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Research articles

Thermoradiation modification of nanostructure of potash-alumo-boron glasses with iron oxide additives

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ABSTRACT

This paper presents the results of thermoradiation modification of nanostructure of potash-aluminum-boron glasses with iron oxide additives studied by optical spectroscopy. The differential spectra of the glass samples have jumps at 240, 320 and 370 nm. The physical mechanism of their formation is suggested. The obtained results show the way for the nanostructure modification useful for magneto-optical and dosimetric applications of the glasses. Valence-coordination transition of iron ions induced by irradiation at elevated temperatures can be used as switching effect in quantum computers.

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1. Introduction

Recently the magnetic nanosystems have caused the interest due to prospects of their use in magneto-optics, dosimetry of ionizing radiation and quantum computer technology [1–3]. Despite their practical value, optical properties of the potash-aluminum-boron glasses (PAB) under irradiation were studied insufficiently. Even more complicate theoretical interpretation is required for thermoradiation phenomena, which allow to modify nanostructure [4,5] for creation of the functional devices and quantum computers.

This work is aimed at revealing thermal, radiation and thermoradiation effects, using phenomenological models of processes in multicomponent glasses with nanostructures and studying optical characteristics of the treated samples.

2. Experimental samples and technique

The glasses of potassium-aluminum-boron oxides PAB-50 ($25K_2O.25Al_2O_3{\cdot}50B_2O_3$) in mol unit and PAB-55 ($22.5K_2O.22.5Al_2O_3{\cdot}55B_2O_3$) both pure and doped by iron oxide were chosen as perspective material for magneto-optics and nanoelectronics. Iron oxide Fe_2O_3 was added with concentrations from 0.1 to 3.0 mass.% over 100% of basic composition of PAB glass. These mixtures were melted at temperature of (1603 ± 5) K.

Samples for optical measurements had the polished area of 1 cm^2 and thickness of (1 ± 0,05) mm. Thermoradiation treatments (TRT) were carried out during 2 h at several elevated temperatures of samples 323, 423, 473, 523, 573 K in the ^{60}Co gamma field at the dose rate of 236 R/s. All samples have received an identical dose of gamma irradiation of $1{,}7{\cdot}10^6$ R. Original installation has been created for TRT with a temperature adjustment accuracy ±0,25 K, regulated heating rate and time of irradiation. Spectra of absorption (transmission) were run at the one-beam spectrophotometer CF-56 in the range from 190 to 1100 nm with margin error of ±3%. Differential or differential ranges $\Delta D = D_1 - D_{treat.}$ were obtained from a series of spectra where D_1- optical density spectra for untreated samples, $D_{treat.}$ - optical density spectra after thermal or thermoradiation treatments.

3. Results and discussion

At first we studied the dependence of optical spectra of PAB-55 glasses on Fe_2O_3 additive concentration varied within 0.1; 0.25; 0.5; 1.0; 1.5; 2.0; 2.5 and 3.0 mass.% above 100 mol%, (Fig. 1, spectra 1 – pure, 2 – 9 doped respectively). The analysis of these spectra shows: for pure glass (1) the edge of absorption at the level D = 1

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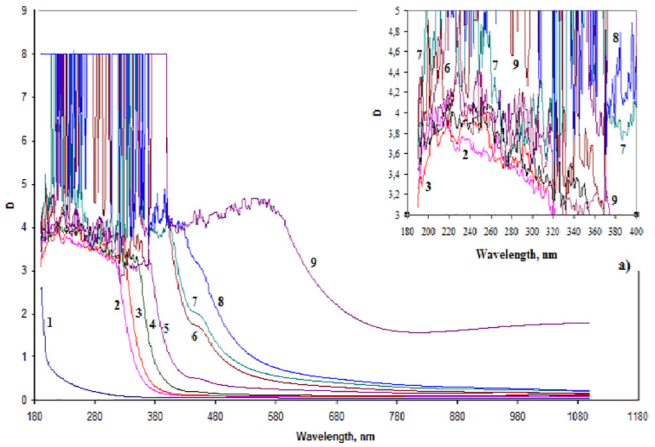

Fig. 1. Concentration dependence of absorption spectra of PAB-55 glasses at 300 K: 1 – 0; 2 – 0,1; 3 – 0.25; 4 – 0,5; 5 – 1,0; 6 – 1,5; 7 – 2,0; 8 – 2,5; 9 – 3,0 mass.% Fe_2O_3; (a) – UV fragments of the spectra at Δλ = 180–400 nm and ΔD = 3–5.

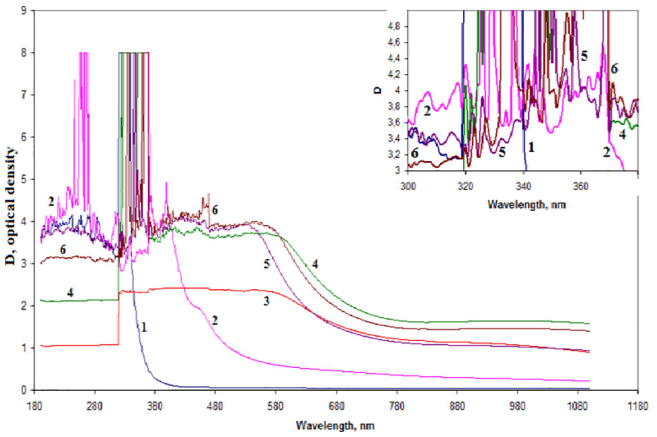

Fig. 2. Absorption spectra of the pure and doped PAB-50 glasses ^{60}Co gamma irradiated at the dose rate of 236 R/s and temperatures: 1 – pure glass at 323 K; 2-doped with 2.0 mass.% Fe_2O_3 at 323 K; doped glasses at 423 K (3); 473 K (4); 523 K (5); 673 K (6).

