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The study of inorganic scintillating materials

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Abstract. The procedure for measuring the temporal characteristics and light output of inorganic scintillating materials excited by β -, γ -, and α -particles from radioactive sources is described. Results of measurements of characteristics are presented for ~30 scintillating compounds including cerium-doped yttrium silicate and scandium borate; europium-doped strontium phosphate; cerium-doped strontium silicate, calcium silicate and magnesium calcium silicate, etc. Upon β - and γ -excitation, cerium-doped scandium borate gives the highest light output with a fluorescent lifetime of 40 ± 4 ns. The highest light output for α -excitation was from cerium-doped yttrium aluminum perovskite, with a fluorescent lifetime of 29 ± 3 ns.

1. Introduction

Inorganic scintillating materials are widely used in detectors for diverse applications ranging from high energy physics to nuclear medicine [1-2]. Better understanding of various scintillation mechanisms has led to innovative new materials for both gamma-ray and neutron detection, and the concept of scintillator design and engineering has emerged, whereby materials are optimized according to the scintillation properties needed by specific applications [3]. Although certain incremental improvements may still be achieved in each property, the main challenge then becomes to find a scintillator material that has a correct combination of properties to meet the needs of a given application rather than to find a scintillator with a single outstanding characteristic.

Intensive efforts to find and study new substances with higher light outputs, shorter fluorescent lifetimes, higher transparencies, smaller radiation lengths, etc., still continue [4-11]. The fluorescence decay lifetime of a scintillating substance is usually determined by the statistical method of single-photon counting [12]. The results obtained by this technique are independent of the shape and dimensions of samples under investigation. Measurements of the light output from substances are carried out under conditions that allow comparisons with results from other samples. For this purpose, crystals are grown to have similar dimensions (~1 cm^3). Identical conditions of light collection on a photomultiplier tube are provided, and the relative light yield is determined by the peak height of the total absorption of γ-quanta of known energy [13,14]. However, it is much easier and less expensive to synthesize a new substance in a powdered form. Based on the results of preliminary tests, one may decide whether or not to grow crystals. Thus, in [14], relative scintillation efficiency and fluorescence decay time were studied using a pulsed (τ<1 ns) monochromatic (E_γ = 22 keV) X-ray beam from the Brookhaven National Laboratory synchrotron source. A powdered scintillating compound was placed in a quartz cuvette and its light output was measured by comparing its response to radiation to the response of a bismuth germanate (BGO) crystal allowing for light absorption in the powder and the

efficiency of interaction of X-rays with the material. As a result, approximately a hundred new inorganic compounds were investigated within a short period.

Using radioactive sources that are more readily available makes it possible to perform measurements with various types of ionizing radiation. In this work, we measured the characteristics of powdered inorganic scintillators using β-, γ-, and α-sources.

2. Measurement procedure

The samples used in the study were prepared by the deposition of a fractionated luminophore powder from aqueous silicate suspension on a 22 mm diameter glass substrate. The 8–10 μm thick luminophore layer made of a powder with a grain size of 2–3 μm provides, on the one hand, identical conditions of light transmission for different samples and elimination of light loss in the scintillator and, on the other hand, necessary efficiency of detection of γ-photons with energy of ~20 keV.

In the device operating in the statistical mode of single-photon counting (figure 1), sample 1 and radioactive source *3* (^{90}Sr, ^{238}Pu) are placed between two photomultiplier tubes.

One of them, PMT_1 is in optical contact with the sample and detects light pulses whose leading edges give the start mark for intervals measured. The stop signal is provided by the other photo-multiplier, PMT_2.PMT-130 photomultipliers with a sensitivity range of 200–650 nm, and good single-electron counting characteristics were used for the measurements. Attenuation of light burst intensity required for single-photon counting on PMT_2 is accomplished with diaphragm 5 and by varying distance L from the sample. Since radioactive source 3 prevented the direct passage of light from the sample to PMT_2, mirror 2 and reflector 4 were applied to provide light collection.

