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## Development of a technique for quantitative comparison of optical power of self-glowing crystals

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The paper describes a method for quantitatively comparing the luminosity of self-glowing samples based on photocurrent measurements. This technique takes into account the spectra of radiation-induced luminescence (self-glow) of the samples under study, as well as spectral functions of the spectrometer and photodetector. Calculations performed by using this technique open up the possibility of quantitatively determining the power of optical radiation from a self-glowing crystal. Also, the proposed method can be used to compare the luminosity of materials subjected to radioactive radiation.

**Keywords:** optical power, radiation-induced luminescence, rare earth ions, zircon, xenotime.

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Many up-to-date electronic devices (most often, microprocessors and memory modules) need not only a prime source of power in the form of either mains or high-capacity batteries, but also constant power supply from low-current sources, such as small chemical power supplies. The main problem in employing chemical power supplies is their relatively short service life (no more than 10–15 years), which means that they need regular replacement throughout the entire service life of the electronic device. When electronic devices are exploited in the mode of limited access (Outer Space, the Arctic, etc.), batteries with a service life of up to 50 years are desired. As a promising solution to this problem, items based on radionuclides are regarded [1,2].

Conversion of the radionuclide decay energy (alpha or beta radiation) into electrical energy can proceed in both direct and indirect ways [3]. Among the indirect methods, utilizing so-called self-glowing crystals should be singled out [4]. A self-glowing crystal is an efficient scintillator activated by an alpha-radioactive isotope, such as <sup>241</sup>Am or <sup>238</sup>Pu. The crystal self-glow is caused by decay of the radioactive isotope introduced into the crystal matrix during its growth [5]. To enable the use of such an optical radiation source as a source of electric current, photoelectric converters are applied. Obviously, it is necessary to select optimal radionuclide–scintillator combinations. The simplest way for this is comparing the output optical powers of crystals self-glow or those of forced luminescence of individual scintillators under the impact of alpha or beta radiation.

The goal of this study was to develop a technique allowing quantitative comparison of the optical powers of self-glowing samples based on measuring the detector photocurrent generated under optical irradiation; thereat,

self-glowing spectral compositions of the samples under study should be taken into account.

In this work, self-glowing crystals of two types were studied: ZrSiO<sub>4</sub>:Tb<sup>3+</sup> and YPO<sub>4</sub>:Eu<sup>3+</sup>. Both the materials were activated in the process of synthesis with the <sup>238</sup>Pu isotope. Detailed description of the methods for obtaining and results of studying these materials were described elsewhere [6]. Table 1 presents the compositions of the studied samples.

The size of the obtained single crystals did not exceed 1 mm. To make the material handling more safe, there were fabricated brass cuvettes 20 mm in diameter with an internal hollow 4 mm in diameter. The hollow was filled with self-glowing single crystals and covered from top with a quartz glass (since the main type of <sup>238</sup>Pu decay is alpha decay, all this ensures safety of handling these materials).

