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**SEARCH FOR NEW SCINTILLATORS  
FOR HIGH ENERGY RESOLUTION  
ELECTROMAGNETIC CALORIMETERS**

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### Abstract

Britvich G.I. et al. Search for New Scintillators for High Energy Resolution Electromagnetic Calorimeters: IHEP Preprint 98-11. – Protvino, 1998. – p. 17, figs. 8, tables 2, refs.: 36.

An analysis of some opportunities to create radiation resistant heterogeneous EM-calorimeters with an energy resolution of about  $\sigma/E \simeq 4 \div 5 \%/ \sqrt{E}$  is given in this paper. Some investigation results of scintillation and radiation characteristics for thin molded plates and new heavy scintillators based on the polystyrene and containing metalloorganic additives are presented. The radiation resistance of thin molded scintillator plates of about 1.1 mm thick containing 2% pTP + 0.05% POPOP has reached a level of about 1.5÷2. Mrad.

### Аннотация

Бритвич Г.И. и др. Исследование новых сцинтилляторов для ЭМ-калориметров с высоким энергетическим разрешением: Препринт ИФВЭ 98-11. – Протвино, 1998. – 17 с., 8 рис., 2 табл., библиогр.: 36.

В работе проанализированы некоторые возможности создания радиационно-стойких гетерогенных ЭМ-калориметров с энергетическим разрешением около  $\sigma/E \simeq 4 \div 5 \%/ \sqrt{E}$ . Приведены результаты исследований сцинтилляционных и радиационных характеристик тонких литевых пластин и новых тяжелых сцинтилляторов на основе полистирола, содержащих металлоорганические добавки. Радиационная стойкость тонких литевых сцинтилляционных пластин толщиной 1.1 мм, содержащих 2% pTP + 0.05% POPOP, достигла значений около 1.5÷2. Мрад.

## Introduction

With the construction of a new generation of high energy particle accelerators (UNK, LHC, RICH, etc.) a great interest to the development of electromagnetic (EM) calorimeters with high radiation resistance (more than several Mrad/yr) [1] and energy resolution of about  $4\div 5 \text{ \%}/\sqrt{E}$  [2,3] has evoked. Homogeneous calorimeters on the basis of heavy crystals most closely satisfy the above conditions [2]. However, they are expensive and have other essential limitations. An appreciable attention is drawn to heterogeneous EM-calorimeters with fiber-optic readout system, so-called sampling-calorimeters, with the purpose of improving their energy resolutions to the levels of homogeneous ones. So, it was experimentally shown that for some heterogeneous EM-calorimeters of the "Shashlyk" and "Bayan" types with fine sampling fractions, their energy resolutions reached a level of about  $\sigma/E \simeq 7 \text{ \%}/\sqrt{E}$  [4,5].

The energy resolution of EM-calorimeters is determined by the influence of a variety of fluctuation sources and their values depend on calorimeters design, types of used photoreceivers, the amount of energy deposited by incoming particles in scintillator layers, etc. In a general case, the energy resolution dependence for EM-calorimeters  $\sigma/E$  as a function of the incoming particles energy  $E$  can be expressed by the formula

$$\sigma/E = a/\sqrt{E} \oplus b/E \oplus c, \quad (1)$$

where  $\oplus$  is the square-law addition,  $a$  is the stochastic term determined by the fluctuation of the deposited energy,  $b$  is the "noise" term determined by the noise of the used photoreceiver,  $c$  is a constant term determined by the light output nonuniformity from scintillator plates, the light output losses in scintillator plates and wavelength shifting (WLS) fibers (if used), the instability of photoreceivers, etc. It was experimentally shown that the constant term value for some sampling EM-calorimeters of the "Bayan" type can be easily made of  $c < 0.5 \text{ \%}$  [4]. Note that  $b$  values give appreciable contributions to expression (1) only with the use of silicon photodiodes in EM-calorimeters. Thus, the energy resolution for well-designed sampling EM-calorimeters [4,5,6] is basically determined by their stochastic terms  $a$ .

Thus, the energy resolution of well-designed EM-calorimeters is determined by the fluctuation of the number of different shower particles. Their contribution to expression (1) can be approximately presented in the following way:  $\sigma/E \propto \sqrt{t_{sc}/f_s}$ , where  $f_s$  is the sampling fraction and  $t_{sc}$  is the scintillator plate thickness expressed in radiation lengths. There exists two ways for the reduction of the sampling fluctuation contribution, i.e. the fact that the fraction of the shower energy deposited in the calorimeter scintillators varies from event to event, to the above expression [7]. The first one is connected with the increase of the so-called sampling fraction  $f_s$ . The second way is connected with the increase of the number of independent sampling layers, i.e. with the decrease of  $t_{sc}$ , for a fixed sampling fraction [7]. Note that most of the well-designed EM-calorimeters with  $\sigma/E < 10\%\sqrt{E}$  had sampling fractions better than 0.2.

**Sampling fraction increase.** In this case the amount of scintillation materials in the calorimeter volume where the showers develop is increased. It means that with the use of ordinary scintillator plates, the  $f_r$  increase leads to an increase of the scintillator plates total thickness which results in the increase of radiation length  $X_o$  and Molier radius  $R_m$  of EM-calorimeters. These factors worsen the ability of such EM-calorimeters work under high rates and secondary particles multiplicity conditions. However, there exists an opportunity for  $f_r$  improvement connected with the use of plastic scintillators containing heavy metals (heavy scintillators). Our simulation of characteristics of some sampling EM-calorimeters [4,6] containing heavy scintillators with the help of the GEANT program [8] showed that in the process of transition from ordinary plastic scintillators with  $X_o \simeq 45$  cm to heavy ones with  $X_o \simeq 15 \div 20$  cm, the energy resolution might be improved 1.3  $\div$  1.5 times and would reach a level of  $\sigma/E \simeq 4.5 \div 5.5 \%/ \sqrt{E}$ .

