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Energy characteristics of scintillators for X-ray introscopy

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In a broad energy range of X-ray radiation ($U = 2 \div 175$ кV), we have studied output characteristics (light output, quantum yield of luminescence, etc.) of scintillators based on ZnSe crystals, as well as scintillators CsI(Tl), CWO, GSO and Al_2O_3(Ti). It has been shown that maximum quantum yield, as well as light output in the energy range $E_x < 80$ keV, is observed for scintillators based on zinc selenide. Their advantages are considered for applications in low-energy detection subsystems of multi-energy X-ray introscopes. Effects of geometrical factors scintillation elements upon output characteristics of such scintillators are discussed.

В широком диапазоне энергий рентгеновского излучения ($U = 2 \div 175$ кВ) изучены выходные характеристики (световыход, квантовый выход люминесценции и др.) сцинтилляторов на основе ZnSe, а также сцинтилляторов CsI(Tl), CWO, GSO и Al_2O_3(Ti). Показано, что максимальный квантовый выход, а также световыход в области энергий $E_x < 80$ кэВ имеют сцинтилляторы на основе селенида цинка. Определены перспективы их применения в низкоэнергетических подсистемах детектирования рентгеновского излучения в мульти-энергетических интроскопах. Изучены геометрические факторы сцинтилляционных элементов, влияющие на выходные характеристики сцинтилляторов.

In modern X-ray introscopy systems (XRIS) for medical and technical tomography, osteodensimetry, customs and security introscopy, etc., the most widely used are radiation detectors of the "photodiode-scintillator" type (PD-S). The efficiency of a PD-S detector is determined both by properties of the scintillation crystal (SC) used and by parameters of the photoreceiver. The signal value Q at the detector output under gamma-quanta flux of energy E_x is [1]:

$$Q = (k_{abs} \cdot e \cdot E_x/h\nu) \times \times (\eta_{sc} \cdot \eta_{pd} \cdot \eta_e \cdot k_{lc} \cdot k_{sm}), \quad (1)$$

where k_{abs} is the fraction of radiation that is absorbed by SC, e is electron charge, $h\nu$ is average energy of scintillation photons, η_{sc} is conversion efficiency of SC, η_{pd} is quantum efficiency of the photodiode, η_e is efficiency of charge carrier collection on PD, k_{lc} is light collection coefficient, k_{sm} is spectral matching coefficient of scintillations in the region of PD sensitivity. Modern technologies allow preparation of PD with parameters close to the theoretical limits, and the problems of production of detectors for XRIS mainly consist in the choice of SC with optimum energy characteristics — quantum efficiency and dependence of the light output upon the detected radiation energy.

An additional condition for achieving higher efficiency of the scintillation transformation by a combined detector is the choice of an optimum thickness, at which the ionizing radiation is fully (commonly considered value is 90 %) absorbed by the detector material. A further way for improving the efficiency is creation of the optimum light collection conditions from the scintillator to the photoreceiver. The light collection coefficient τ depends upon the scintillator shape, its refraction coefficient (n), absorption factor (α, cm^{-1}); also important is the use of additional reflecting surfaces, optical adhesive between the scintillator and the photoreceiver, etc.

The commonly used scintillators for detection of low-energy X-ray radiation include such single crystalline scintillators like CsI(Tl), NaI(Tl), CWO, etc. It would be of great interest to use scintillators of other types, in particular, semiconductor scintillators based on isovalently doped ZnSe. These scintillator materials are non-hygroscopic, have high conversion efficiency, as well as good spectral matching with Si photodiodes and other advantages.

The present work was aimed at establishing conditions for optimized uses of scintillators of different types CsI(Tl); CWO; ZnSe(IVD), where IVD — O, Te, Cd; Al_2O_3(Ti); GSO) in XRIS of special purpose (tomographic, industrial, two-energy systems for security/customs inspection) in the X-ray radiation energy range of E_x = 2–150 keV. The concentration of activating dopants in the crystals was 0.1–0.5 % (mol.). The crystals were grown by conventional methods.

