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Optical properties and generation of white light in Dy^{3+}-doped lead phosphate glasses

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Dy^{3+}-doped lead phosphate (PbPKANDy: $P_2O_5+K_2O+Al_2O_3+PbO+Na_2O+Dy_2O_3$) glasses were prepared by melt quenching technique and their optical properties have been studied. Judd-Ofelt parameters have been evaluated for 1.0 mol% Dy_2O_3-doped lead phosphate glass and inturn derived radiative properties for excited luminescent levels of Dy^{3+} ions. The yellow-to-blue emission intensity ratios and CIE chromaticity coordinates were calculated which have been used to evaluate white light emission as a function of the activator (Dy^{3+}) ion concentration. The observed non-exponential decay nature and quenching of lifetime for higher Dy^{3+} ion concentration ($\geq$1.0 mol%) have been attributed due to energy transfer of dipole–dipole type between excited and unexcited Dy^{3+} ions. The PbPKANDy glasses exhibit better luminescence properties which are suitable for generation of white light.

1. Introduction

Rare earth (RE)-doped glasses have attracted persistent research interest as they are easy to shape and have high transparency, low production cost, and better thermal stability. These glasses have been playing significant role for the development of optical devices including optical fibers and amplifiers, visible and infrared solid state lasers, waveguides, display devices, etc. [1–3]. Though different spectroscopic characterizations have been studied by the modification of chemical composition to improve the performance of optical devices, still there is a demand for new host materials with higher efficiency. Among different glass hosts, oxide based phosphate glasses have unique physical and chemical properties, which render them useful for a wide range of practical applications. Unfortunately, their relatively poor chemical durability precludes their practical utilization. Therefore, it has been noticed that the addition of metal oxides (MO) such as ZnO or PbO, resulting in the formation of M–O(–P) bonds, leads to improvement of the chemical durability and thermal stability of the phosphate glasses and enhance the radiative transition rates of RE^{3+} ions [4–7]. Hence, lead based phosphate glasses can be considered as promising materials for applications in optoelectronics.

Among the RE ions, trivalent dysprosium (Dy^{3+})-doped glasses have been considered as promising materials for two-color phosphors and white light emission since Dy^{3+} ion possesses intense emissions at blue (486 nm, ${}^4F_{9/2}\rightarrow{}^6H_{15/2}$) and yellow (576 nm, ${}^4F_{9/2}\rightarrow{}^6H_{13/2}$) regions. Further, it is well known that the ${}^4F_{9/2}\rightarrow{}^6H_{13/2}$ transition of Dy^{3+} ions is hypersensitive and therefore its intensity strongly depends on the nature of the host, whereas the intensity of magnetic-dipole allowed ${}^4F_{9/2}\rightarrow{}^6H_{15/2}$ transition is less sensitive to the host. Hence, at a suitable environment, the intensity ratio of these yellow to blue (Y/B) transitions will be tailored such that the Dy^{3+}-doped materials would generate white light.

In this work we report the optical properties of lead phosphate glasses containing different concentrations of

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Dy^{3+} ions. The Judd-Ofelt (JO) intensity parameters have been determined from the absorption spectrum, which are used to evaluate the radiative properties such as spontaneous transition probability, luminescence branching ratio, radiative decay time and stimulated emission cross-section for the $^4F_{9/2}$ emission level of Dy^{3+} ion. The calculated radiative properties derived from JO theory are compared with the experimental results obtained from emission spectra. The Y/B ratios and color coordinates have been calculated and the utility of the present glasses for white light emission has also been discussed. The influence of dopant concentration on the lifetimes is presented as well.

2. Experimental details

Lead phosphate glasses with composition in mol%, $44P_2O_5+17K_2O+9Al_2O_3+(24-x)PbO+6Na_2O+xDy_2O_3$ (x=0.1, 0.5, 1.0 and 2.0 mol% referred as PbPKANDy01, PbPKANDy05, PbPKANDy10 and PbPKANDy20, respectively) were prepared by the conventional melt quenching technique using reagent grade $Al(PO_3)_3$, KPO_3, PbO, Na_2CO_3 and Dy_2O_3 as starting materials. About 20 g of the batch composition was thoroughly ground in an agate mortar and this homogeneous mixture was taken into a platinum crucible and heated in an electric furnace at 1150 °C for 1 h 30 min. The melt was air quenched by pouring it onto a preheated brass mold and kept for annealing at 380 °C for 12 h in order to remove thermal strains and then slowly allowed to cool to room temperature (RT). After that, these glass samples were polished to get good transparency and flat surfaces for optical measurements.