It is well known that heavy plastic scintillators are widely used for the registration efficiency improvement of soft  $\gamma$ -quanta and neutrons [9,10,11]. Recently, the use of heavy polystyrene (PS) scintillators doped with metalloorganic additives at high concentration levels was proposed to improve the registration efficiency of soft  $\gamma$ -quanta in EM-showers and, hence, the energy resolution [12] of modern EM-calorimeters [4,6].

**Sampling frequency increase.** Some opportunities for the energy resolution improvement of EM-calorimeters are provided by the method of increasing the number of independent layers. It means a decrease both in the absorber  $t_a$  and scintillator  $t_{sc}$  plates thicknesses with the preservation, at the same time, the sampling fraction at a fixed level [7]. However, this method appreciably enhances the cost of EM-calorimeters modules caused by the cost increase of all plates (and not in the last turn — scintillator ones), a complication of calorimeters design, etc. It should be noticed here that the improvement of EM-calorimeters energy resolution via increasing their sampling frequency also has its limitations. We consider that the development of a simple design of EM-calorimeters of the "Bayan" type [4] can be a solution for the above problems. The injection into the mold technique for mass production of thin scintillator plates [13], both ordinary and heavy, may be used. Note that the decrease of scintillator plates thicknesses may result also in the improvement of their radiation resistance [14].

This work is devoted to the study of characteristics of thin scintillator plates fabricated by the injection into the mold technique as well as bulk-polymerized scintillator samples

with metalloorganic additives containing tin, lead, bismuth and mercury. The opportunity to apply heavy scintillation materials for the energy resolution improvement of modern EM-calorimeters is also considered.

## 1. Samples fabrication and measurement techniques

### 1.1. Samples fabrication by bulk-polymerization technique

The monomer was de inhibited through a column and distilled in vacuum. Then, the monomer was placed in cylindrical glass ampoules and doped with metalloorganic additives containing such heavy metals as Sn, Pb, Bi and Hg together with sets of scintillation additives 2 % pTP + 0.05 % POPOP or 1÷6 % PPO + 0.05 % POPOP. The chemical formulae of the used additives and maxima of their wavelengths emission  $\lambda_{em}$  are indicated in Appendix. The glass ampoules with the solutions of about  $\simeq 100 \text{ cm}^3$  were sealed off after pumping-out the air during 10÷15 min and polymerized in a polysilicone bath at a temperature of about  $T \simeq 180^\circ\text{C}$  during 24 hours [15].

Preliminary measurements showed that the process of darkening caused by the decomposition of metalloorganic additives at this temperature of polymerization (the temperature of decomposition for TPB additive is about  $T \simeq 160^\circ\text{C}$ ) was observed in the samples containing TPB additive. However, the use of activators, which lower the temperature of polymerization to  $T \simeq 60^\circ\text{C}$ , for example DAK, makes it possible to fabricate heavy scintillators with good optical properties containing this additive as well as other ones with low decomposition temperatures. The decomposition temperatures of other metalloorganic additives TPT, TPL and TPM appeared to be reasonably high, i.e.  $T > 180^\circ\text{C}$ . This provided the samples on their base of good optical properties. Note that the bulk-polymerization technique insures the production of plastic materials with the highest level of the light attenuation lengths of  $70 < l_o < 240 \text{ cm}$  for scintillators with  $\lambda_{em} \simeq 410 \div 430 \text{ nm}$  provided the temperature stable additives are used.

### 1.2. Scintillators fabrication by the injection into the mold technique

Except for the bulk-polymerization there exists a highly productive technology of plastic scintillators fabrication by the injection into the mold technique [13]. According to it, the fabrication process is performed with the use of injection machines, which provide an opportunity for the production of scintillator plates of various sizes and configurations. The granulated polystyrene of 115 PSM trade mark with the granule size of about  $\sim 5 \text{ mm}$  and a set of scintillation additives 2% pTP + 0.05% POPOP were thoroughly mixed. Then the mixture was loaded into an injection machine and heated to  $T \simeq 220 \div 240^\circ\text{C}$ . Appropriate quantities of the melt under a pressure of about 30÷80 MPa were injected into an injection mould where the plates were formed during 1÷2 min [13]. The sizes variation of thus manufactured plates did not exceed  $\pm 0.1 \text{ mm}$ .

### 1.3. Measurement of new scintillators light outputs

The 5 mm thick disks with a diameter of about 30 mm were cut out of the polymerized blocks. The scintillation in samples was excited by electrons from a radioactive source of  $^{90}\text{Sr}+^{90}\text{Y}$  with an activity of about  $10^7$  Bk. The sample and radioactive source were placed on the entrance window of a photomultiplier (PMT) FEU-110 which had a multialkali photocathode with an averaged quantum efficiency of about  $p_m \simeq 19\%$  in a wavelengths interval  $\lambda = 410 \div 490$  nm [16].

The samples light outputs were determined on the base of photocurrent measurements in comparison with that from a standard crystal anthracene with  $\lambda_{em}=447$  nm. The accuracy of these characteristics was determined on the basis of repeated measurements and did not exceed  $\pm 5\%$ .

On the other hand, for many detectors of ionizing radiation the main parameter which determines their applicability is the value of photon yield, i.e.  $w$ -energy that is necessary for the formation of a emission photon. For example, in photoemultions a similar value is about  $w \simeq 100$  eV/grain, for gas-discharged detectors —  $w \simeq 35$  eV/electron-ion pair, in silicon detectors —  $w \simeq 3.75$  eV/electron-hole pair [17].

It is well known [4] that in order for the scintillator materials not to appreciably influence the energy resolution of EM-calorimeters, the used scintillator plates should have  $w \leq 250$  eV and light attenuation lengths  $l_o \geq 6 \div 8$  cm.