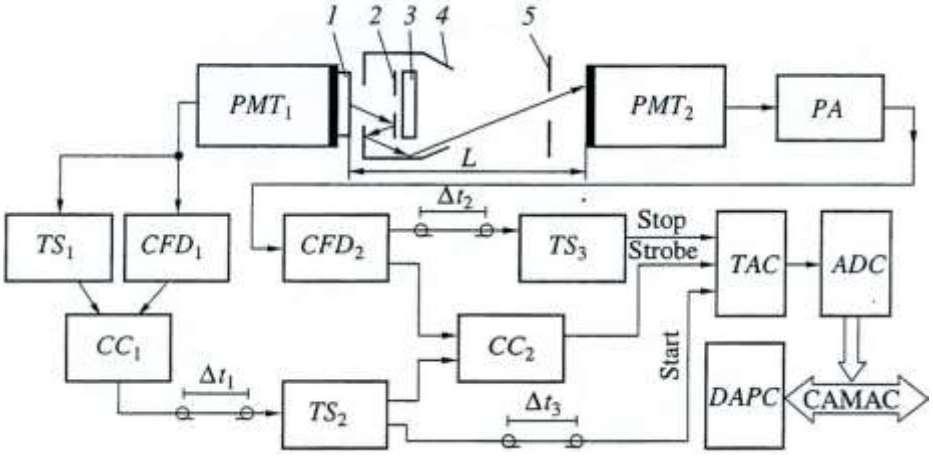

Figure 1. Diagram of the setup for measurements of fluorescence lifetime of scintillating materials. PA is the preamplifier; CFD_1 and CFD_2 are the riding-threshold shapers, TS_1 and TS_2 are the threshold shapers; CC_1 and CC_2 are the coincidence circuits; TAC is the time-to-amplitude converter; ADC is the analog-to-digital converter; DAPC is the data acquisition and processing complex; Δt_1, Δt_2, and Δt_3 are the matching delay lines; (1) sample, (2) mirror, (3) radiation source; (4) reflector, (5) diaphragm.

To form the start signal, a two-threshold detection system is used that includes a constant-fraction discriminator, threshold shaper (TS_1), and coincidence gate. The constant- fraction discriminator with a low (~2 photoelectrons) threshold accomplishes accurate timing of the instant of excitation of the sample. In the case of β-activation, the timing starts from the pulse front of the Cerenkov radiation produced in the glass substrate and determining the front of the overall light pulse. The threshold of shaper TS_1 is established at a level of ~0.5 of the signal peak height.

Pulses from PMT_2 (stop channel) after the amplification with a preamplifier pass to discriminator CFD_2 having the detection threshold in the "valley" of single electron pulse distribution. Relative efficiency of detection of scintillation flashes by this channel is established at a level of 1/20 by the

ratio of counts from the coincidence circuits CC_1 and CC_2, thus providing the probability of <2.5% for detecting many-electron events.

The start and stop signals are transmitted to the time-to-amplitude converter (*TAC*) triggered by a strobe pulse from the coincidence circuit CC_2. Timing of signals is accomplished with cable delay lines. Threshold shapers TS_2 and TS_3 are used to restore the front and the amplitude of the start and stop signals, which are distorted by delay lines. Measurements on the luminophore-free glass substrate used as a Cerenkov light emitter gave the time resolution of the device as $\sigma = 0.68$ ns.

Fluorescent lifetime's τ_2 and $\tau_3$are determined as parameters of approximation of time spectrum by two exponentials (figure 2)

The total uncertainty of determination of τ_2 does not exceed 10%. In order to verify the procedure, we measured the fluorescence lifetime of the KS-425-1 commercial luminophore, which is a constituent of a type V-2 electrooptical transducer, upon excitation with monoenergetic electrons. To synchronize the instant of luminophore excitation with the start pulse, a silicon carbide light-emitting diode was used (flash duration <2 ns) which illuminates the photocathode of the electrooptical transducer.

The value measured in this manner agrees, within the limits of accuracy, with the τ_2 and τ_3 values obtained upon excitation with a ^{90}Sr β-source.

To measure the afterglow intensity *F* of a substance, an additional delay ($\Delta\tau_3$) of 5 μs was introduced to the start channel. Time spectra with and without the delay are built-up during the same period, and the *F* value is determined as the ratio of the number of events in these spectra within the interval τ_2 for the given substance. The accuracy of determination of *F* is ~20%.

The light yield from samples is measured by two methods. The first one is based on the use of a ^{90}Sr β-source. For β-particles, the ionization energy loss d*E*/d*x* in a thin layer of a scintillator depends mainly on its density. Relative light yield accurate to within ~20% is determined from the ratio of counts in the start and stop channels (introducing a correction for density of the material).

