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Hybrid scintillators on the basis of ammonium salicylate and ammonium dihydrogen phosphate for neutrons detection

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Scintillation properties of hybrid scintillators on the basis of ammonium salicylate and ammonium dihydrogen phosphate obtained using different methods for registration of fast neutrons and gamma-radiation have been investigated. Sensitiveness to fast neutrons of scintillation hybrid crystals ADP:NH_4Sal is proportional to the concentration of ammonium salicylate in the matrix of ADP. The highest neutron sensitiveness is shown by co-doped hybrid scintillation single crystals ADP:NH_4Sal/Tl.

Исследованы сцинтилляционные свойства гибридных сцинтилляторов на основе салицилата аммония и дигидрофосфата аммония для регистрации быстрых нейтронов и гамма-излучения. Сцинтилляторы получены различными способами. Чувствительность к быстрым нейтронам сцинтилляционных гибридных кристаллов ADP:NH_4Sal пропорциональна концентрации салицилата аммония в матрице ADP. Наибольшую чувствительность к нейтронам проявляют соактивированные гибридные сцинтилляционные монокристаллы ADP:NH_4Sal/Tl.

1. Introduction

Today in devices for fast neutrons registration in mixed (n/γ)-fields the well known organic scintillation crystals stilbene, anthracene, paraterphenyl and plastic scintillators (polystyrene) are mainly used. Interaction of neutrons with organic scintillator material causes the appearance of recoil protons and light flash. However these scintillators react similarly on γ-quanta which almost always accompany neutrons in nuclear transformations and present in large content in the mixed radiation fields [1]. Moreover, organic single crystals have high cost and limited accessibility. For development of new generation of fast neutron detectors the new, effective, cheap and accessible scintillation materials are required.

Organic single crystals of ammonium salicylate NH_4Sal are promising material samples for fast neutron registration [2]. They exceed their inorganic analogues in sensitiveness to fast neutrons however they are very hygroscopic and have low mechanical strength. Along with this it is known that thallium doped ammonium dihydrogen phosphate crystals reveal the selective sensitiveness to fast neutrons in the mixed (n/γ)-radiation flow [3]. The crystals NH_4Sal and ADP are grown from the water solutions under temperature decrease or solvent evaporation. In comparison with NH_4Sal ADP crystals are less hygroscopic and their growth technology is well-developed.

In the present work the results of investigation of scintillation properties of hybrid scintillators on the basis of ammonium salicylate and ammonium dihydrogen phosphate

obtained by different methods for registration of fast neutrons and gamma-radiation are presented.

2. Experimental

The samples for investigation have been obtained:

— by dispergating into an organosilicone matrix or compressing in the view of plates both microcrystalline powder of ammonium salicylate NH_4Sal and co-crystallized powder of dihydrogen phosphate and salicylate of ammonium ADP–NH_4Sal;

— by growing of single crystals:

• single crystals of ammonium salicylate NH_4Sal;

• single crystals of ammonium dihydrogen phosphate doped by ammonium salicylate – ADP:NH_4Sal;

• single crystals of ammonium dihydrogen phosphate co-doped by ammonium salicylate and thallium — ADP:NH_4Sal/Tl.

The organosilicone matrix on the basis of oligomethylvinylsiloxane has been vulcanized by addition polymerization reaction. The scintillation crystalline powder has been obtained by cocrystallization from the mixed solution of ADP–NH_4Sal. The powder consisted of microcrystals of ammonium salicylate and dihydrogen phosphate/salicylate of ammonium with the size of up to 1 mm.

Pressed samples have been obtained by hot pressing at the temperatures of 80–100°C.

Crystals have been grown by the temperature decrease from the water solutions of ammonium salicylate and ammonium dihydrogen phosphate with addition of NH_4Sal.

3. Measurements and results

The sample sensitiveness to neutrons of plutonium-beryllium source with activity of $5 \cdot 10^6$ neutron/s has been investigated. Measurements have been conducted by the multiplier phototube Hamamatsu R7056 and by multichannel impulse analyzer. The lower threshold of registration was 5 keV at the scale of NaI(Tl) that allows to cut off impulses caused by the radiation running from (Pu–Be)-source [4]. The measurement scheme is presented in Fig. 1. The sample neutron sensitiveness has been estimated according to the velocity of impulse counting of the neutrons scaled to the neutron flow and the sample area [5].

Fig. 2 presents amplitude spectra at irradiation of the crystals by neutrons (Pu–Be source) (2), γ-quanta (^{60}Co) (1) and background (3). A neutron spectrum does not have express photopeak because a power spectrum of neutrons of Pu–Be source is continual. γ-Spectrum and the background are located in the area of the small channels and are not overlapped with the area of the neutron spectrum.

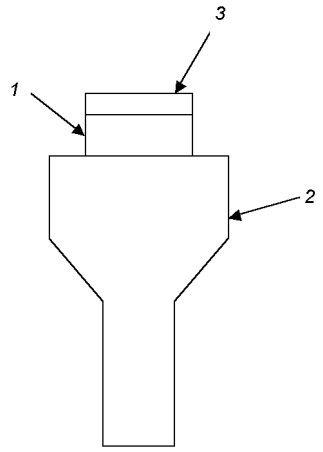

Fig. 1. Scheme of the experiment performance: *1* — sample under investigation; *2* — multiplier phototube; *3* — radioactive source.