In this work we also measured  $w$ -energy for new scintillators. The  $w$  value is determined from the formula

$$w = \frac{\Delta E}{n_{ph}}, \quad (2)$$

where  $\Delta E$  is the energy deposited by the ionizing radiation in the scintillator,  $n_{ph}$  is the number of photons in the scintillation burst. Here  $n_{ph}$  is determined from the formula

$$n_{ph} = \frac{n_{ph.e}}{p_m \kappa}, \quad (3)$$

where  $n_{ph.e}$  is the number of photoelectrons registered by the photoreceiver,  $\kappa$  is the light collection coefficient (the ratio of the number photons past through the receiver to the total number of photons in the scintillation burst),  $p_m$  is the averaged quantum efficiency of the photoreceiver for the scintillator emission spectrum with  $S(\lambda)$ . Here  $p_m$  is determined by the expression

$$p_m = \frac{\int p(\lambda)S(\lambda)d\lambda}{\int S(\lambda)d\lambda}, \quad (4)$$

where  $p(\lambda)$  is the quantum sensitivity of the photoreceiver.

A PMT FEU-143 with the known value of the quantum efficiency  $p$  as a function of the wavelength  $\lambda$  [18] was used as a photoreceiver. The internal conversion electrons from radioactive sources of  $^{57}\text{Co}$ ,  $^{137}\text{Cs}$  and  $^{207}\text{Bi}$  were used as the sources of irradiations. These radioactive sources provided us with a set of the deposited energy  $\Delta E$  values of 114.2; 481, 561, 622.6, 975 and 1047 keV.

#### 1.4. Light attenuation lengths measurements of new scintillators

The measurement of light attenuation lengths  $l_o$  in scintillator plates was carried out in accordance with the following technique. The scintillator plates with the sizes of  $400 \times 50 \times 1.1 \text{ mm}^3$  covered with aluminized Maylar were placed in a light-tight box. The plates sides of  $50 \times 1.1 \text{ mm}^2$  were coupled to a PMT FEU-110. The PMP photocurrent was measured. The scintillator plates were excited with a  $^{106}\text{Ru}$  radioactive source having an activity of about  $10^7$  Bk. In order to reduce the irradiation intensity, a collimator of  $30 \times 3 \text{ mm}^2$  was used. The collimator could be moved along the scintillator plates and stopped in a required position  $l$ . Results of the light attenuation measurements were fitted by the function

$$I(l) = I_o \exp(-l/l_o), \quad (5)$$

where  $I_o$  is the initial light intensity,  $I$  is the amount of light reached the photoreceiver,  $l_o$  is the light attenuation length and  $l$  is the thickness of the absorbing layer. Equation (4) corresponds to the well known law of Lambert-Beer. The averaged light attenuation lengths  $l_o$  were determined in an interval  $2.5 < l < 15$  cm. The accuracy of  $l_o$  measurements was about  $\pm 7\%$ .

#### 1.5. Decay time measurements

The characteristic scintillation curves were measured by the single photoelectrons counting technique [19]. PMTs FEU-143 had semitransparent SbCs photocathodes and their first dynodes had  $A^3B^5$  (gallium phosphide) covering. Such PMP were used both in the "Start" and "Stop" channels. The PMTs dividers were specially adjusted with the purpose of providing the best single-electron peak resolutions. So, in the "Start" and "Stop" channels the full widths at the half of its height for single-electron peaks were about 57 % and 53 %, correspondingly.

Monoenergetic electrons from a radioactive source  $^{207}\text{Bi}$  placed in a thin film were used for the samples excitation. Amplitude compensating shapers (both in the "Start" and "Stop" channels) with thresholds of sensitivity at a level of  $\sim 0.1 A_{max}$  were used. Here  $A_{max}$  was the maximum amplitude of single-electron pulses. Differential discriminators were also used in the same channels in order to register events near the maximum of the energy deposition in the scintillators, i.e.  $0.9 \div 1.1 A_{max}$ .

The time resolution of the above experimental setup was about 0.5 ns. Calibrated cable lines were used for the graduation of our setup time scale. The accuracy of the decay time measurements  $\tau$  was about  $\pm 5\%$ . Note that our experimental setup did not allow us to measure the decay times  $\tau \geq 1000$  ns.

#### 1.6. Radiation resistance tests

The radiation resistance of our scintillator plates was investigated in a flux of  $\gamma$ -quanta with the energy of  $E_\gamma = 662$  keV from radioactive sources of  $^{137}\text{Cs}$  with a dose rate of about 5.2 rad/s (or 150 Mrad/yr) in air at  $T \simeq 20^\circ\text{C}$ . The accuracy of the dose determination did not exceed  $\pm 10\%$ . Taking into account the prolonged time ( $\sim 10$  yr) of

scintillators operation under the irradiation in EM-calorimeters, it is clear that the study of the samples relative light output  $I/I_o$  just after the end of irradiation (in 20÷30 min) as well as in  $t_{rec}=23$  days of recovery in air [15,21,20] became an essential part of our radiation properties investigation. For thin scintillator plates with thickness  $h=1.1$  mm, the relative light output from points in a range  $2.5 < l < 15$  cm was measured.

## 2. Experimental results

### 2.1. Scintillation and radiation properties of new heavy scintillators

Some results of the light output measurements for ordinary and new heavy PS-scintillators are summarized in Table 1. At the beginning of Table 1 scintillation and radiation characteristics for some standard PS-scintillators are presented. The light output of a standard scintillator 1 is about 45 % in comparison with the crystal anthracene. Note that the maximum concentration of TPL and TPT which can be introduced in bulk-polymerized PS-scintillators are about 7.5÷10 % and 15 %, respectively.

**Table 1.** Scintillation and radiation properties of new heavy PS-scintillators prepared by the bulk-polymerization technique.

