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Radioluminescence properties of nanocomposite scintillators with BaF₂ fillers

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Abstract. In this paper, studies of the luminescence properties of nanocrystalline BaF₂ samples synthesized by laser ablation and pulse electron beam evaporation method are presented. The measurements of X-ray excited luminescence (XEL) showed the dependence between luminescence intensity and the shape of the spectrum on the morphology and particle size. Also, studies of X-ray excited luminescence, decay curves and optical transmittance for nanocomposite materials containing BaF₂ nanopowder are presented. Barium fluoride nanopowder, obtained by pulsed electron beam evaporation method is characterized by a lower intensity than the initial microcrystalline powder, but at the same time, XEL spectrum of the nanocomposite material with this nanocrystalline filler is more intense, then that for nanocomposite material with initial powder.

1. Introduction

Organic scintillation materials are characterized by the simplicity of manufacturing technology, which allows to create detectors with large volumes and different sizes, but at the same time it is much lower in the efficiency of energy absorption and light yield in comparison with inorganic materials. In this connection, one of a promising direction for the development of scintillation materials is the creation of composite scintillation materials that combine the main advantages of organic and inorganic scintillators [1]. One of the main tasks in this case is the selection of inorganic filler. One of the most promising fillers is BaF₂, whose single crystal has a high light yield in the self-trapped exciton (STE) band with a maximum at 300 nm, a short decay time (with a decay time constant 0.9 ns [2] in the core-valence luminescence band CVL) with maximum at 222 nm, Also important that the value of the refractive index for barium fluoride close to corresponding value for organic materials used as a matrix. It is necessary to investigate dependence between particle size and radioluminescent properties. In our previous paper [3], we presented the results of a study of the luminescence properties of CaF₂ synthesized by laser ablation method [4]. For nanoscale BaF₂ synthesized by the chemical method it was shown that the luminescence intensity at the STE band is more critically dependent on the particle size than at CVL band [5]. At the same time, for a small particle size (with average particle size 20 nm), a decrease in the total luminescence intensity is observed. In addition to the properties of nanocrystalline particles themselves, it is important to understand the mechanisms of absorption, transformation and emission of energy between filler and organic matrix. In work [6]



studies of luminescent properties of nanocomposites based on nanopowder BaF₂, obtained by chemical method, placed in a polystyrene matrix are given. In this paper, we present studies of the radioluminescence properties of nanocrystalline BaF₂ samples obtained by laser ablation method (LA) [4] and pulsed beam electron evaporation method (PEBE) [7]. Also optical and radioluminescence properties of nanocomposite materials with epoxy resin as an organic matrix and nanopowders fillers are presented.

2. Samples and experiment

2.1. Samples preparation

All nanopowder samples were synthesized from microcrystalline powder (BaF₂@RAW) with average particle size 35 μm. Synthesis of all nanopowders was carried out at the Institute of Electrophysics UB RAS. The BaF₂@LA sample, with average particle size 64 nm, was obtained by laser ablation technique using a pulsed-periodic CO₂ laser with a peak radiation power of up to 11 kW and a pulse duration of 150-300 μs. The BaF₂@PEBE sample, with average particle size 3-5 nm, was produced by pulsed electronic beam evaporation method (PEBE) on “Nanobeam-2” installation equipped with electron gun, operated at 40kV with pulse length 100 μs. Preparation of samples of nanocomposite materials was carried out in the chemical laboratory of Scientific and Educational Centre “Nanomaterials and nanotechnologies”. The required amount of BaF₂ was added to the epoxy resin, which was then heated and stirred. Then hardener was added to the POPOP (1,4-bis (5-phenyloxazol-2-yl)benzene) solution in ethyl acetate. After this, all the ethyl acetate was evaporated, the remaining hardener with POPOP was added to the epoxy resin. The resulting mixture was stirred and degassed, and then left at room temperature for 24 hours to harden. Thus, several samples of a cylindrical shape with a base diameter of 30 mm and a height of 6 mm, with different wt% of POPOP and fillers prepared by the LA, PEBE methods and initial micropowder BaF₂ were obtained. The marking and description of the samples are given in table 1.

