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X-ray induced effects in Sm^{3+}-doped $ZnO-P_2O_5$ glass for radiation measurements

Go Okada[a], Hirokazu Masai[b], Aya Torimoto[b], Safa Kasap[c] and Takayuki Yanagida[a]

[a]*Graduate School of Materials Science, Nara Institute of Science and Technology, 8916-5 Takayama-cho, Ikoma, Nara 630-0192, Japan*
[b]*Institute of Chemical Research, Kyoto University, Gokasho, Uji, Kyoto 611-0011, Japan*
[c]*Electrical and Computer Engineering, University of Saskatchewan, 57 Campus Dr., Saskatoon, SK S7N5A9, Canada*

We have studied X-ray induced effects on $ZnO-P_2O_5$ glass doped with Sm^{3+} ion. The glass was synthesized by a conventional melt-quenching method in an ambient atmosphere. We have investigated the following three different effects induced by X-ray irradiation in this glass, which can be used for radiation sensing applications. First, although the as-prepared glass sample is highly transparent and homogeneous, upon irradiation with X-rays, the sample becomes slightly brownish due to an appearance of color centers induced by X-ray irradiation which can be measured as an optical absorption in the UV-Vis range. By utilizing the extent of absorption change by X-ray irradiation, it is possible to measure the incident radiation dose from about 0.5 to over 20 Gy. Second, with the Sm^{3+} ion included as a luminescent centre, the glass sample emits orange-red light upon X-ray irradiation. The integrated light emission is proportional to the incident X-ray dose, hence it can be used as a scintillator plate for online radiation monitoring. The confirmed exposure dose detection range for this use is approximately 1 -300 R. Last, when an irradiated sample is heated at elevated temperatures from 100 to 400 °C, it shows a thermally-stimulated luminescence (TSL). The emission intensity increases with the accumulated X-ray dose delivered prior to the measurement, so our sample has a function to act as a TSL dosimeter as well. Over the radiation dose range we have tested, the X-ray induced effects above show linear response against the incident dose. The linear response is of particular importance for dosimetry applications since it enables us to calibrate the response of sample to radiation dose with high accuracy.

Key words: Samarium, Zinc phosphate glass, X-ray

Introduction

Ionizing radiations, e.g. X-rays, γ-rays, neutrons etc., have been used in a wide range of applications such as radiography, cancer therapy, nuclear physics, and so on. Since radiations are "invisible", it always requires radiation sensing techniques, which vary depending on the type of application it is used for. Inorganic phosphors are often used to measure high-energy radiations since these phosphors convert the incident radiation photons and high energy particles into light, which can be indirectly detected using conventional photodetectors. These phosphors may be categorized into two types which are often termed as scintillator and storage phosphor. The former converts the incident radiation into photons almost instantly [1] whereas the latter stores the absorbed radiation energy in the form of photoelectrically generated charges trapped locally. The trapped charges can then be detrapped by stimulation (either optically or thermally) followed by optical emission. Such emission by optical stimulation is termed as optically-stimulated luminescence (OSL) [2] while that by thermal stimulation is the so-called thermally-stimulated luminescence (TSL) [3].

In this research, we have studied X-ray induced effects in a Sm-doped zinc-phosphate glass for radiation measurements. The zinc-phosphate glass studied in this work is of interest since X-ray irradiation often induces absorption bands in many materials, and we expect it is easier to measure such effects in this host glass, for, due to the wide band gap energy, zinc-phosphate glass is highly transparent over the spectral range from near infrared to ultraviolet. In addition, Sn-doped zinc-phosphate glass shows over 80% of photoluminescence (PL) quantum yield [4]. Optical materials doped with Sm have been of distinct interest for practical applications including high-resolution and large-dynamic-range radio-hotoluminescence dosimeter for radiation therapy [5, 6], high-resolution X-ray scintillator plate for medical imaging [7], high density optical memory [8], and light harvesting in solar cells [9].