In this study, we considered energy characteristics of scintillators, such as the absolute energy yield of the X-ray luminescence (η), light output as function of energy $I(E_x)$ and sample thickness $I(d)$ at different values of the energy E_x of the X-ray radiation and different shapes of the scintillator samples.

Measurements of the light output ($I_{l.o.}$) were carried out in the current mode under irradiation using a REIS X-ray source (I = 50 μA, U = 45 kV) with BS-1 microfocal tubes. To determine statistical error of the measurements, data were used that had been obtained using different X-ray tubes. The optical radiation power was recorded by a "Kvarts-1" device with a receiver of silicon photodiode FD-288 with known spectral distribution of sensitivity. The samples used were discs with polished top and bottom side and grinded rims, 2 mm thick and of diameter corresponding to the diameter of the photodiode sensitive area. As a reference, a CsI(Tl) crystal was used, with its light output 55000 h/meV.

Measurements of X-ray luminescence light output as function of energy $I(E_x)$ were carried out using 10×10×0.7 mm^3 scintillator samples. As excitation sources, we used X-ray devices IRI with W-tubes (voltage range on the tube from 50 to 200 kV) and REIS-I (Ag-tube BS-1, U = 2–50 kV). A FEU-100 PMT was used as receiver, with its spectral sensitivity range from 200 nm to 800 nm. The sample was located directly on the anode of the X-ray tube. The measurements were carried out both in the case of the polished output window and of the grinded one. In addition, studies were carried out of the effects of a TAVEK reflective coating of 0.05 mm thickness.

Absolute energy yield of scintillation materials. The absolute energy yield (η), defined as ratio of the total energy of the luminescence photons to the energy spent for their excitation, is the most important and universal characteristic of X-ray luminophores and scintillation materials. Direct methods of measuring the absorbed dose in the current measurement mode (conversion efficiency (η_{conv})) include early works on determination of the absolute energy yield by means of chemical actinometry [2]. In practice, many scintillators operate in the spectrometric (pulse) mode; therefore, spectrometric methods for determination of the absolute energy yield, scintillation efficiency (η_{scint}), are widely used, based upon calibration of the spectrometric circuit and defined either by the one-electron PMT level or by number of electrons that are generated in a silicon detector [3–4].

In this work, we determined absolute values of the light output of scintillation crystals under X-ray excitation in the current measurement mode. All possible factors affecting the true value of the scintillator light output were intended to be accounted for. As reference, a sample with known light output value was used. Such measurement procedure allows accounting for contributions from both fast and slow luminescence components and is the most close to the real operation conditions of scintillators in introscopic systems.

In this case, the light yield value should be described by the following expression:

$$\eta = I_{l.o.}/k_{sm} \cdot k_{lc} \cdot k_{abs}, \tag{2}$$

where $I_{l.o.}$ is light output, k_{sm} is spectral matching coefficient between scintillator radiation and photoreceiver sensitivity; k_{lc} is light collection coefficient of the scintillator sample; k_{abs} is coefficient of X-ray absorption in the substance.

Table 1. Calculated values of coefficients k_{sm}, k_{lc}, k_{abs}, experimental data of the light output $I_{l.o.}$, and absolute light yield η values for scintillator samples

Scintillator crystal	k_{sm}	k_{lc}	k_{abs}	$I_{l.o.}$	η		
					calculated from (2)	with respect to CsI(Tl)	ph/meV
CsI(Tl)	0.77	0.115	1	0.00296±0.00005	0.0334±0.0006	1.0	55000
ZnSe(O)	0.87	0.076	1	0.00226±0.00005	0.0342±0.0008	1.024±0.03	56000±2000
ZnSe(Te)	0.89	0.077	1	0.00303±0.00005	0.0442±0.00075	1.323±0.02	73000±1500
ZnSe(Cd)	0.88	0.076	1	0.00255±0.00005	0.0382±0.0007	1.142±0.02	63000±2000
CWO	0.73	0.09	1	0.00085±0.00004	0.0125±0.0006	0.374±0.01	20500±1000
GSO	0.66	0.12	1	0.00045±0.00004	0.0057±0.0007	0.171±0.01	9500±600
Al_2O_3(Ti)	0.9	0.08	1	0.00034±0.00004	0.0046±0.0004	0.141±0.01	7500±600

Thus, the problem of determination of the absolute value of the light output of scintillators is reduced to measuring η and calculating the coefficients in (2).