Table 1. Investigated samples of nanomaterials.

| Sample name | POPOP, wt% | BaF ₂ – filler type |
|-------------|------------|--------------------------------|
| N@P0002@NF | 0.0002 | - |
| N@P0018@NF | 0.0018 | - |
| N@P018@NF | 0.018 | - |
| N@P018@PEBE | 0.018 | PEBE, 0.87 wt% |
| N@P018@RAW | 0.018 | RAW, 0.87 wt% |
| N@P018@LA | 0.018 | LA, 0.87 wt% |

2.2. Experimental setups

The X-ray excited luminescence (XEL) spectra of BaF₂ powders were performed with ASSI ROPCM setup (Automatic System for Science Investigation of Radiation and Optical Properties of Condensed Matter) equipped with photomultiplier FEU-106 (operated at 1800 kV with 200–800 nm wavelength range) and MDR-2 monochromator with 1200 and 600 grooves/mm diffraction gratings. Excitation was provided by X-ray source URS-1,0 with BSV-2 X-ray tube (W-anode, 30 kV, 14 mA). For investigation of luminescence decay properties the samples emission was excited by MIRA-2D pulse electron gun (energy 150 keV with pulse duration about 20 ns) and recorded by photomultiplier FEU-97 and oscilloscope Tektronix TDS5034B with 50 ohm input load. Equipment used for decay curves investigation has time constant measurements limit at 25 ns. Measurement of the optical absorption spectra of the samples was performed using an optical spectrometer Shimadzu UV-2450.

3. Experiment and discussion

3.1. BaF_2 luminescence measurements

The X-ray excited luminescence spectra (XEL) and decay curves measured at the 300 nm band for all nanopowders are shown in figure 1. Spectra of all samples contain asymmetric band with a maximum of 300 nm which can be associated with STE luminescence. The weak band with a maximum at 222 nm is very well known a CVL band.

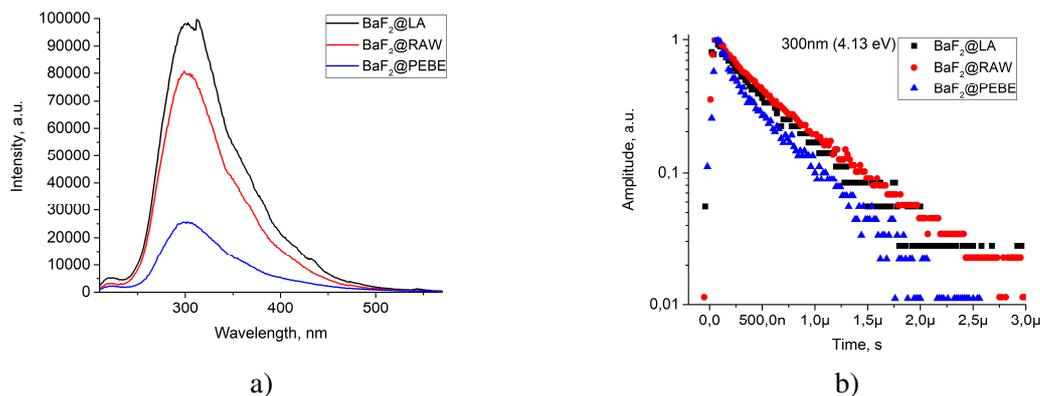


Figure 1. XEL spectra of nanocrystalline samples (a) and decay curves measured at 300 nm (b).

The sample with the smallest average particle size BaF₂@PEBE is characterized by the lowest luminescence intensity. The maximum luminescence intensity is observed for a nanocrystalline powder obtained by laser ablation method (BaF₂@LA). Also all samples are characterized by approximately the same intensity of the CVL band. Measurements of the decay curves at the 300 nm band indicate the presence of two luminescence components in BaF₂@PEBE and BaF₂@LA samples with decay time constants values of 40 ns and 500 ns respectively.

3.2. Nanocomposite materials luminescence measurements

Figure 2 shows the optical transmission spectra and XEL spectra of the nanocomposite materials.

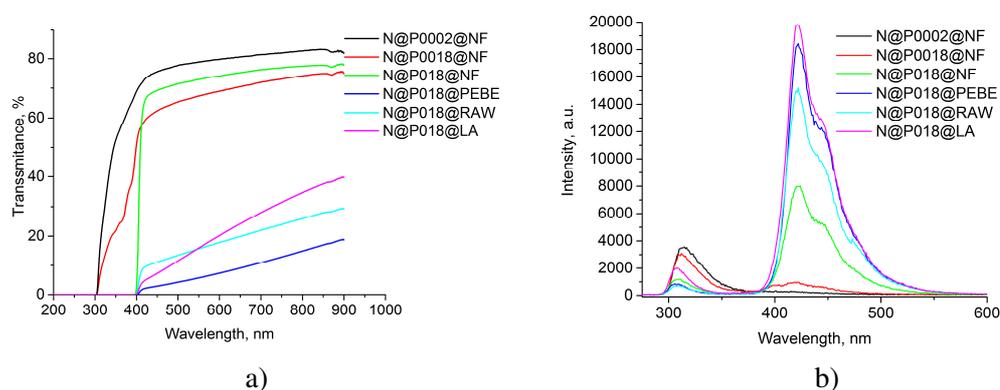


Figure 2. The optical transmission spectra (a) and XEL spectra (b) of the investigated nanocomposite samples.