N	Scintillator	$I_o$ , %	$I/I_o$ , 3.4 Mrad	$I/I_o$ , 10 Mrad	$I/I_o$ , 23 days rec.
1	2% pTP + 0.05% POPOP	45.0	0.51	0.24	0.53
2	2% PPO + 0.05% POPOP	39.6	0.48	0.21	0.46
3	2% pTP + 0.05% POPOP + 5.0% BPM	18.9	0.67	0.40	0.55
4	2% pTP + 0.05% POPOP + 5.0% TPT	29.3	0.50	0.30	0.51
5	2% pTP + 0.05% POPOP + 15.% TPT	9.0	0.34	0.19	0.49
6	2% pTP + 0.05% POPOP + 2.5% TPL	32.4	0.35	0.21	0.57
7	2% pTP + 0.05% POPOP + 5.0% TPL	24.8	0.55	0.31	0.54
8	2% pTP + 0.05% POPOP + 7.5% TPL	16.7	0.18	0.10	0.43
9	1% PPO + 0.05% POPOP + 7.5% TPL	15.3	0.31	0.14	0.42
10	3% PPO + 0.05% POPOP + 7.5% TPL	21.6	0.36	0.22	0.47
11	6% PPO + 0.05% POPOP + 7.5% TPL	32.4	0.40	0.18	0.45

As is clear from Table 1, our heavy PS-scintillators 3÷8 showed lower levels of their light outputs  $I_o$  in comparison with the standard scintillators (without metalloorganic additives), i.e. all our metalloorganic additives are scintillation quenchers. So, the increase of the additive concentrations (TPL and TPT)  $> 5$  % results in an appreciable loss of heavy scintillators light output. However, the increase of a primary scintillation additive (PPO) concentration in samples 10-11 to levels  $\simeq 3\div 6$  % results in an increase of the scintillators light outputs which are close to the value of sample 2, i.e. without metalloorganic additives. It is obvious that in order to increase the light output of heavy scintillators, one should also use such primary additives that can be solved in the monomer

with a concentration  $>5\%$ . Besides, the high concentration of some primary additives also improves the radiation resistance of plastic scintillators [15,20].

The introduction of metalloorganic additives results in a change of the relative light output  $I/I_o$  of new heavy scintillators measured just after the end of irradiation in comparison with the values for the samples prepared without the above additives. Taking into account the recovery processes, levels of the radiation resistance of our heavy scintillators are comparative with those of samples 1-2. These experimental results are in a good agreement with the results of work [12]. However, a more detailed conclusion can be only made after an investigation of the radiation resistance of prolonged samples of new scintillators at dose rates that are comparable with those expected at physical setups.

The introduction of metalloorganic additives by the bulk-polymerization technique is not an unique method of heavy scintillators production. The injection into the mold technique allows one to introduce TPL additive in PS-scintillators with the concentration  $1.5\div 2$  time higher over the bulk-polymerized ones. So, we prepared optically transparent samples of heavy scintillators with the sizes  $400\times 50\times 1.1\text{ mm}^3$ , density of  $\rho = 1.072\text{ g/cm}^3$  and a TPL concentration of about  $15\%$  (equivalent to  $\text{Pb} = 4.07\%$  by weight). The radiation length of such scintillators reached a level of about  $X_o \simeq 27\text{ cm}$ . It is obvious that with the help of this technique, the TPT concentration can reach a level of about  $30\%$ , i.e. the radiation length will be near  $X_o \simeq 20\text{ cm}$ . It is close enough to the above determined levels of  $X_o = 15\div 20\text{ cm}$ . Note that this technique also permits the concentrations of primary additives, for example pTP, PPO, etc., in plastic scintillators to be increased.

Fig. 1 presents some experimental and calculated dependencies of the linear absorption coefficients  $\mu$  as functions of the energy of  $\gamma$ -quanta in a range  $10\text{ keV} < E_\gamma < 200\text{ keV}$  for ordinary and heavy molded PS-scintillator plates. For comparison, a similar characteristic for heavy polyvinyltoluene BC-452 scintillators [10] is presented in Fig. 1. The calculated curve  $\mu$  (in Fig. 1) for ordinary PS-scintillator was taken from work [10] and the curve for the heavy scintillator plate was calculated on the basis of the formula from work [22]

$$\mu = \rho \sum_i \mu_{m,i} \omega_i, \quad (6)$$

where  $\rho$  is the media density,  $\mu_{m,i}$  is the mass attenuation coefficient of  $i$  component having the mass fraction  $\omega_i$ . Fig. 2 shows the relative absorption efficiencies of  $\gamma$ -quanta from a  $^{238}\text{Pu}$  radioactive source. The  $\gamma$ -quanta pass through three thin scintillator plates ( $h = 1.1\text{ mm}$ ) with TPL additive at a concentration of about  $15\%$  and for the same number of ordinary plates, i.e. without TPL additive. The experimental results show that our heavy scintillators most effectively absorb  $\gamma$ -quanta in an energy region  $10\div 150\text{ keV}$ , i.e just where the maximum of the energy deposition for the soft  $\gamma$ -quanta of EM-showers occur. Note that due to higher densities of our heavy scintillators, the light output from EM-calorimeters should be  $1.4\div 1.7$  times higher in comparison with the use of a similar light output plastic scintillator, but without metalloorganic additives.

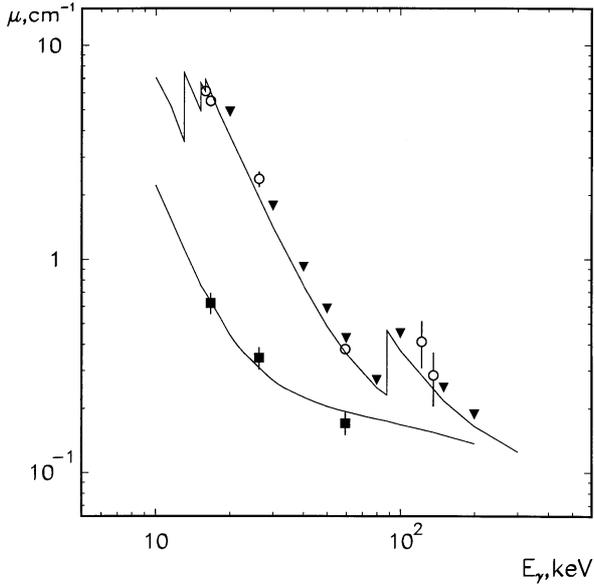


Fig. 1. Linear attenuation coefficient  $\mu$  for thin molded scintillator plates in a flux of  $\gamma$ -quanta from a  $^{238}\text{Pu}$  as a function of their energy.  $\circ$  - for heavy scintillators containing 4.07%Pb,  $\blacksquare$  - for ordinary scintillators and  $\blacktriangledown$  - for heavy scintillator BC-452. Solid curves — results of calculations (see the text).