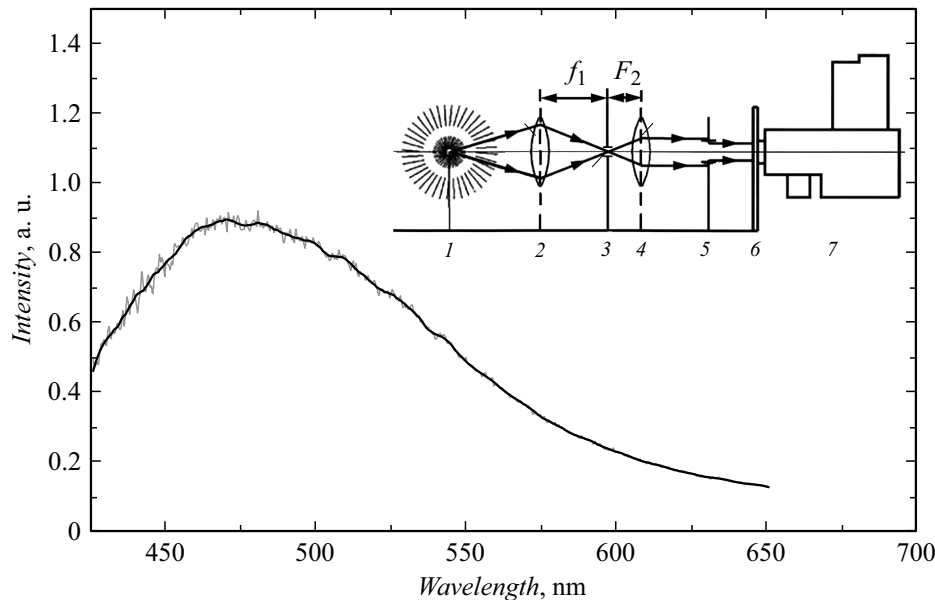
The self-glowing spectra of the obtained samples were measured on a special optical bench (its schematic diagram is shown in the inset to Fig. 1). The signal was detected by using an optical spectrometer with photoelectron multiplier FEU-106.

Since the samples exhibit a low level of glowing, they were mounted directly in front of the diaphragm (position 3 in the Fig. 1 inset). The bench instrument function is shown in Fig. 1. The spectral function was determined by using an incandescent lamp with a DC power supply; the lamp spectral characteristic was measured with DFS 36 (LOMO).

To measure the photocurrent generated by the light from the samples, a special setup was designed and constructed. It is a light-proof box inside which there are installed a sample holder and optical radiation detector based on a silicon photodiode [8]. Between the self-glowing crystal and detector there is a deflecting shutter necessary for measuring the photodetector dark signal. The setup layout is presented

**Table 1.** Compositions of the self-glowing samples under study

Sample	Activator content, wt.% (activator type)	Radionuclide content ( <sup>238</sup> Pu), wt.%
ZrSiO <sub>4</sub> :Tb <sup>3+</sup> , <sup>238</sup> Pu	0.3 (Tb <sup>3+</sup> )	0.02
YPO <sub>4</sub> :Eu <sup>3+</sup> , <sup>238</sup> Pu	1.7 (Eu <sup>3+</sup> )	0.1



**Figure 1.** Instrument function of the self-glowing measurement bench. The inset presents the basic optical scheme of the bench: 1 — light source, 2 — light collimator, 3 — diaphragm (entrance slit), 4 — quartz lens for creating a parallel light beam, 5 — exit slit for creating a parallel beam of a specified diameter, 6 — fasteners for installing the optical spectrometer, 7 — optical spectrometer of a unique design [7].

in Fig. 2, *a*. Fig. 2, *b* presents the function of the silicon detector spectral sensitivity determined by comparing with the reference silicon photodiode calibrated at VNIIOFI.

Self-glow spectra were obtained for both samples. To correctly compare the self-glowing spectra of two samples, the background was subtracted, and the measured self-glowing intensity was divided by the optical bench instrument function at each spectrum point. Spectra obtained as a result of those transformations are presented in Fig. 3.

Spectrum of the ZrSiO<sub>4</sub>:Tb<sup>3+</sup> sample exhibits bands associated with transitions between the Tb<sup>3+</sup> levels; in the spectrum of the YPO<sub>4</sub>:Eu<sup>3+</sup> sample, bands associated with transitions between the Eu<sup>3+</sup> levels are observed. Luminescence bands of the two samples are observed in different optical ranges of the spectrum.

Using the setup described above (Fig 2, *a*), photocurrent ( $J_f$ ) produced by the detector under the impact of each crystal self-glowing was measured.

Photocurrents obtained for each sample are shown in the Fig. 3 insets. The averaged measured photocurrents were  $J_{f1} = 2.7 \cdot 10^{-8}$  A for the ZrSiO<sub>4</sub>:Tb<sup>3+</sup> sample and  $J_{f2} = 1.2 \cdot 10^{-7}$  A for the YPO<sub>4</sub>:Eu<sup>3+</sup> sample. However, to quantitatively compare optical radiation powers of both

samples, it is necessary to consider a number of parameters, first of all, the photodetector spectral function.