The samples were placed directly between the X-ray tube cathode and the photoreceiver, without additional optical contact between the sample and photoreceiver, and reflecting coatings of the passive surfaces of the sample. The light output $I_{l.o.}$ value was determined as ratio of the recorded energy flux of solar radiation to the falling flux of the X-ray energy.

Spectral matching coefficient was determined as

$$k_{sm} = \int I(\lambda)S(\lambda)d(\lambda) / \int I(\lambda)d(\lambda), \tag{3}$$

where $I(\lambda)$ is the spectral distribution of the scintillator emission, $S(\lambda)$ is spectral sensitivity of the photoreceiver.

Calculations of k_{lc} for sample scintillators were carried out using the calculation algorithm, which uses the Monte-Carlo method for solution of the problem of light collection in the scintillator volume [5]. As initial data for calculations, we used the geometry and size of the samples, degree of the surface treatment, absorption coefficient of the material α, refraction coefficient; account for the boundary between the sample and the photoreceiver was also made. Scintillations inside the sample were considered as surface-adjacent, as the penetration depth of X-ray radiation of energy $E_x \approx 30$ keV is substantially less than the sample thickness. Therefore, k_{abs} was taken as unity.

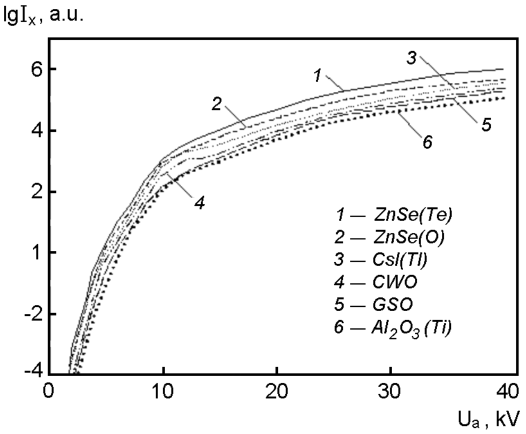

Fig. 1. X-ray luminescence light output I_x as function of the X-ray source anode voltage for different scintillators of thickness 0.7 mm.

In Table 1, values of coefficients k_{sm}, k_{lc}, k_{abs} are presented, as well as experimental data on values of $I_{l.o.}$ and the absolute light yield values obtained using (2).

Energy dependence of X-ray luminescence light output. Low-energy sensitivity limit has been determined for different scintillators. Fig. 1 shows X-ray luminescence light output as function of energy corresponding to the X-ray tube voltage from 2 to 40 kV.

Fig. 2 shows light output I_x values of crystals ZnSe(IVD), CsI(Tl) as function of the X-ray source anode voltage for samples of thickness 0.7 mm and 4.0 mm in the energy range corresponding to X-ray tube voltages from 50 to 175 kV.

Analysis of dependences of the light output values upon the X-ray excitation energy (Fig. 1–2) have shown that:

a) all scintillators are characterized by a sharp sensitivity rise (up to 7–8 orders of magnitude) in the energy range corresponding to X-ray tube voltages from 3 to 10 kV,