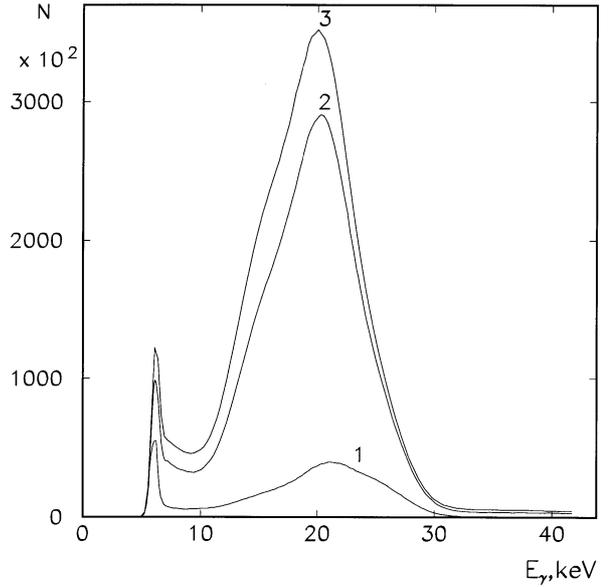


Fig. 2. Absorption spectra of  $\gamma$ -quanta from a  $^{238}\text{Pu}$  after some absorbers. All measurements were conducted with the help of a silicon semiconductor detector during 1000 c. 3 — In absence of a absorber; 2 — after ordinary scintillators with  $h=3.42$  mm; 1 — after heavy scintillators with 4.07% Pb and  $h=3.42$  mm.

## 2.2. Scintillation and radiation characteristics of molded plates

We chose 130 molded scintillator plates for this investigation. Their scintillation and radiation characteristics were submitted to the following subsection. The samples of  $\varnothing 20 \div 30$  mm were cut out from the chosen plates for comparison with other material experimental data.

### 2.2.1. Light output and light attenuation

Some distributions of the light attenuation lengths  $l_o$  and light output  $I_o$  for the scintillator plates are shown in Figs. 3a-3b. Their joint distribution is presented in Fig. 4. Note that here  $N$  is the number of counts per channel. As is clear from Figs. 3-4, our thin molded scintillator plates (without heavy metal additives) have a light output ( $I_o$ ) distribution of about 19 % from its averaged value and light attenuation lengths ( $l_o$ ) distribution — 45 %. Note that the light attenuation lengths are in a range of  $16 < l_o < 32$  cm.

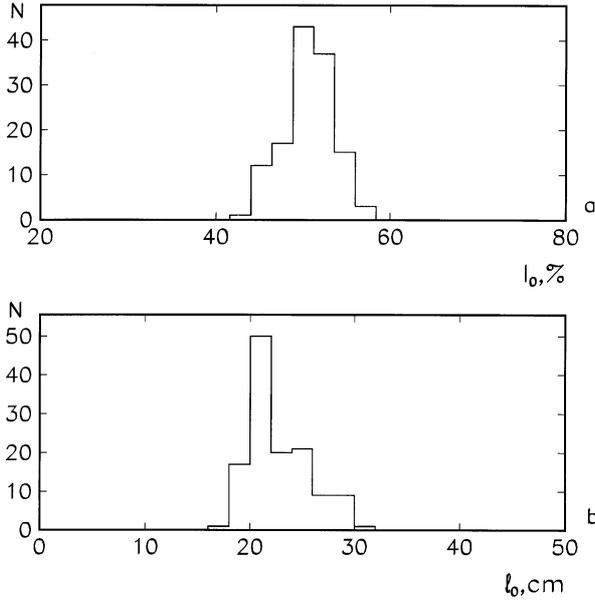


Fig. 3. Light output  $I_o$  distribution for 130 thin molded ordinary scintillator plates (a). Light attenuation lengths  $l_o$  distribution of the same plates (b).

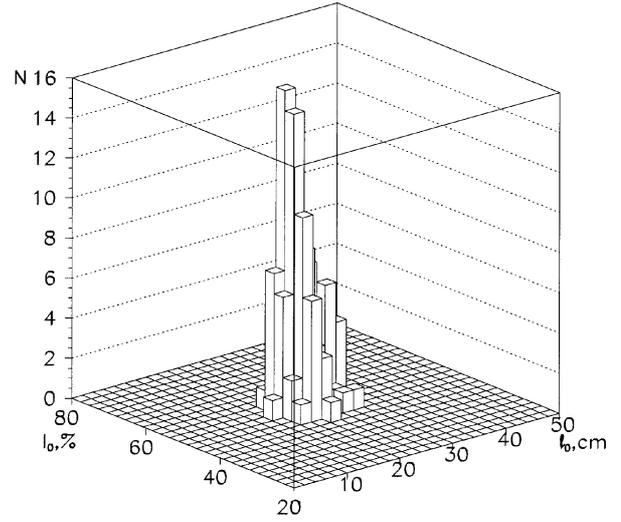


Fig. 4. 2D distribution of light attenuation lengths  $l_o$  and light outputs  $I_o$  for the same 130 plates.

**Table 2.** Optical and scintillation characteristic of some scintillators.

Scintillator	Light output $I_o, \%$	Light attenuation $l_o, \text{cm}$	w-energy, eV/photon	Decay time $\tau, \text{ns}$
Anthracene	100		65[23] 60.6±4.3 <sup>*)</sup>	27[24] 21.55±0.97 <sup>*)</sup>
NE-110	60[9] 61.0±2.9 <sup>*)</sup>	70-240	107.8±5.4[25] 100[9] 65.9±4.7[26] 82.3±4.7 <sup>*)</sup>	3.3[9] 3.28±0.11 <sup>*)</sup>
BC-452 <sup>**) (5%Pb)</sup>	32 [10]	NA	200±10 <sup>*)</sup>	2.1[10]
BC-400	65 [10]	190 <sup>***)</sup>	100±10 <sup>*)</sup>	2.4[10]
PSM-115	53.3±2.5 <sup>*)</sup>	16÷32 <sup>*)</sup>	109±10 <sup>*)</sup>	2.78±0.10 <sup>*)</sup> 2.76±0.11 <sup>***)</sup>
PSM-115 (4.07%Pb)	21.6±1.0 <sup>*)</sup>	12-15 <sup>*)</sup>	250±27 <sup>*)</sup>	2.47±0.10 <sup>*)</sup> 2.39±0.11 <sup>****)</sup>

NA – optical characteristics for this material are not presented by the manufacturing firm;

<sup>\*)</sup> – this work;

<sup>\*\*)</sup> – scintillation additives of BC-400 are used in BC-452;

<sup>\*\*\*)</sup> – for molded scintillator plates with sizes 120x200x5 mm<sup>3</sup> (this work);

<sup>\*\*\*\*)</sup> – after a dose of about 2.5 Mrad (this work).