The absorption edge of the epoxy resin is near 300 nm (figure 2a). With an increase in the POPOP concentration, the «sorption edge of the samples shifts to 400 nm. Samples with inorganic fillers have less optical transmission. For all samples in the XEL spectrum (figure 2b), a band with a maximum in

the 310 nm region is observed. At the same time, for samples with a higher concentration of POPOP, there is a small shift to the region of smaller wavelengths (N@P018@NF, N@P018@PEBE, N@P018@RAW, N@P018@LA). The luminescence in this band is due to epoxy resin. As the concentration of POPOP increases, the XEL intensity decreases. It's caused by absorption maximum of POPOP in this range [8]. A large luminescence intensity value for N@P018@LA sample can be associated with an increase in the luminescence intensity of the STE BaF₂@LA band observed in figure 1a. The luminescence bands with maxima in the region of 421 nm and 450 nm are associated with the emission of POPOP [8]. This maximum is practically not observed for the samples N@P0002@NF, N@P0018@NF and obvious for N@P018@NF, N@P018@PEBE, N@P018@RAW and N@P018@LA. Samples containing inorganic additives are characterized by a higher intensity of these bands. The luminescence intensity for a sample with a microcrystalline filler (N@P018@RAW) in the 420 nm band is two times higher than for a sample with a similar concentration of POPOP without a filler (N@P018@NF). N@P018@LA sample has the highest luminescence intensity which agrees with the data obtained for the nanopowder (figure 1a). But at the same time, the nanocomposite material with BaF₂@RAW filler (N@P018@RAW) exhibits a lower intensity than the nanocomposite material with BaF₂@PEBE (N@P018@PEBE). This may be due to the fact that the processes of energy transfer between the filler and the spectrum shifter are more effective for particles with a smaller size. Measurement of the decay curves (figure 3) in all bands reveals only one component with average time decay constant about 30 ns associated with the luminescence of POPOP. This experimental result requires additional study.

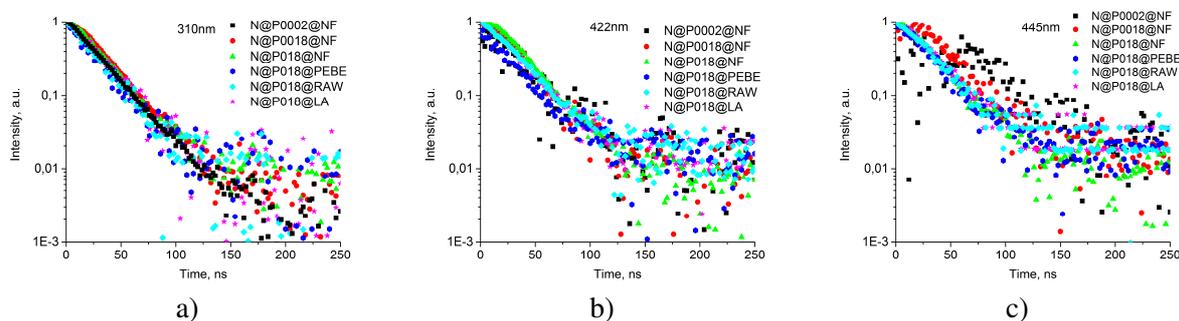


Figure 3. Decay curves measured at 310 (a), 422 (b) and 445 nm (c) for nanocomposite samples.

4. Conclusion

The carried out researches show that the BaF₂ nanopowder synthesized by laser ablation method, with an average particle size of 64 nm, characterized by highest intensity of luminescence. The sample of BaF₂@PEBE is characterized by a lower intensity than the microcrystalline powder, but at the same time, XEL of the nanocomposite material with the BaF₂@PEBE filler is more intense. This may indicate a more efficient process of energy transfer from nanocrystalline BaF₂ particles to the POPOP. The obtained data of XEL spectra are in good agreement with the data obtained in [6] for nanocomposite materials based on BaF₂ nanopowder obtained by the chemical method. The difference in the shape of the spectrum is due to the various organic matrices used.

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