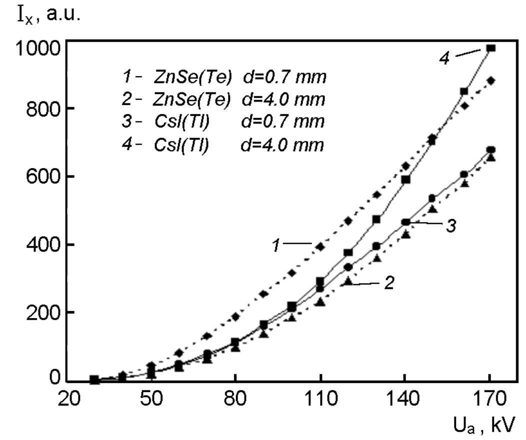

Fig. 2. Light output I_x of crystals ZnSe(IVD), CsI(Tl) as function of the X-ray source anode voltage and sample thickness d.

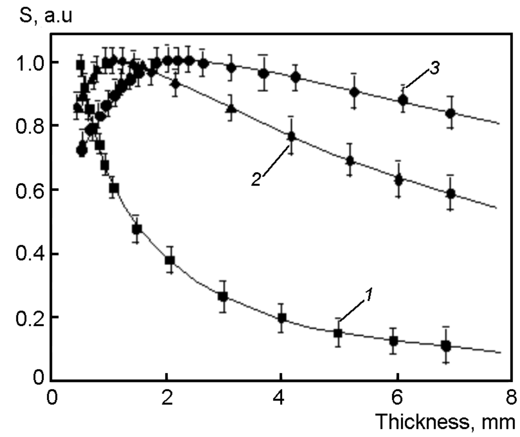

Fig. 3. Relative light yield S as function of ZnSe(IVD) scintillator thickness under X-ray excitation: 1 — U = 20 kV, 2 — U = 50 kV, 3 — U = 100 kV.

with subsequent monotonous increase of the light output with increasing energy;

b) for scintillators of 0.7 mm thickness, the light output is higher when a grinded output window is used. The use of TAVEK reflective coating increases I_x by 80–100 %;

c) maximum values of I_x in the low-energy range (up to 70–80 keV) are observed for ZnSe(IVD) scintillators, which is in an agreement with the above results on the absolute light yield;

d) upon thickness changes of CsI(Tl) scintillators from 0.7 mm to 4.0 mm at small energies of X-ray radiation (voltage on the tube up to 100 kV), light output values remain unchanged. With ZnSe(IVD) scintillators, such changes are observed, which is related to substantial absorption of intrinsic radiation in the crystal volume. The absorption of intrinsic radiation, in combination with low values of the effective atomic number Z_{eff} of ZnSe(IVD), is also the reason why the light output at energies above 70 keV becomes lower than the values for CsI(Tl) (Fig. 2).

Effect of substantial intrinsic absorption in ZnSe(IVD) scintillators are confirmed by the dependences of the relative light yield upon thickness of ZnSe(IVD) scintillators at different X-ray excitation energies (Fig. 3).

It can be seen from Fig. 3 that at energies below 70 keV the use of ZnSe(IVD) scintillation crystals of thickness higher than 2–3 mm is not reasonable because of strong absorption of the intrinsic optical radiation — $\alpha = 0.1–0.3$ cм$^{-1}$, which leads to a fall in detector sensitivity. Another parameter that is disadvantageous with ZnSe(IVD) crystals at high energies is their high refractivity coefficient (n = 2.58). This leads to substantial light losses because of low n value (1.4–1.6) of the optical contact between the scintillator and the photoreceiver (the optimum value would be $n_{opt.} \approx 1.9$), as well as because of high fraction of the "captured" light inside scintillators of "regular" forms (parallelepiped, cylinder, etc.).