In Table 2 we present the  $w$ -energy for our molded PS-scintillator plates and new heavy scintillators containing 4.07÷5% Pb. In Table 2 the  $w$ -energy for polyvinyltoluene scintillators NE-110, BC-400 and BC-452 and crystal anthracene are also indicated for comparison reasons. The experimental data submitted to Table 2 show that the intrinsic light output from ordinary molded plates  $I_o$  is comparative with those of PS-scintillators manufactured by Kuraray [27]. The  $w$ -energies of molded PS-scintillator plates are enough for their application in modern EM-calorimeters. It is well known that the light attenuation in all the PS-scintillators is very strong in a range of  $0 < l < 1.0$  cm. In order to avoid the nonuniformity of response in modern EM-calorimeters using such scintillators, the light needs to be collected close to where it is produced [7].

Note that in our heavy molded scintillator plates we used the cheapest TPL which underwent the minimum level of purification. Note that the metalloorganic additive concentration in our materials is comparative with those levels in similar materials manufactured by others [9,10]. The light attenuation lengths  $l_o$  of our heavy molded plates are also sufficient for their application in modern EM-calorimeters.

The experimental data submitted to Table 2 shows that all the organic scintillators have appreciable variations of their optical and scintillation characteristics from sample to sample.

### 2.2.2. Radiation resistance of ordinary thin molded plates

It is well known that the value of radiation resistance depends on many factors including the thickness of plates [14]. This is connected with the oxygen diffusion into the volume of plates and its influence on the formation and recovery of colouration centers in plastic materials during their irradiation as well as in the recovery process in air [28]. On the basis of works [14,28] we can assume that the thinner scintillator plates, the higher their radiation resistance. As far as we know the radiation resistance for such thin molded scintillator plates was not investigated earlier.

We studied the radiation resistance of 20 thin ordinary molded plates ( $h = 1.1$  mm) which were casually chosen from the above 130 plates after their scintillation and optical characteristics had been measured (see 3.2.1). Figs. 5-6 show the influence of irradiation on the scintillation and optical characteristics of the molded plates measured just after the irradiation and after one day of recovery. Note that the comparison of radiation characteristics for ordinary and heavy (containing TPL) molded scintillator plates was presented earlier in work [12] and the investigation of an aging effect on some heavy scintillators in [11].

For the determination of the radiation degradation causes, it is convenient to analyze both the light attenuation lengths and light outputs of scintillator plates as functions of the dose. So, Fig. 5 shows that an appreciable and sharp fall of  $l_o$  and  $I_o$  begins after receiving doses of  $\geq 1$  Mrad. On the other hand, it is well known [29] that the radiation resistance of the used scintillation additives pTP and POPOP (in the toluene solution) is higher than for the above dose. All this allows us to assume that for PS-scintillators at this dose (about 1.0 Mrad) the concentration of highly excited states and radicals

reaches a critical level after which the process of cross-links formation [30,31] begins to prevail. The formation of an appreciable amount of cross-links in PS-scintillator plates should result in the appearance of sharp losses of the light attenuation and efficiency of the energy transfer from the PS basis to the scintillation additives, i.e. this should result in a reduction of the plates light outputs.

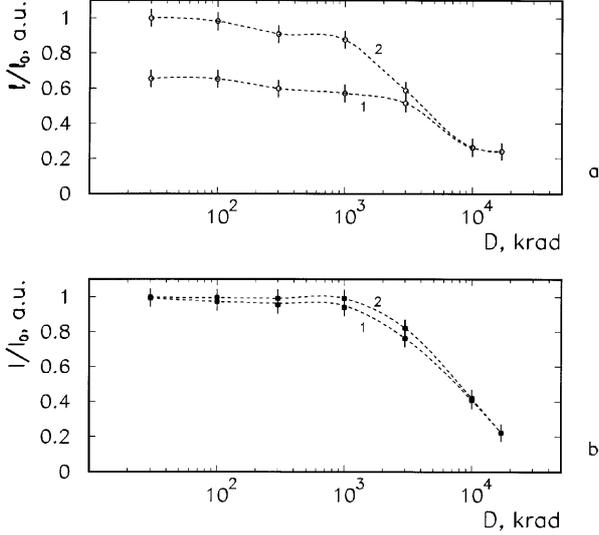


Fig. 5. Relative light attenuation lengths  $l/l_0$   $\circ$  (a) and light outputs  $I/I_0$   $\blacksquare$  (b) for thin molded ordinary scintillator plates with  $h=1.1$  mm as functions of the dose. 1 - measured just after  $\gamma$ -irradiation and 2 - after one day of recovery. Dotted curves through experimental points were drawn to guide the eye.

