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Photoluminescent properties of aqueous solutions of triglycine sulfate of various concentrations

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Experimental results of studying photoluminescence of aqueous solutions of triglycine sulfate at different concentrations are presented. The excitation and emission spectra, characteristic wavelengths, and concentration dependence of intensity are established. The obtained data indicate the stability of luminescence centers and potential suitability of triglycine sulfate solutions for using them in scintillation detectors.

Keywords: triglycine sulfate, luminescence, spectroscopy, scintillator.

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Liquid scintillators are widely used in astrophysical research and reactor experiments with antineutrinos [1,2]. Development of up-to-date large-scale neutrino facilities enhances the need for large-volume detectors; this makes urgent the problem of searching for low-cost and highly efficient liquid scintillators. At present, such detectors employ as solvents aromatic hydrocarbons [2–8], mixtures of saturated aromatic hydrocarbons [9–11], and other organic compounds. Liquid scintillators are typically organic substances dissolved in organic solvents; however, their use is associated with a number of restrictions related to fire hazards and negative environmental impacts.

In this regard, research aimed at developing liquid scintillators based on aqueous solutions becomes of particular importance [12,13]. Such scintillators will have not only improved environmental characteristics but also a number of functional advantages, including the ability to detect the direction of the interacting particle motion. Despite their significant potential, water-based liquid scintillators have not yet gained wide application. Therefore, consideration of an alternative option for the liquid scintillator based on aqueous solutions is a promising task.

Crystalline triglycine sulfate (TGS) exhibits hydrogen-bonded ferroelectric properties, intense photoluminescence emission [12–15], and good water solubility. Under the excitation by ultraviolet radiation (310 nm), a broad luminescence band (300−700 nm) with the maximum at 442 nm was detected in the TGS crystal. Paper [13] discusses possible thermoluminescence mechanisms and shows that optical transition involving the SO_4^{2-} center occurs in the range of 300−400 nm. However, the difference in data on the excitation and emission wavelengths provided by different studies evidences the results' ambiguity. Papers [14,15] demonstrate the influence of doping additives to the TGS crystal on the phase transition optical properties and temperature. For instance, introduction of dye increases the phase transition temperature by about 50 °C and causes an increase in the intensity of blue-shifted luminescence [14]. In the case of doping with croconic acid $C_5H_2O_5$, additional absorption bands arise in the UV and visible spectrum regions, which are absent in pure TGS [15]. However, fluorescent properties of TGS aqueous solutions have not yet been studied. Thus, this paper presents new results of studying luminescence spectra of pure TGS crystals and aqueous solutions in a wide range of excitation and emission spectra with a view to creating a highly efficient scintillator for underground neutrino experiments.

The photoluminescence properties were studied using luminescence spectrometer PerkinElmer LS55. The spectrometer's photomultiplier tube (PMT) was optimized for the wavelength range of 250 to 800 nm, its slit width was adjustable within the range of 2.5 to 20 nm. The wavelength was measured accurately to ±0.5 nm; relative error of multiple measurements (6−10 repetitions) was $\sim 0.1\,\%$. The device was equipped with the FL WinLab software.

The samples for studying were fabricated from a high-purity TGS monocrystalline ingot and distilled water. The TGS aqueous solution concentrations varied from 20 to 80 g/l, which is significantly lower than the room-temperature TGS water-solubility limit equal to 150 g/l [16]. Therefore, no formation of crystalline nuclei was observed in the solutions, and these solutions were homogeneous phases.

The excitation and photoluminescence spectra were measured on the samples of a pure TGS crystal and its aqueous solutions with concentrations of 20, 40, 50, 60, 70 and 80 g/l in the wavelength range from 300 to 800 nm.

