g. ^{6}Li.

For penetrative gamma radiation, a material with high density is required. Characteristics of some inorganic materials usually used for the detection of photons of gamma radiation have been previously discussed (see [5], for example). Detectors based on NaI(Tl) crystals have the best characteristics for high resolution gamma-spectroscopy. The combination of two of above mentioned scintillation materials (NaI(Tl) and LiI(Eu), or stilbene and NaI(Tl)) allows us to detect simultaneously different types of ionizing radiation.

We have chosen a 25 mm type 9110FLA ETL photomultiplier for the detection of thermal neutron and a 52 mm type 9272B ETL photomultiplier for the detection of background gamma radiations. The signal d (see Fig.1) from the next to the last dynode output was formed for amplitude analysis according to above-mentioned procedure. In this case the anode (control) output a did not used.

4. RESULTS AND DISCUSSION

To test the proposed technique of selective detection neutron and gamma radiation (see Fig. 1) we have carried our a series of measurements of amplitude spectra from a Ø(40×40) mm stilbene detector irradiated by the combined neutron and gamma source (Pu-Be), the mono-energetic D-T neutron source and standard sources of gamma radiation ^{22}Na, ^{60}Co, ^{137}Cs, ^{152}Eu and ^{232}Th.

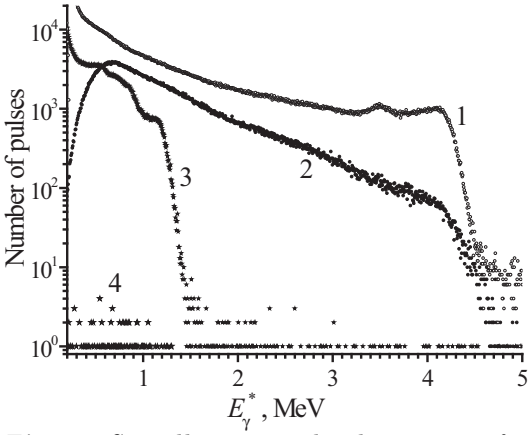

Fig.2. *Scintillation amplitude spectra of a Ø40 mm×40 mm stilbene detector irradiated by the sources of ionizing radiation: ^{239}Pu-Be (curves 1 and 2), ^{252}Eu (curves 3 and 4). Curves 1 and 3 have been measured without the neutron control signal, but curves 2 and 4 have been measured with the neutron control signal*

Curve 1 on Fig.2 shows the total recoil protons and recoil electrons spectrum of stilbene irradiated by Pu-Be (discrimination is not used). Curve 2 shows the proton spectrum of Pu-Be (control from neutron output). Curve 3 shows the recoil electrons spectrum of stilbene irradiated by ^{252}Eu measured without the neutron control signal, and curve 4 shows this spectrum measured with the neutron control signal, respectively. A negligible number of pulses for curve 4 confirm that discrimination procedure discussed here is clearly effective. The X-axis is calibrated in the scale of scintillation amplitudes obtained from photons of gamma radiation of energy E_γ^*. Compton edge energies E_{comp} for sources with energies E_γ has been obtained according to a well-known formula [6].

The following expression can used to regenerate the neutron spectrum with energies E_n [7] in scale of scintillation amplitudes:

$$\varphi(E_n) = -\frac{E_n}{\varepsilon(E_n)} \times \left.\frac{du(E_p)}{dE_p}\right|_{E_p=E_n}, \quad (1)$$

where $u(E_p)$ is a recoil proton spectrum (measured for $E_p \le E_n$), $\varepsilon(E_p)$ is an efficiency of detection of different energy neutrons. Expression (1) does not take into account the non-linearity of scintillation response to recoil protons [5]. Expression (1) gives the neutron spectra $\varphi(E_n)$ in non-linear E_n scale. Therefore the results obtained still hold the information concerning features of scintillator response, as well as the information about "specific quenching". The calculation (1) is inapplicable to the case of study a spectrum of unknown neutron source, because the non- linearity of scintillation response has to be considered in such a case. It allows obtaining the scintillation response when the neutron source with know spectrum is used [7]. Therefore we used a radionuclide ^{239}Pu-Be source. It spectrum is thoroughly studied. See i.e. the theoretical calculations of the energy spectrum of Pu-Be neutron source [8], experimental works in which such a source was tested by stilbene scintillators and nuclear emulsions [9, 10], etc. The peaks 1, 2, 3, 4, 5, 6, 7, 8, 9, 10 and 11 on Fig. 3 correspond to well-known energies of neutrons (0.85, 2.0, 3.1, 4.2, 4.9, 6.4, 6.7, 7.3, 7.9, 8.6 and 9.7 MeV respectively [7-10]) emitted by a ^{239}Pu-Be source.