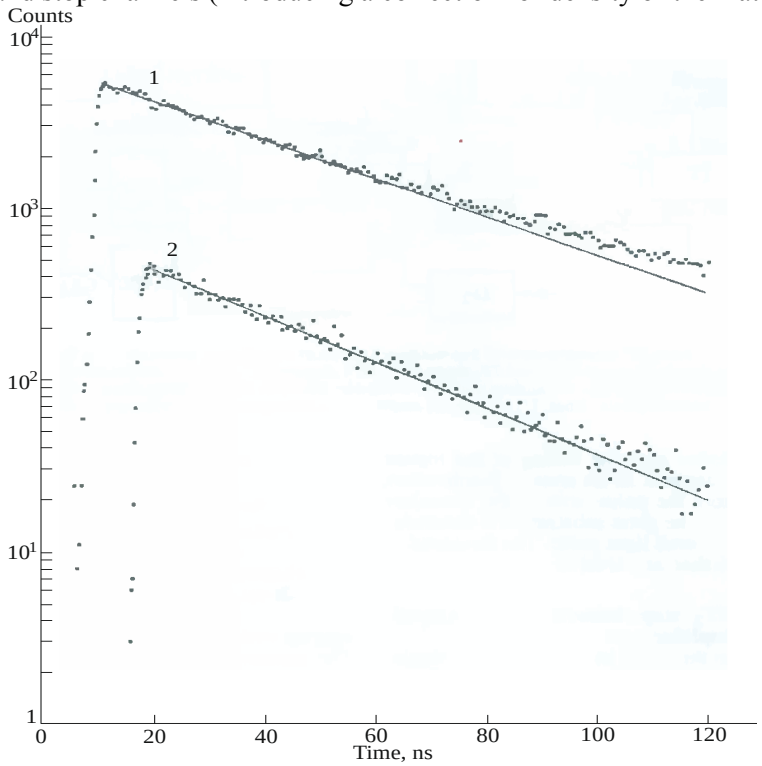

Figure 2. Temporal spectra of fluorescence decay of scintillating materials upon ^{90}Sr β-excitation: (1) KS-425-1 (two-exponential decay law), (2) and $YAlO_3$: Ce (one-exponential decay law).

More accurate determination of the light yield is performed with the use of γ-sources of low energies: ^{65}Zn (E_γ= 8.1 keV), ^{113}Sn (E_γ= 24,7keV), and ^{241}Am ($E_{\gamma1}$ =13.2 keV, $E_{\gamma2}$ = 17.8 keV, $E_{\gamma3}$= 59.5 keV) [15]. The relative light yield for α-particles was determined by using a ^{238}Pu source. Because the range of α-particles from this source exceeds the thickness of scintillation screens under study, the mode of total energy absorption is achieved by varying the distance from the α-source to the luminophore so that to obtain the required energy of α-particles at the expense of ionization loss in air.

3. Results

The characteristics of ~30 types of scintillating substances were measured on the apparatus. Tables 1 and 2 list these characteristics for β- or γ-excitation and for α-excitation, respectively. Rows 2–22 in Table 1 and rows 2–8 in Table 2 present the data on luminophores prepared in the Mendeleev University of Chemical Technology (the wavelengths at the maxima of emission spectra were also measured there). The $YAlO_3$: Ce crystal and Scandium borate, produced at the Research Institute of Nuclear Problems of Belarussian State University (Minsk), was kindly presented by the Joint Institute for Nuclear Research (Dubna).