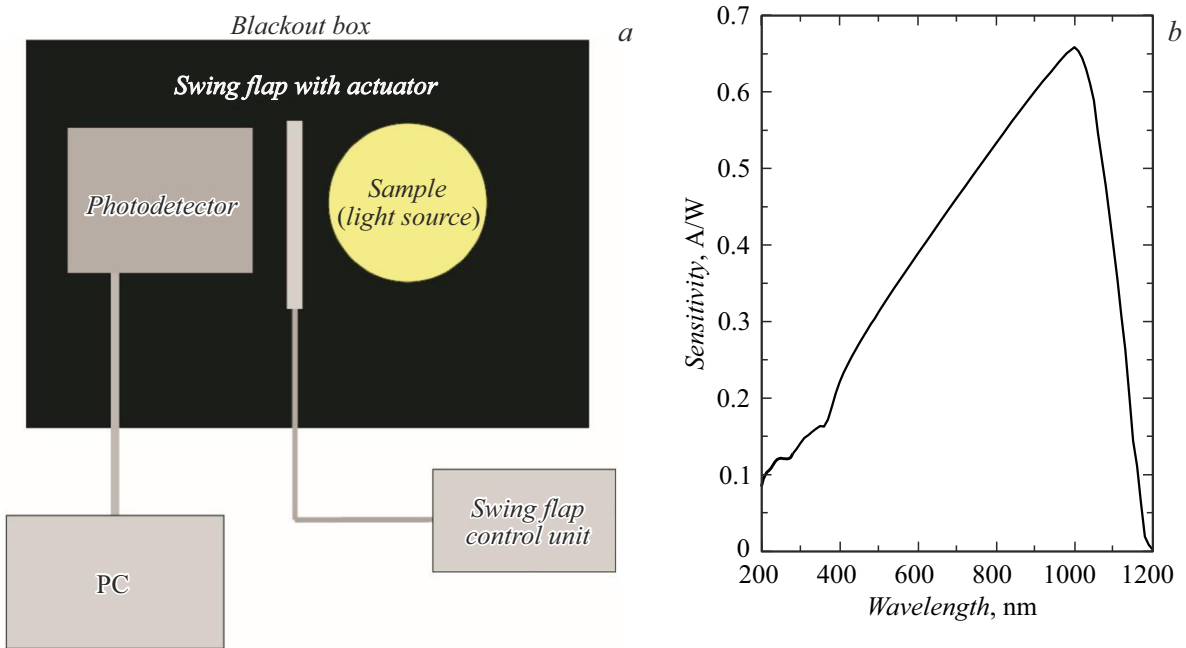
To find the total sample luminosity, it is necessary to take into account, among others, the geometry of sample arrangement in the photocurrent measurement setup. A fairly simple geometric model was created, which comprised a square photodetector and circular light source of non-zero size spaced by a distance greater than their characteristic dimensions. When all the parameters are taken into account, it becomes possible to determine the fraction of sample radiation getting on the detector:  $g \approx 0.021$ .

Power of the samples self-glowing was determined according to the following procedure. Energy contribution to the total energy luminosity of the sample from each spectrum range between adjacent points was calculated as