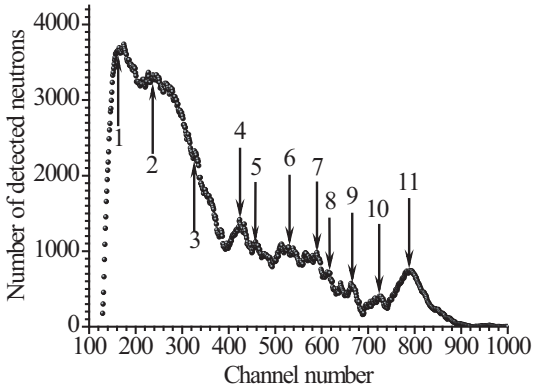

Fig.3. *Neutron spectra reconstructed from the recoil proton spectra (see Fig.2, curve 2) of ^{239}Pu-Be source for a Ø(40×40) mm stilbene detector*

We investigated the spectrum of scintillation amplitudes of stilbene detector irradiated by a D-T mono-energetic neutron source using the above-mentioned procedure of n/γ-discrimination. Fig.4 shows the results of these measurements.

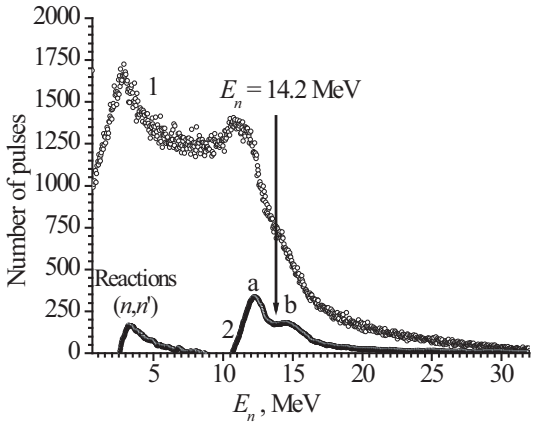

Fig.4. *Scintillation amplitude spectra of a Ø(40×40) mm stilbene detector irradiated by a D-T neutron source: curve 1 is the scintillation spectrum of recoil protons, curve 2 is derived spectrum of 14.2 MeV neutrons*

Curve 1 on Fig.4 shows the experimental scintillation spectrum of recoil protons generated by fast neutrons in the detector. Curve 2 is the neutron spectrum of a D-T source obtained by the procedure of numerical differentiation followed by smoothing of experimental data of curve 1 (see expression 1). The high-energy peak of curve 2 is the reconstructed neutron signal from 14.2 MeV neutrons. One can see it splitting on two peaks, *a* and *b*. The amplitude relationship between these peaks is about 1.2 that is in a good agreement with the value of anisotropy of stil-

bene signals obtained during an excitation along and perpendicular to crystallographic plane *ab* [3]. In our experiments, the measuring system was excited along crystallographic plane *ab* and this type of excitation was more probable. The neutrons were scattered by the neutron shielding excited the crystal perpendicular to crystallographic plane *ab*.

Fig.5 shows the scintillation spectra obtained during ^{6}LiI(Eu) detectors irradiation by thermal neutrons (the sources of fast neutrons in paraffin sphere) and photons of gamma radiation from a ^{137}Cs radionuclide source. Neutrons generate monochromatic α-particles and therefore an α-scintillation spectrum is obtained. It is accompanied by a γ-spectrum of background radiation. Amplitude discrimination is enough to separate these two spectra, because for thin ^{6}Li(Eu) crystals a γ-scintillation spectrum is narrow and located in the range of low amplitudes.