Refractive index (n) measurements were performed using an Abbe refractometer at sodium wavelength (589.3 nm) with 1-bromonapthalene ($C_{10}H_7Br$) as contact liquid. The density (d) was measured by Archimedes' principle using water as an immersion liquid. All these measurements were carried out at RT. The physical properties of PbPKANDy10 glass are presented in Table 1.

Optical absorption spectrum in the range of 340–1900 nm was recorded using Perkin Elmer Lambda-950 spectrophotometer. Emission spectra were obtained by exciting the samples with the 457.9 nm line of Ar^+ laser. The fluorescence decay rates were detected using a mechanical chopper with a multi-channel scalar interfaced to a personal computer that recorded and averaged the signal at RT.

Table 1
Physical properties, average nephelauxetic ratio ($\overline{\beta}$) and bonding parameter (δ) for PbPKANDy10 glass.

Properties	PbPKANDy10
Optical path length, l (cm)	0.182
Density, d (g/cm^3)	3.680
Concentration, N ($\times 10^{20}$ ions/cm^3)	2.970
Refractive index, n	1.617
$\overline{\beta}$	1.005
δ	-0.457

3. Results and discussion

3.1. Absorption spectra

The absorption spectra of RE^{3+} ions serve as a basis for understanding their radiative properties. Absorption spectrum of PbPKANDy10 glass was originated from the $^6H_{15/2}$ ground level to various excited levels belonging to the $4f^9$ electronic configuration of the Dy^{3+} ion (see Fig. 1). The observed free-ion energy levels involved in $4f^9$–$4f^9$ transitions are analysed and assigned by means of the free-ion Hamiltonian (H_{FI}) model [8–12] from the absorption and emission spectra following the same procedure reported in our earlier work [12–14].

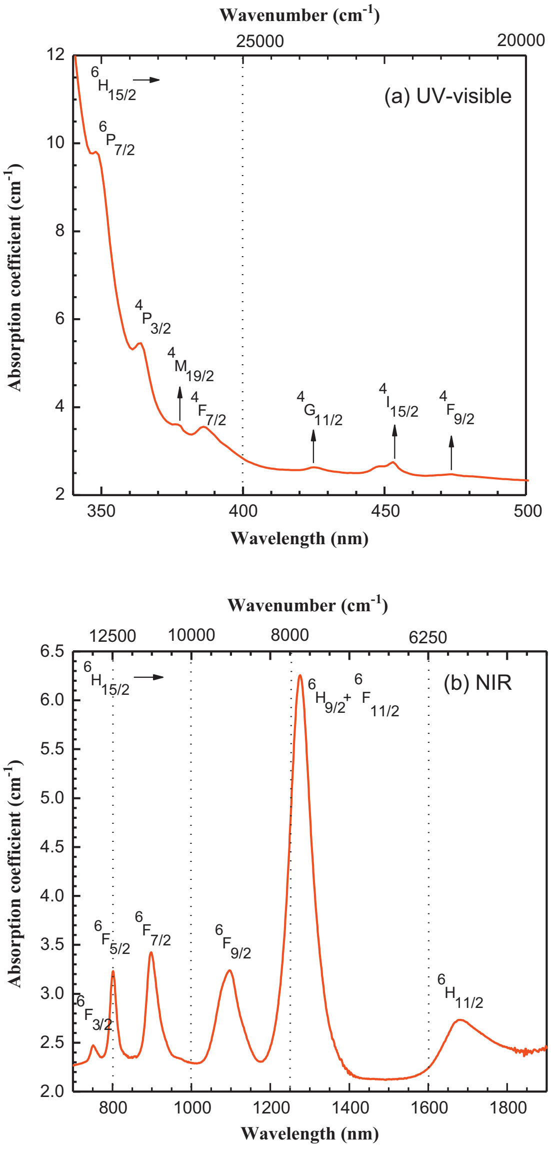

Fig. 1. Optical absorption spectra of PbPKANDy10 glass in the regions.