Table 1. Scintillating materials and their characteristics on β-excitation

№	Scintillatortype	λ_m, nm (color)	τ_2, ns τ_3, ns	L_{rel},%	F,%
1	KS-425-1 [Y_2SiO_5 : Ce (1.5 wt%)]	410	38±4 51±4	100	~0.03
2	Y_2SiO_5: Ce + BaF_2	410	39±4	86	0.03
3	Y_2SiO_5: Ce (2 wt %)	380	35±3	29	0.03
4	$2YPO_4$: Ce	365	33±3	5	0.6
5	$Y_{0.8}{\cdot}La_{0.1}{\cdot}Cd_{0.1}{\cdot}PO_4$: Ce	365	39±4	6	0.03
6	$SrO{\cdot}SiO_2$: Ce (0.04 mol %)	410	48±5	27	0.31
7	$SrO{\cdot}SiO_2$: Ce (0.04 mol %) with additional calcination at, t = 1250°C for 1 h	410	45±4	5	1
8	$Sr_3(PO_4)_2$: EU_2O_3 (0.6 wt%) t = 100°C	425	375±35	89	—
9	$Sr_3(PO_4)_2$: EU_2O_3 (0.6 wt%) t = 1200°C	425	395±40	89	—
10	$Sr_3(PO_4)_2$: EU_2O_3 (0.6 wt%) t = 1300°C	425	405±40	83	—
11	$Sr_3(PO_4)_2$: EU_2O_3 (1.2 wt%) t =1300°C	425	389±40	44	—
12	3CaO $2SiO_2$: Ce (0.1 mol %)	410	39±4	22	0.3
13	3CaO $2SiO_2$: Ce (0.1 mol%) with additional calcination at, t = 1250°C for 1 h.	410	47±5	4	0.6
14	2CaO MgO· $2SiO_2$: Ce, calcination for 1.5 h, t=1250°C	380	48±5	4	2
15	2CaO MgO· $2SiO_2$: Ce, calcination for 3 h, t = 1250°C	380	47±5	4	0.7
16	$2CaAl_2O_3{\cdot}SiO_2$: Ce (0.015 mol %)	400	56±5	4	—
17	$CaZrO_3$: Ti	White	276±30	37	—
18	$BaZrO_3$: Ti	White	40±4 959±90	31	—
19	$ScBO_3$: Ce (0.8 mol %)	390	33±3	62	0.03
20	$ScBO_3$: Ce (2.5 mol %)	390	32±3	60	0.02
21	$ScBO_3$: Ce (1.5 mol %) + BaF_2 (0.3 mol %)	390	38±3	67*	0.03
22	$ScBO_3$: Ce (1.5 mol %) + BaF_2 (1 mol %)	390	39±4	75*	0.04
23	$ScBO_3$: Ce (Minsk)	393	40±4	156*	0.03
24	$YAlO_3$: Ce (3 x 3 x 0.1 mm^3 crystal) (Minsk)	347	32±3	98*	0.03
25	CsI(Tl) (3 x 3 x 0.6 mm^3 crystal) (Minsk)	430	710±50	100*	—

Table 2. Scintillating materials and their characteristics on α-excitation

№	Scintillator type	λ_m, nm (color)	τ_2, ns τ_3, ns	L_{rel},%	F,%
1	KS-425-1	410	29 ±3	100	0.03
2	Y_2SiO_5: Ce, t = 1350°C	410	28 ±4	69	0.03
3	Y_2SiO_s: Ce, t = 1350°C	410	30±3	116	0.03
4	Y_2SiO_5: Ce + NF_4, t = 1350°C	410	20 ± 2	69	0.02
5	Y_2SiO_5: Ce + BaF_2, t = 1350°C	410	26 ± 2	89	0.02
6	Y_2SiO_5: Ce + BaF_2, t = 1300°C	450	24 ±2	57	0.03
7	Y_2SiO_5: Ce + BaF_2,t = 1350°C,	White	21 ±2	121	0.02
8	$ZrGeO_4$: Ti·P	430	110 ± 10 500 ± 50	75	2
9	KO-425 (Y_2SiO_5: Ce, Gd)	410	45 ±4	72	0.04
10	$ScBO_3$: Ce (Minsk)	393	37 ±4	131	0.03
11	$YAlO_3$: Ce (Minsk)	347 [8]	29 ±3	216	2

4. Conclusion

An analysis of the results shows that the highest light yield upon β-excitation is provided by cerium-doped scandium borate (a sample from Minsk). Its light yield is higher than that of CsI(Tl). For α-excitation, the cerium-doped yttrium aluminum perovskite (YAP:Ce) crystal has unique light yield characteristics. For scintillators intended for the identification of charged particles, the α/β ratio is of practical importance. We determined this ratio for Y_2SiO_5: Ce, $YAlO_3$: Ce, and $ScBO_3$: Ce as 0.19, 0.46, and 0.16, respectively. Tables 1 and 2 also show the dependence of luminescent lifetimes of inorganic scintillators on the type of particles exciting the scintillations. This time is shorter for α- than for β-excitation; the difference is especially pronounced in the case of the KS-425-1 luminophor.

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