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SILICATES OF ALKALINE-EARTH ELEMENTS ACTIVATED RRE-IONS – PROMISING MATERIALS FOR SCINTILLATION AND LUMINESCENCES APPLICATIONS

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Summary. *The composition of the glass scintillation materials was elaborated. The effects of rare earth ions upon spectral and luminescent properties of the glasses and glass ceramics were established.*

Introduction. There is a strong demand for development of new inorganic luminescent materials which nowadays shows tremendous progress through nano-engenering of the materials. Among variety of inorganic materials silicates are seems to be prospective to construct new phosphors. They find numerous application as luminescent materials in a crystalline phase. The rare earth (RE) containing single-crystalline RE_2SiO_5 and $RE_2Si_2O_7$ doped with Ce-ions are good scintillation materials [1]. However, in a glass form, silicates possess several limitations, especially lower thermal conductivity and difficulty to stabilize activating ions in a needed valent state. Modification of the glass net with different additives, especially La-ions, allows doping however, stabilization of needed valent state still remains the problem. Glass ceramic materials possess several advantages combining the properties of crystals and glass, especially the high thermal conductivity. Their production is an industrial domain which promotes future development of the materials since sixties [2]. However, at the development of luminescent glass ceramics materials several matters still have to be answered. They are: transformation of the luminescent properties at the material crystallization; conservation of the valent state of the luminescent ions like Ce^{3+}, $Eu^{2+,3+}$ and Tb^{3+}; search of the most advanced composition for the glass ceramic to create high efficiency phosphors.

Materials and Methods. Development of the laboratory technology of producing silicate glass $xMO–ySiO_2$ (M = Mg, Ba, Ca, Sr; x = 1, 2, 3; y = 1, 2, 3) with stoichiometric composition and to characterization of their luminescent properties were the aim of our research. Moreover, the MO–SiO_2 (M = Mg, Ba, Ca) systems were choosed due to their capability to be manufactured in several states: amorphous, as glass-ceramics, and in a crystalline form.

$xMO–ySiO_2$ (M = Mg, Ba, Ca, Sr; x = 1, 2, 3; y = 1, 2, 3) glasses were obtained by two different approaches – solid state and modificated sol-gel.

By solid state approach we mean the heat treatment of a mixture of powders of MCO_3 (M = Mg, Ba, Ca, Sr) and SiO_2. The synthesis was performed at maximum temperature 1450° C in the gas furnace (furnace atmosphere contains a high concentration CO) for 2 h. Obtained samples were annealed at 500 °C for 4 h in the muffle furnace.

The modificated sol-gel approach was lied in heat treatment of precursor obtained by hydrolysis of TEOS in the presence of thin MCO_3 powder (M = Mg, Ba, Ca, Sr). All components were taken in a stoichiometric ratio. Nobertherm LHT 04/18 (up to 1800°C) oven has been used to prepare glass samples in reducing, neutral and oxidizing atmosphere.

Glass-ceramics has been obtained by heat treatment of mother glass in Nobertherm LHT 04/18 oven at different temperatures. The heat treatment duration and its temperature were different for different mother glass composition.

Results and Discussion. The samples of the glasses and glass-ceramics based on stoichiometric compounds with composition $xMO–ySiO_2$ (M = Mg, Ba, Ca, Sr; x = 1, 2, 3; y = 1, 2, 3) were obtained in present work. Synthesis conditions (glass transition temperature), crystallization ability and luminescence properties of obtained glasses depend on the $MO:SiO_2$ (M = Mg, Ba, Ca, Sr) ratio.

The influence of glass composition and crystallization conditions on photoluminescence of the glasses was studied by comparing the photoluminescence spectra, spatial distributions of photoluminescence parameters, and quantum efficiency.

Tendency for scintillation for stoichiometric compounds in $BaO–SiO_2:Ce$ system was established. The highest light yield was registered for glass-ceramics with composition $BaO–2SiO_2:Ce$ (λscint. = 440 nm). Radiation hardness for these compounds has been studied [3]. Possibility of activating of $BaO–SiO_2$ system by Ce^{3+}, Eu^{3+} and Tb^{3+} ions was demonstrated (Figure 1).

a b c

Figure 1.Developed glass ceramics samples obtained with different RE activators (a – Ce^{3+}, b – Tb^{3+}, c – Eu^{3+}) at excitation with 390 nm LED light

It was observed that the quantum efficiency of the glass ceramics is higher than that of the corresponding glass. The investigation of spatial distributions of photoluminescence parameters with submicron spatial resolution revealed that heat treatment of the glasses results in a decrease in sample homogeneity. Needle-like structures were observed.

The compounds in the system $CaO–SiO_2:Ce$ are characterized by absence of scintillation at saving high photoluminescence intensity. The bands of excitation luminescence at 340 nm and luminescence at 450 nm are making these compounds promising for applying as a luminescence shifter in combination with CeF_3 scintillation crystalls.

The laboratory production technology of RE doped glass and glassceramics has been developed. It has been established that the investigated systems have tend to the homogeneous nucleation. The crystallites sizes in these systems depend on the glass annealing temperature. Developed glass-ceramic systems in dependence of the composition can be used as a promising material for use as a scintillators or high luminosity phosphors with excitation in UV spectral region.

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