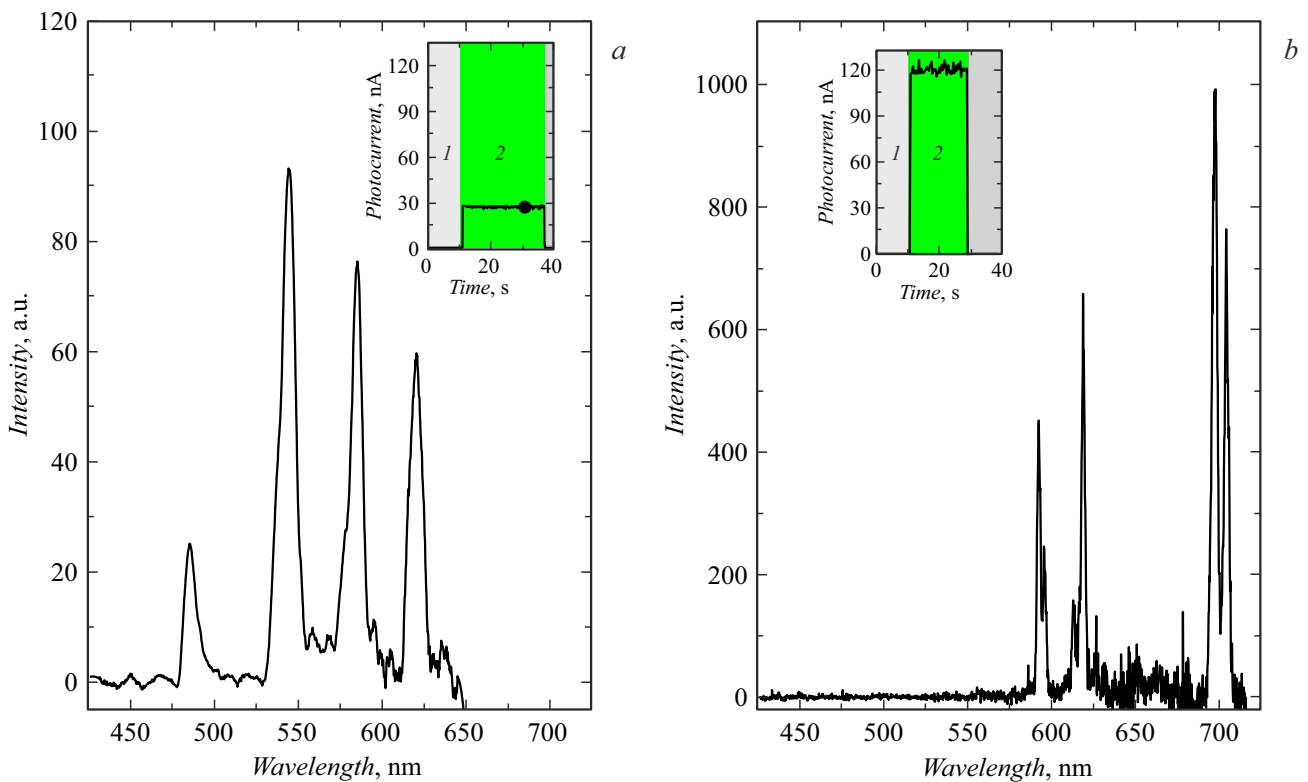
$$E(\lambda) = I(\lambda) \frac{hc}{\lambda}, \quad (1)$$

where  $\lambda$  is the wavelength [nm];  $I(\lambda)$  is the number of photons detected by the spectrometer FEU during 1 s for wavelength  $\lambda$  (taking into account the bench instrument function);  $h$  is the Planck constant;  $c$  is the light speed.

As shown in the Fig. 1 inset, the bench for measuring self-glowing spectra comprises an entrance and exit slits. The



**Figure 2.** *a* — layout of the setup for measuring optical power of self-glowing samples; *b* — spectral function of the silicon detector.



**Figure 3.** Self-glowing spectra of samples  $\text{ZrSiO}_4:\text{Tb}^{3+}$  (*a*) and  $\text{YPO}_4:\text{Eu}^{3+}$  (*b*). The insets present the time dependences of photocurrent  $J_f$  measured with a silicon photodetector in a light-proof box. Gray areas 1 — the detector is closed with a shutter, green areas 2 — the shutter is open (the color version of the figure is presented in the electronic version of the article).