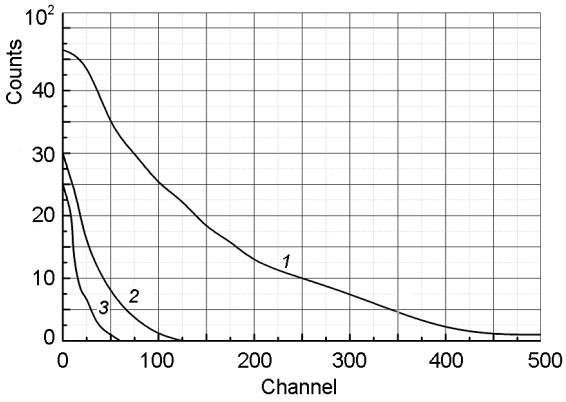

Fig. 2. Neutron amplitude spectra (Pu–Be)-source (*1*), γ-quanta (^{60}Co) (*2*) and background (*3*).

The mechanism of interaction of neutrons (having a few MeV of energy) with the doped ADP crystal consists in elastic scattering of neutrons on the hydrogen nuclei with formation of recoil protons. The hydrogen content in the ADP crystals is $\approx 10^{21}$ cm^{-3}. As a result of *np*-scattering in anionic groups $(H_2PO_4)^-$ hydrogen vacancies appear and hole center $(HPO_4)^-$ is formed. Formation of the autolocalized hole is accompanied by electron capture on the activator. Subsequent electron and hole recombination results in scintillation.

The values of neutron sensitiveness for the samples obtained by the different methods are presented in the Table. The percentage of ammonium salicylate is indicated in the mixed solution of ADP–NH_4Sal.

As it can be seen from the Table the neutron sensitiveness of hybrid scintillators depends on the method of the samples production. Single crystals of ammonium salicylate shown the highest neutron sensitiveness. In the hybrid single crystal samples ADP:NH_4Sal the sensitiveness decreases along with an increase of the content of ammonium salicylate in the solution that apparently is connected with association of salicylate-ions in solutions, for example, with formation of ionic pairs [6]. As a consequence, the coefficient of distribution of ammonium salicylate decreases with the increase of its concentration in the solution.

For comparison of activator influence on the neutron sensitiveness of doped ADP crystals the crystals co-doped by ammonium salicylate and thallium were grown (1 wt.% of each component in the solution). As it can be seen, the neutron sensitiveness of single crystals ADP:NH_4Sal/Tl is a few higher than the one of crystals doped only by ammonium salicylate (at the level of pressing NH_4Sal).

The sensitiveness of organosilicone composite materials and pressings obtained from the crystalline powder of ammonium salicylate does not differ significantly. Organosilicone composite materials and pressings obtained from the co-crystallized powder of dihydrogen phosphate and salicylate of ammonium show the sensitiveness increase in correlation with an increase of ammonium salicylate content in the samples. It is easy to understand because the cocrystallized powder is a mechanical mixture of the initial components, but undoped ADP does not possess scintillation properties.

Table. Sensitiveness of hybrid scintillators on the basis of salicylate and dihydrogen phosphate of ammonium to neutrons from (Pu–Be) source ($E > 0.75$ MeV)

Scintillator material	Sensitiveness 10^{-4}, imp·n^{-1}·cm^2
Polystyrene + 1.5 % *n*-terphenyl + 0.01 % POPOP (standard sample)	250
NH_4Sal single crystal	220
NH_4Sal organosilicone composite	170
NH_4Sal pressing	180
ADP + 1 % NH_4Sal single crystal	160
ADP + 12 % NH_4Sal single crystal	70
ADP + 25 % NH_4Sal single crystal	20
ADP + 1 % NH_4Sal + 1 % thallium single crystal	180
ADP + 25 % NH_4Sal organosilicone composite	32
ADP + 90 % NH_4Sal organosilicone composite	97
ADP + 25 % NH_4Sal Pressing	92
ADP + 90 % NH_4Sal Pressing	163

4. Conclusion

In spite of the small amount of the built-in salicylate-ions the hybrid crystals demonstrate the presence of scintillation properties depending on the concentration of C_{Sal}. Codoped hybrid scintillation single crystals ADP:NH_4Sal/Tl are characterized by the highest neutron sensitiveness which is comparable with the sensitiveness of the organic single crystals of ammonium salicylate.

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Гібридні сцинтилятори на основі саліцилату амонію та дигідрофосфату амонію для детектування нейтронів

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Досліджено сцинтиляційні властивості гібридних сцинтиляторів на основі саліцилату амонію та дигідрофосфату амонію для реєстрації швидких нейтронів та гамма-випромінювання. Сцинтилятори виготовлено різними способами. Чутливість до швидких нейтронів сцинтиляційних гібридних кристалів ADP:NH_4Sal пропорційна концентрації саліцилату амонію у матриці ADP. Найбільшу чутливість до нейтронів мають соактивовані гибридні сцинтиляційні монокристали ADP:NH_4Sal/Tl.