begins at 200 nm, and the band of transparency begins with 300 nm to 1100 nm. This spectrum was taken as the standard for reference undoped sample. Even insignificant addition of 0.1 mass.% Fe_2O_3 shifts the edge of absorption down 140 nm at the level D = 1 (spectrum 2). With increase in the additive up to 1.0 mass.% there appears a weak absorption band at 440 nm corresponding to Fe^{3+} ions. Further shift at the level D = 2.5 sharply increases due to formation of clusters $[Fe_2O_3]^n$ (Table 1), and influence of clustering begins at 2.5 mass.% Fe_2O_3. The absorption edge shift in case of 3.0 mass.% stands at 456 nm and the contribution from light scattering at charges clusters reveals clearly (spectrum 9, Fig. 1). Moreover, very broad band at 780–1100 nm is identified as $[FeO]^x$, corresponding to $[Fe^{2+}$— $Fe^{3+}]$ interactions. Such non-linear changes demonstrate clustering $[Fe_2O_3|Fe_3O_4]$ (440–700 nm) (hematite-magnetite) and agree with results of ours [2,4] and others [3,5,6].

The estimated sizes of nanoparticles in structures $[FeO]^x$, $[Fe_2O_3]^y$ and $[Fe_3O_4]^z$ of [9–10] [1.2–9] – X (nm); [1–8] – Y (nm); [3.2–27] – Z (nm), where X, Y and Z are the model calculated values of number of the main structure units in crystal or amorphous glass [2]. In particular, we estimated the sizes of nanoparticles of Fe^{2+} ions in tetrahedral structure and Fe^{3+} ions in octahedral structure: $X_{min} = Y_{min} = 6$; $X_{max} = Y_{max} = 16$, and obtained for [FeO] structure from [7.5–54] nm to [20–144] nm; for $[Fe_2O_3]$ structure from [6.3–48.3] to [16.8–128.8] nm; for ferromagnetic component $[Fe_3O_4]$ with the maximum sizes from 52 nm to 432 nm.

As the data on separate thermal and radiation treatments have been discussed earlier [4], we were focused in this work on thermoradiation treatment within 2 h at the dose rate of 236 R/s of PAB- glasses doped with Fe_2O_3 and also differential absorption spectrum. As have shown experimental data TRT of PAB glasses with 1.0 mass.% Fe_2O_3 actually weak changes the spectra at temperatures of 473, 523, 573 °C at the UV-edge 380–400 nm and in all visible range 400–900 nm, including near IR 900–1100 nm. It means the absence of clustering at these temperatures and concentration of Fe_2O_3. At 2.0 mass.% Fe_2O_3 (Fig. 2, spectrum 1 – non-irradiated sample) for PAB-50 glass processed in the gamma field at temperatures of 323, 423; 473; 523; 573 K, corresponding to spectra 2–6. It is shown, that the absorption band at 420 nm, according to Bates theory of field ligands, corresponds to structure – $[Fe^{3+}O_6]$ and increases with TRT and at the same time there is a jump band in the range 300–580 nm which corresponds, apparently, to Fe_3O_4. In the references, both bands continue growing up to the temperature of 573 K.

Figs. 3 and 4 show differential absorption spectra of PAB-50 glasses with 1.0 and 2.0 mass.% Fe_2O_3 depending on gamma irradiation temperature from which the spectrum of pure PAB-50 was subtracted. Regions with negative absorption for Fig. 3 have a width of 10 nm whereas for Fig. 4 – 20 nm, which correlates with 2 times increase in concentration of Fe_2O_3. As for Fig. 5, in - the first, separation of clusters of the Fe_3O_4 type corresponds to the absorption band at 400–680 nm.

Unlike Fig. 3 and 4 it is selected the negative absorption bands in case of switching of wavelength λ_s = 320 nm stretching to 680 nm with a width of 240 nm here. Besides, the second band of the negative absorption with a width of 35 nm, Fe_3O_4 which is actually relates to transition at the point of 370 nm (switching effect) $[Fe_2O_3] \leftrightarrow [Fe_3O_4]$.

The special program (code) of processing [7], decomposing initial nonlinear curve dependences of D or ΔD (Figs. 1–5) to symmetric Gaussian curves, determined parameters of Gaussian curves: magnitude A_i, wavelength at the maximum X_j, half width $\Delta X_{j,1/2}$ for each.

The program is expected determination of the sizes and quantity of nanoparticles in the given range of lengths of waves of graphical representation. As on Smakula's formulas for each Gauss-curves number of particles it is proportional to the area under a curve, adding

Table 1
Shift of edge of fundamental absorption for PAB-55 glass.

	At the level D = 1				
Fe_2O_3, mas.%	0	0,1	0.25	0,5	1,0
Δλ, nm	0	140	152	172	200
	At the level D = 2,5				
Fe_2O_3, mas.%	1,5	2,0	2,5	3,0	
Δλ, nm	224	228	272	456	

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