The excitation wavelength was selected based on the maximum intensity of excitation spectrum of the pure TGS crystal and its aqueous solution. Dispersion dependence of excitation intensity of the TGS crystal and its aqueous solution (Fig. 1) shows that the most prominent excitation maximum of the crystal is located in the ultraviolet spectrum region at the wavelength of 310 nm, which is consistent

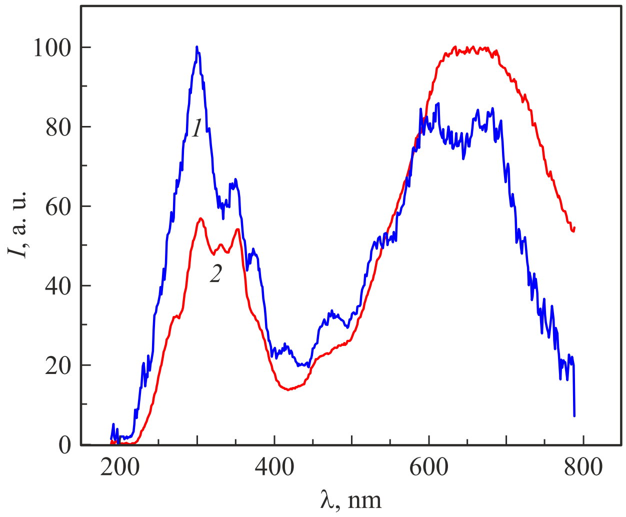

Figure 1. Excitation spectra of crystalline TGS (*1*) and TGS aqueous solution 50 g/l in concentration (*2*) under excitation at the wavelength of 310 nm.

with the data of [14]. Therefore, for measuring emission spectra in the standard scanning mode there were selected the fixed excitation wavelength of 310 nm and excitation monochromator slit width of 5 nm.

Fig. 2 presents the survey photoluminescence spectra of crystalline TGS and its aqueous solution 50 g/l in concentration under the excitation at 310 nm. As shown in Figs. 1 and 2, excitation and emission spectra of the crystalline and aqueous solutions are consistent with each other.

To reveal the degree of coincidence between the crystal and solution photoluminescence spectra, high-sensitivity synchronous scanning of the excitation and emission spectra was performed. The measurements were carried out at constant shift $\Delta\lambda = 10$ nm between the excitation and emission wavelengths during measurements in the range of 300–750 nm; the aqueous solution concentration was 80 g/l (Fig. 3).

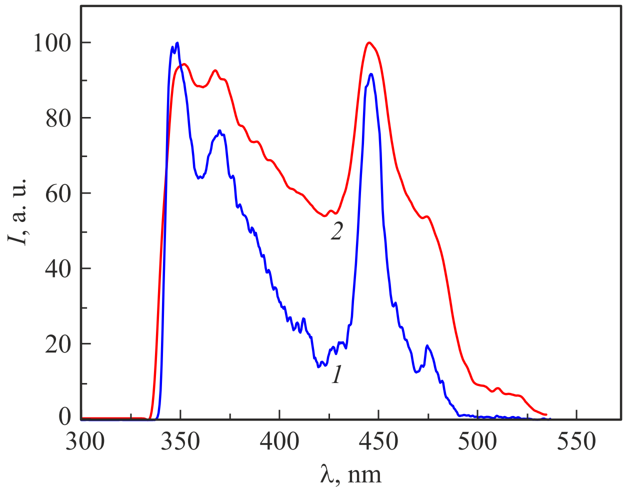

Figure 2. Photoluminescence spectra of crystalline TGS (*1*) and its aqueous solution 50 g/l in concentration (*2*) under excitation at the wavelength of 310 nm.