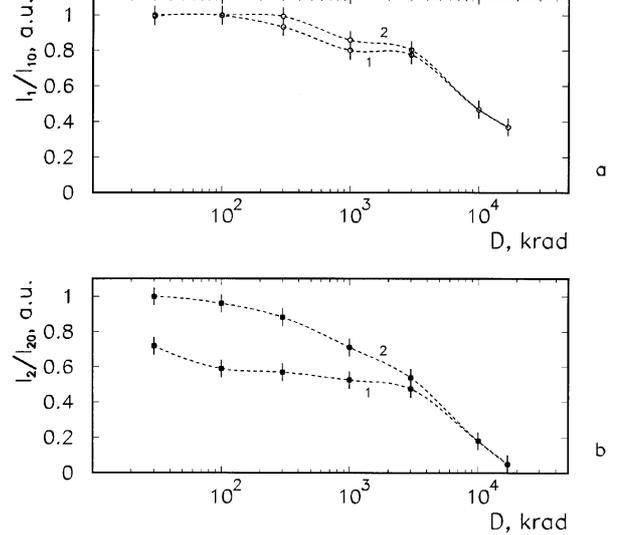


Fig. 6. Relative light outputs  $I_1/I_{10}$  from  $l_1=2.5$  cm  $\circ$  (a) and  $I_2/I_{20}$  from  $l_2=15$  cm  $\blacksquare$  of thin molded ordinary scintillator plates with  $h=1.1$  mm as functions of the dose. 1 - measured just after  $\gamma$ -irradiation and 2 - after one day of recovery. Dotted curves through experimental points were drawn to guide the eye.

For practical applications, it is more convenient to analyze the light output from the nearest  $I_1$  ( $l_1=2.5$  cm) and a distant  $I_2$  ( $l_2=15$  cm) points to the scintillator plates for the evaluation of the radiation resistance, etc. So, the knowledge of  $I_1$  as a function of the dose (Fig. 6a) allows us to evaluate the radiation resistance of modern EM-calorimeters. The comparison of  $I_1$  and  $I_2$  as functions of the dose (Figs. 6a-6b) shows a strong dependence of the radiation resistance measure on the thickness of absorbing layer to the photoreceiver (or WLS fibers), i.e. from the scintillator plates sizes. An essential difference of these characteristics behavior is observed after one day of recovery. In Fig. 6,  $I_{10}$  and  $I_{20}$  present the initial levels of light output from the nearest and distant points on the scintillator plates, respectively.

For the above application of EM-calorimeters let us define the radiation resistance for scintillator plates as such a dose of continuous irradiation at which they lose about 10÷15 % of their initial light outputs provided that all recovery processes have been completed. As is clear from Fig. 6 thin molded scintillator plates with  $h=1.1$  mm show

the radiation resistance of about  $1.5 \div 2$ . Mrad. Due to the influence of oxygen in air on the process of recovery, our thin plates show  $1.5 \div 2$ . times better level of the radiation resistance relative to similar levels for molded plates (about 1. Mrad) with  $h=3. \div 3.8$  mm [20,21]. Note that there is an opportunity of the further improvement of the radiation resistance to a level of  $D \simeq 2 \div 2.5$  Mrad connected with the reduction of molded plates thickness to  $h=0.5 \div 0.4$  mm. However, a further progress in this direction has its limitations caused by the light attenuation deterioration and increase of nonuniformity of the light output from thin plates below the required level, the influence of oxidation processes of surface layers under irradiation on the light attenuation and light output [28], etc.

As is clear from Fig. 7, characteristic times of the light attenuation  $l_o$  recovery (as well as for  $I_1$  and  $I_2$  not shown in Fig. 7) for thin molded scintillator plates with  $h=1.1$  mm is about  $t_{rec}=1$  day. The experimental data are close to the same characteristic for molded scintillator plates with  $h=3. \div 3.8$  mm [20,21]. Note that  $I_o$  for thin plates does not practically recover in time after the irradiation.

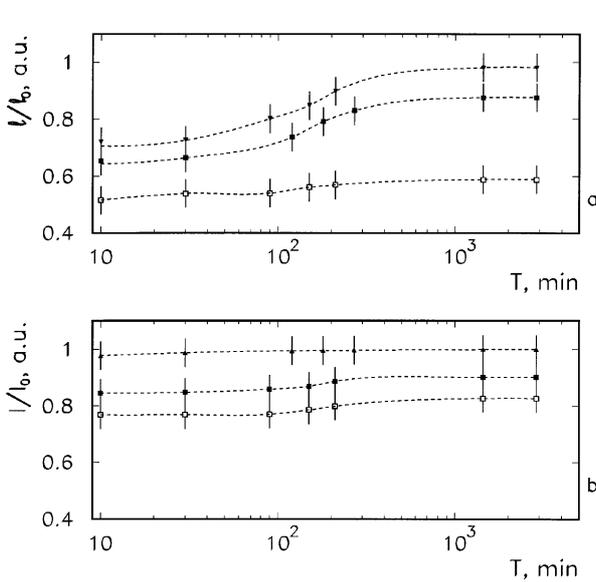


Fig. 7. Relative light attenuation lengths  $l/l_o$  recovery (a) and light outputs  $I/I_o$  recovery (b) of thin molded ordinary scintillators with  $h=1.1$  mm.  $\blacktriangledown$  - after a dose of 100 krad,  $\blacksquare$  — 1 Mrad and  $\square$  — 3 Mrad. Dotted curves through experimental points were drawn to guide the eye.

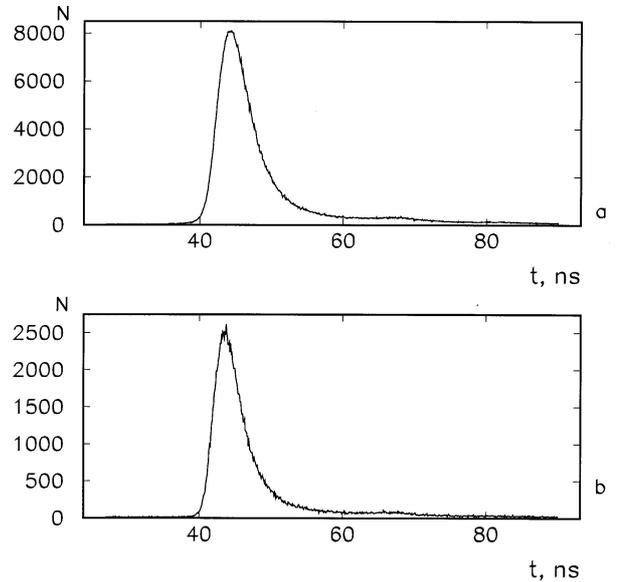


Fig. 8. Scintillation decay curves for thin molded ordinary (a) and heavy (b) scintillator plates excited by conversion electrons from a  $^{207}\text{Bi}$ .