Dependence of the light output values upon the scintillator shape. One of the ways to improve sensitivity of "scintillator-photodiode" detectors is the use of scintillator element shapes that ensure optimum light collection conditions. For this purpose, calculations have been carried out of the light collection coefficient τ for different scintillator shapes. As a specific example, we used ZnSe(IVD). The value of τ (the fraction of light that passed through the output window of the scintillator) was determined by Monte-Carlo calculations. The calculation algorithm accounted for the sample geometry, absorption in the scintillator material (α = 0.1–0.2 cm^{-1}), refraction coefficient (n = 2.58), the light scattering indicatrix at the crystal — reflective coating boundary, as well as a number other parameters. Different shapes of ZnSe(Te) scintillators were considered, such as polyhedrons — parallelepiped, parallelepiped with a rounded rib, as well as tetrahedral pyramid, tri- and hexahedral prisms, hemispheres. Values of τ for scintillation elements with an output window corresponding to the sensitive area of a silicon photodiode (S = 1 cm^2) are presented in Table 2.

Table 2. Calculated values of light collection coefficient for scintillator elements of different shapes with output window area $S = 1$ cm^2 (Monte-Carlo method)

Scintillator shape	Light collection coefficient, τ	
	Scintillations uniformly distributed over the volume	Scintillations confined to the surface-adjacent layer
Parallelepiped (cube)	0.144	0.145
Parallelepiped with rounding	0.339	0.371
Pyramid, 60°	0.60	0.668
Pyramid, 45°	0.49	0.51
Hemisphere	0.457	0.652
Trihedral prism (variant 1)	0.357	0.378
Hexahedral prism (variant 1)	0.202	0.221
Trihedral prism (variant 2)	0.147	0.146
Hexahedral prism (variant 2)	0.145	0.147

It can be seen from the Table that variation of the scintillator shape can lead to noticeable (up to 3 times) in the light collection coefficient value. The lowest τ values are observed for scintillators of "correct" forms (parallelepiped, cylinder, prisms, pyramid with a 45° vertex angle (angular reflector)), which can be explained by large fractions of light captured inside the volume. In the case of a tetrahedral pyramid with a 60° angle at the vertex and a hemisphere, effective "mixing" of light in the space of angles, which favors good output of light from the crystal.

In the general case, these results can be explained by a theoretical model, according to which light propagation inside a crystal can be described by the mathematical billiard theory [6]. According to this model, optimization of the light output can be realized by finding such a geometry for the scintillator that would ensure a cross-over from the regular dynamics of light propagation in the scintillator volume to non-regular stochastic mixing.

Experimental arguments in support of this model have been found. Specifically, it has been shown that rounding of vertexes and ribs of ZnSe(Te) and CsI(Tl) scintillator crystals of rectangular and cylindrical shape (i.e., passing from the regular light propagation dynamics to the chaotic case) leads to increases in the light output up to 20 % .

The obtained energy characteristics in combination with other scintillation characteristics of the materials studied have shown that, for application in detectors of X-ray introscopes and tomographs, the most preferable crystals are CsI(Tl), CWO и ZnSe(IVD). Due to their high light output values and low effective atomic number Z_{eff}, ZnSe(IVD) crystals are the most suitable and promising for the low-energy subsystem of XRIS detectors. Accounting for the high absorption level of intrinsic radiation ($\alpha \approx 0.1 \div 0.2$ cm^{-1}), which causes high losses of light at higher crystal thickness, optimum conditions and shapes have been determined for ZnSe(IVD) crystals, ensuring optimum conditions for efficient scintillation transformation in the crystal.

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Енергетичні характерістики сцинтиляторів для рентгенівської інтроскопії

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У широкому діапазоні енергій рентгенівського випромінювання ($U = 2 \div 175$ кВ) вивчено вихідні характеристики (світловий вихід, квантовий вихід люмінесценції та ін.) сцинтиляторів на основі ZnSe, а також сцинтиляторів Cs(Tl), CWO, GSO і Al_2O_3(Ti). Показано, що максимальний квантовий вихід, а також світловий вихід в області енергій $E_x < 80$ кеВ мають сцинтилятори на основі селеніду цинку. Визначено перспективи їхнього застосування у низькоенергетичних підсистемах детектування рентгенівського випромінювання у мультиенергетичних інтроскопах. Вивчено геометричні фактори сцинтиляційних елементів, що впливають на вихідні характеристики сцинтиляторів.