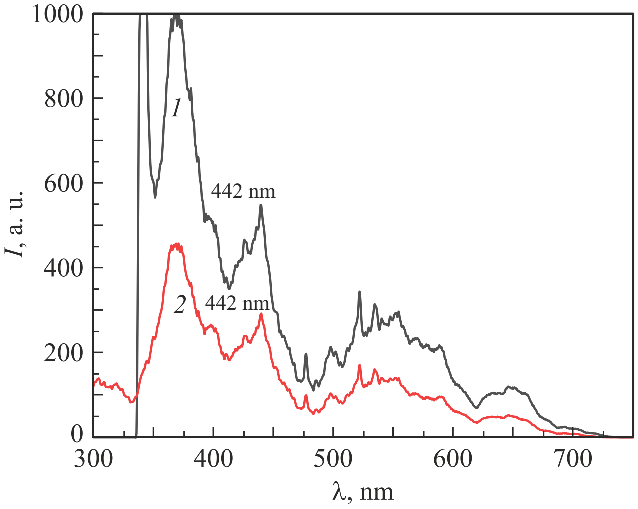

Figure 3. Emission spectra of the pure TGS crystal (*1*) and aqueous solution 80 g/l in concentration (*2*) obtained by synchronous scanning with fixed shift $\Delta\lambda = 10$ nm between the excitation and emission wavelengths during measurement.

The spectrum fine structure exhibits a complete coincidence of the crystalline sample's peaks with those of the aqueous solution; the peaks remain the same when the solution concentration changes. Apparently, this is due to that the electronic structure of TGS molecules in solution does not change with varying concentration in water. If new intermolecular interactions were to arise between TGS molecules in solution, this would lead to a shift in spectra. Apparently, the TGS molecule does not dissociate being dissolved in water, and two formula units that constitute the TGS crystal unit cell (including SO_4^{2-} and two glycine molecules) are preserved in the solution. Thus we may assume that the following energy pairs are retained in the solution: electron capture center–hole luminescence center and hole capture center–electron luminescence center [13]. The authors of [13] have noted that it is the centers involving SO_4^{2-} that may promote additional short-wavelength luminescence (300–400 nm) whose contribution remains significant in the aqueous solution.

To construct the concentration dependence of the intensity of TGS aqueous solutions, there was selected the 442 nm wavelength corresponding to the most prominent photoluminescence spectrum peak of the TGS aqueous solution. Fig. 4 presents the concentration dependence of the photoluminescence intensity of TGS aqueous solutions.

At those concentrations, the dependence is well approximable by a linear function, which is important for devices intended as scintillation detectors. The increase in concentration results in a deviation from linearity; first an increase and then a decrease in intensity is observed. Therefore, to make TGS aqueous solutions usable as short-

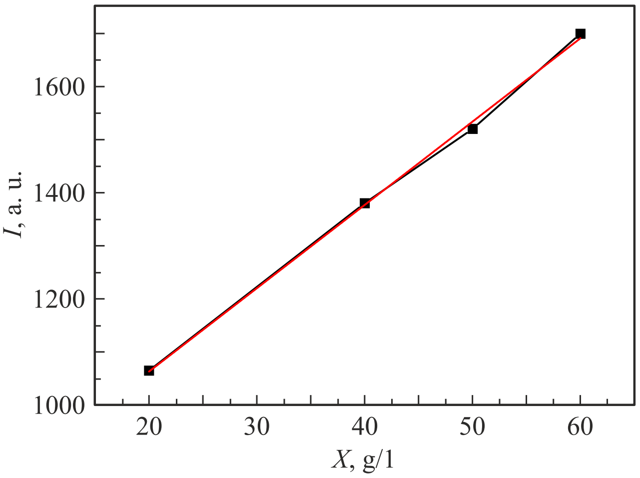

Figure 4. Concentration dependence of the photoluminescence intensity at the emission spectrum wavelength of 442 nm for distilled-water TGS solutions 20 to 60 g/l in concentration.

wave radiation receivers, concentrations ranging from 20 to 60 g/l should be used.

Analysis of photoluminescent properties of crystalline TGS and its aqueous solution has shown that the maximum excitation intensity of the TGS crystal and its aqueous solution is located at the wavelength of 310 nm. The maximum photoluminescence intensity of the TGS aqueous solution is observed at the wavelength of 442 nm. In this study, concentration dependence of the fluorescence intensity of aqueous solution with the TGS concentration varying from 20 to 60 g/l has been constructed for the first time. The results obtained may be used to create scintillation detectors for converting X-ray and gamma radiation into visible light. Luminescence of the TGS aqueous solutions makes it a promising material for medical imaging, diagnostics, dosimetry, biosensors, etc.

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Conflict of interests

The authors declare that they have no conflict of interests.

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