Fig. 6 shows that after receiving doses less than  $D_{cr} = 0.3$  Mrad only weak changes of all characteristics are observed in thin molded scintillator plates. It allows us to speak about an opportunity of practically complete restoration of all characteristics of our plates through  $t_{rec}$ . Only partial changes of the above characteristics appear after doses  $0.3 <$

$D < 1$ . Mrad. They are caused by the influence of the oxidation processes under the irradiation. Significant changes of all characteristics are observed after doses  $\geq 1.0$  Mrad.

Note that the maximum time of receiving doses less than  $D_{cr}$  under continuous irradiation in physical setups is reasonably long. This allows us to realize the proposed [32] periodic mode of irradiation and recovery. This working mode may be suitable for many experimental setups at new accelerators UNK (POLEX [33]), LHC (CMS [2], FELIX [3]), etc. with the purpose of improving the radiation resistance of their EM-calorimeters.

### 2.2.3. Decay times of new scintillators

Some decay curves of new scintillators are presented in Figs. 8a-8b. The main decay times  $\tau$  were determined on the basis of the above experimental data. Some results of these measurements are summarized in Table 2 where  $\tau$  for well known scintillators are also given. As is clear from Table 2 our PS-scintillators containing heavy metal additives show only a small reduction of  $\tau$  connected with the scintillation quenching processes caused by the presence of these additives. Note that similar processes of the scintillation quenching caused by the appearance of colouration centers under irradiation also results in a small reduction of the decay time (Table 2).

## 3. Conclusion

The above calculated and experimental results show a real opportunity for the creation of heterogeneous EM-calorimeters on the basis of the "Bayan" and "Shashlyk" types with the energy resolution of  $\sigma/E \simeq 5\%/\sqrt{E}$  and high levels of the radiation resistance with the usage thin ( $\leq 1.1$  mm) ordinary scintillators.

It has been shown that the problems of creation are easier to resolve when thin molded plastic scintillators containing high concentrations  $\geq 30\%$  of metalloorganic additives are used. However, the introduction of such high concentrations of additives in the polystyrene base is near to the limit of the injection into the mold technique. A further progress in the direction of improvement of the energy resolution of EM-calorimeters to  $\sigma/E \leq 5\%/\sqrt{E}$  can be connected with the application of new scintillation materials, other methods of plastic scintillators preparation, etc. which are presented below.

Our investigations have shown that the radiation resistance of thin molded scintillator plates with thickness 1.1 mm containing 2% pTP + 0.05% POPOP reaches a level of  $1.5 \div 2$ . Mrad. There are some opportunities for further improvements of their radiation resistance connected with the reduction of plates thickness.

With increased concentrations of primary scintillation additives to a level  $\geq 6\%$ , the light output of heavy scintillators can remain at a level comparable with the light output of scintillators without heavy metal additives. So, the light output of our best new heavy polystyrene scintillator doped with 7.5% tetraphenyl lead + 6% PPO + 0.05% POPOP is 0.82 relative to the level of the light output of the scintillator without the heavy metal additive.

The satisfaction of the ecological requirements for the process of fabrication of large volumes of heavy scintillators containing the above described additives and as well as low cost requirements to materials for the new EM-calorimeters can be fulfilled most completely only with the use of tetraphenyl tin. It is the cheapest additive and all its decomposition products are not toxic. Besides, it should be noticed that the maximum concentration of tetraphenyl tin in the bulk-polymerized polystyrene reaches a level of 15% by weight and for molded polystyrene — to 25÷30%. This is almost 1.5÷2 times higher in comparison with another promising additive — tetraphenyl lead. Note that the absorption efficiencies for soft  $\gamma$ -quanta for the above heavy metals containing scintillators are reasonably close [22]. All this even more advocates the use of metalloorganic additives containing tin from other ones.

One of the simplest way to increase the maximum concentration of metalloorganic additives in scintillators to a level  $\geq 30\%$  is connected with the increase of the polystyrene temperature (to 220÷240°C) during the process of the injection into the mold. However, the progress in this direction has its limitations caused by the decomposition of used additives.

Another possible way to increase the maximum concentration of the metalloorganic additives in scintillators to a level  $\geq 50\%$  is connected with the use of vinyl derivatives of the metalloorganic additives which co-polymerize with the styrene molecules.

The third possible way to increase the maximum concentration of metalloorganic additives in scintillators to a level of about 30% is connected with the use of liquid metalloorganic additives whose solution concentrations in the styrene are usually higher than powders.

Here we may use other alternative heavy scintillation materials for the above application in modern sampling EM-calorimeters: composite plastic scintillators, i.e. plastic scintillators filled with heavy crystals powders ( $BaF_2$ ) [34] with their concentrations to  $\sim 40 \div 50\%$ . However, their optical characteristics will require further improvement.

Other alternative heavy scintillation materials that might be used in modern sampling EM-calorimeters [4,6] with the aim of improving the energy resolution and radiation resistance are transparent ceramic scintillator plates based on  $Bi_4Ge_3O_{12}$ ,  $CeF_3$ , CsJ, etc. inorganic scintillators. Note that  $Bi_4Ge_3O_{12}$  and  $CeF_3$  bases showed very high levels of the radiation resistance  $\gg 10$  Mrad [35,36].

Note that other metalloorganic additives containing heavy metals like Cd, In, etc. can be used in plastic scintillators. Such scintillators are of an appreciable interest for the effective registration of a wider class of particles: neutrons (in wider energy range), neutrino, etc.

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**The list of abbreviated names of the used additives**

DAK	–	dinitryl 2,2-iso-di-iso-oil acid
TPT	–	tetraphenyl tin
TPL	–	tetraphenyl lead
TPB	–	tetraphenyl bismuth
BPM	–	biphenyl mercury
pTP	–	paraterphenyl, $\lambda_{em} = 340$ nm
PPO	–	2,5-biphenyloxazole, $\lambda_{em} = 365$ nm
POPOP	–	1,4-bis-[2-(5-phenyloxazolyl)]-benzene, $\lambda_{em} = 420$ nm

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