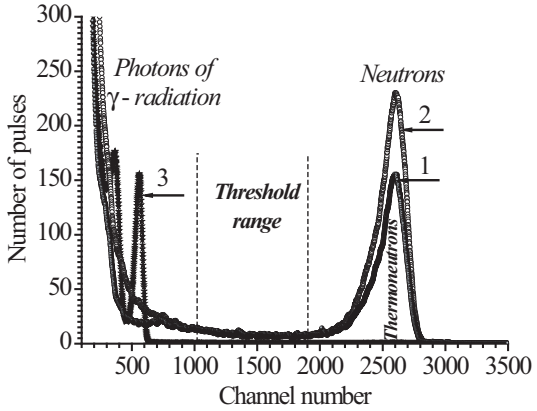

Fig.5. *Scintillation amplitude spectra from a Ø20 mm×3 mm LiI(Eu) detector irradiated by neutron sources have been introduced inside moderator (a paraffin sphere 15 cm in diameter): ^{239}Pu-Be (curve 1), ^{252}Cf (curve 2) and irradiated by a source of photons of gamma radiation ^{137}Cs (curve 3)*

5. CONCLUSIONS

We have tested and optimized the set of devices for fast and thermal neutron detection that can be used as the basis for measuring a range of parameters including hydrocarbons, heavy metals and other elements in both geological and environmental applications. The technique of discrimination of ionizing radiation by their pulse shape allows the separate detection and with high degree of accuracy both of thermal neutrons scattering from the material under investigation and secondary photons of gamma radiation. These photons are generated in reactions of inelastic scattering of fast neutrons or radiative capturing of thermal neutrons, and give information about the chemical composition of a material [5].

It should be noted that in geological applications the content of water (one of the most effective moderator of neutrons in nature), difference in density, porosity of soil, and some other factors might have a significant effect on the results of any measurements, especially in the case of quantitative analysis. So, for complex systems, data processing and interpretation is a separate task, and therefore has been discussed separately.

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АППАРАТУРА ДЛЯ РАЗДЕЛЬНОЙ РЕГИСТРАЦИИ НЕЙТРОНОВ ДЛЯ ГЕОЛОГИЧЕСКИХ ПРИМЕНЕНИЙ

Д.Х. Бэйкер, Н.З. Галунов, В.Г. Костин, Е.В. Мартыненко, О.А. Тарасенко

Изложены основные принципы и аппаратура для регистрации быстрых и тепловых нейтронов для современных задач геологии и радиоэкологии. Раздельная регистрация медленных нейтронных импульсов и быстрых импульсов гамма-излучения органических сцинтилляторов осуществлялась путем дискриминации ионизирующего излучения по форме сцинтилляционного импульса. Аттестация аппаратуры производилась с помощью источников ^{252}Cf и ^{239}Pu-Be , а также D-T источника быстрых нейтронов с энергией 14.2 МэВ, как в лабораторных условиях, так и в реальных условиях ее применения в ряде стран Европейского Союза.

АПАРАТУРА ДЛЯ РОЗДІЛЬНОЇ РЕЄСТРАЦІЇ НЕЙТРОНІВ ДЛЯ ГЕОЛОГІЧНИХ ЗАСТОСУВАНЬ

Д. Х. Бейкер, М.З. Галунов, В.Г. Костін, Є.В. Мартиненко, О.А. Тарасенко

Розглянуті основні принципи та апаратура для реєстрації швидких і теплових нейтронів для сучасних задач геології й радіоекології. Роздільна реєстрація повільних нейтронних імпульсів і швидких імпульсів гама-випромінювання органічних сцинтиляторів здійснювалася шляхом дискримінації іонізуючого випромінювання за формою сцинтиляційного імпульсу. Атестація апаратури проводилася за допомогою джерел ^{252}Cf та ^{239}Pu-Be , а також D-T джерела швидких нейтронів з енергією 14.2 МеВ, як у лабораторних умовах, так і в реальних умовах її застосування в ряді країн Європейського Союзу.