**Table 2.** Calculations of luminosity for samples ZrSiO<sub>4</sub>:Tb<sup>3+</sup> and YPO<sub>4</sub>:Eu<sup>3+</sup>

Sample	Measured photocurrent $J_f$ , A	Photocurrent calculated from self-glowing spectra $J_t$ , A	Total optical power, W
ZrSiO <sub>4</sub> :Tb <sup>3+</sup> , <sup>238</sup> Pu	$2.7 \cdot 10^{-8}$	$3.6 \cdot 10^{-15}$	$3.6 \cdot 10^{-6}$
YPO <sub>4</sub> :Eu <sup>3+</sup> , <sup>238</sup> Pu	$1.2 \cdot 10^{-7}$	$1.6 \cdot 10^{-14}$	$1.2 \cdot 10^{-5}$

use of slits provided achieving high spectral resolution in the spectra presented in Fig. 3, but made the radiation intensity significantly decreasing on the way from the source to spectrometer. In addition, the source in the bench is located at a much greater distance from the spectrometer than from the silicon detector in the optical power measurement setup. The combination of these factors leads to that the theoretical optical power of the source, which can be obtained by summing energy contributions of each measured spectrum range, is only a small part of the real optical power of the source.

Next, by integrating the product of the silicon photodetector sensitivity function  $s(\lambda)$  [A/W] by function  $E(\lambda)$  (see formula (2)), we obtained photocurrent  $J_t$  which is to be produced by the photodetector if the latter is mounted in the optical bench for measuring self-glowing spectra in place of the spectrometer and the optical bench is assumed to be absolutely transparent for all wavelengths:

$$J_t = \int s(\lambda)E(\lambda)d\lambda. \quad (2)$$

What is important is that, in order to verify the correctness of the described method, the ratio between theoretical photocurrents  $J_{t2}/J_{t1}$  of the studied samples was compared with the experimentally obtained ratio  $J_{f2}/J_{f1}$  of photocurrents of these samples. The calculations showed that

$$\frac{J_{t2}}{J_{t1}} \approx \frac{1.6 \cdot 10^{-14}[\text{A}]}{3.6 \cdot 10^{-15}[\text{A}]} \approx 4.4, \quad \frac{J_{f2}}{J_{f1}} \approx \frac{1.2 \cdot 10^{-7}[\text{A}]}{2.7 \cdot 10^{-8}[\text{A}]} \approx 4.4, \quad (3)$$

which confirms the correctness of executed operations.

After that, the ratio between the current measured by the photodetector and theoretically calculated one ( $J_f/J_t$ ) was found for each sample. Average value of this ratio for two samples appeared to be  $R = 7.5 \cdot 10^6$ . This value can be used to directly determine the total optical power of any weakly glowing object whose luminosity is induced by external or internal excitation under the condition of complete repetition of the optical bench parameters: distance between the source and spectrometer, sizes of the entrance and exit slits, etc. Thus, total optical power  $P$  of the sample can be calculated as

$$P = \frac{R \int E(\lambda)d\lambda}{g}. \quad (4)$$

Table 2 presents the results obtained by applying the described technique to samples ZrSiO<sub>4</sub>:Tb<sup>3+</sup> and YPO<sub>4</sub>:Eu<sup>3+</sup>.

Thus, the paper reports the developed a technique for comparing optical radiation powers of samples with taking into account their radiation spectra. This makes it possible to compare the radiation optical powers of sources with complex radiation spectra; this is very important in working with materials activated by rare earth ions. Calculations carried out via the proposed technique showed that the total optical power of the ZrSiO<sub>4</sub>:Tb<sup>3+</sup> sample was  $3.6 \cdot 10^{-6}$  W, while that of the YPO<sub>4</sub>:Eu<sup>3+</sup> sample was  $1.2 \cdot 10^{-5}$  W. The bench and technique described in this paper may be used also for comparative analysis of other weakly glowing samples whose luminosity is induced by external or internal excitation.

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## Conflict of interests

The authors declare that they have no conflict of interests.

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