Experimental

Sm^{3+}-doped $ZnO-P_2O_5$ ($70ZnO-30P_2O_5-0.5Sm_2O_3$ in mol%) glass was prepared by a conventional melt-quenching technique. Batches consisting of ZnO and $(NH_4)_2HPO_4$ were first calcinated at 800 °C for 3 hrs. The heat-treated glass precursor was mixed with Sm_2O_3 at room temperature, and then melted in a platinum crucible in an ambient atmosphere using an

*Corresponding author:
Tel : +81-743-72-6144
Fax: +82-743-72-6147
E-mail: go-okada@ms.naist.jp

electric furnace (Super-Burn SH, Motoyama). After melting at 1100 °C for 30 min, the melt was quenched on a stainless steel plate preheated at 200 °C. The synthesized glass sample was then annealed at 415 °C for 1 hr to relieve the internal stress, cut into the size of approximately 10 × 10 × 0.5 mm^3, and polished for the following measurements.

Optical transmission spectrum was measured using a spectrophotometer (V-670, JASCO) to derive the absorbance via -ln(T_t/T_i), where T_t and T_i are the transmitted and incident light intensities of normal incidence, respectively. X-ray induced luminescence (XL), or scintillation, spectra were measured using a lab-constructed set-up as described below. X-rays from the X-ray generator was delivered directly to the sample. The consequent emission as XL was guided, through an optical fiber (727-733-2447, Ocean Optics), to the CCD-based spectrometer (QE Pro, Ocean Optics) to measure the spectrum. Here, the applied voltage and current to the X-ray tube was fixed to 40 kV and 5.2 mA, respectively. TSL was measured using a reader (TL-2000, NanoGray) [10]. The measurements were carried out over the temperature range of 50-400 °C with the heating rate of 1 °C/s. Dose values quoted refer to entrance dose in air at the surface of the sample.

Results and Discussion

Fig. 1. illustrates the optical absorbance spectra of a 0.5% Sm-doped ZnO-P_2O_5 glass. It is highly transparent from the near-infrared range through the ultraviolet. The fundamental absorption edge is located approximately at 218 nm. There are absorption lines around 400 nm as well as in the 1000-1600 nm range. These are attributed to the intra 4f shell transitions by Sm^{3+} as typically observed in various Sm-doped materials. Once the samples were irradiated by X-rays, the sample becomes brownish due to the appearance of additional absorption in the UV-Vis. range. Such X-ray induced absorption, for example, is commonly observed in phosphate glasses. The P-O bonding network capture electrons and holes generated by X-ray irradiation to become F centers [11]. Fig. 2 shows the induced absorption versus the delivered X-ray dose. The X-ray induced absorption increases with the X-ray dose delivered. The linear relationship was confirmed over the dose range from 0.5 Gy to over 20 Gy.

Fig. 3 shows the XL spectrum of 0.5% Sm-doped ZnO-P_2O_5 glass sample in contrast with that of non-doped sample. It is clearly seen that, with the Sm activator, strong light emission is present in the orange to near infrared range. The origins of these emissions are the 4f-4f transitions of Sm^{3+}. The spectral emission range is where the quantum efficiency of Si-based photodetector is high. Fig. 4 shows the response of XL signal integrated against the incident X-ray dose. It was confirmed that the XL response is linear over the exposure dose range of 1-300 R.

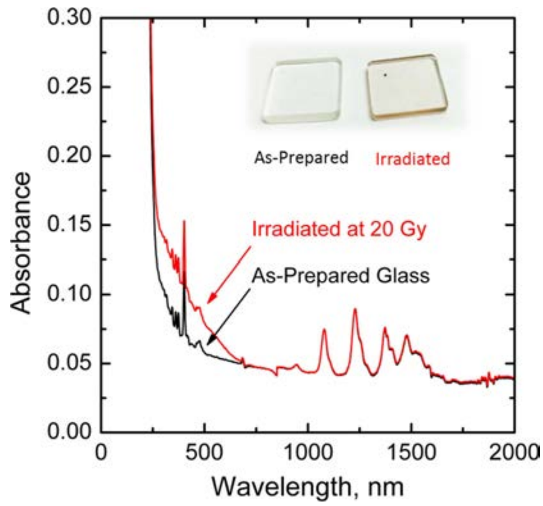

Fig. 1. Optical absorbance of as-prepared and irradiated ZnO-P_2O_5 glasses doped with 0.5% Sm. The inset is the sample photograph.

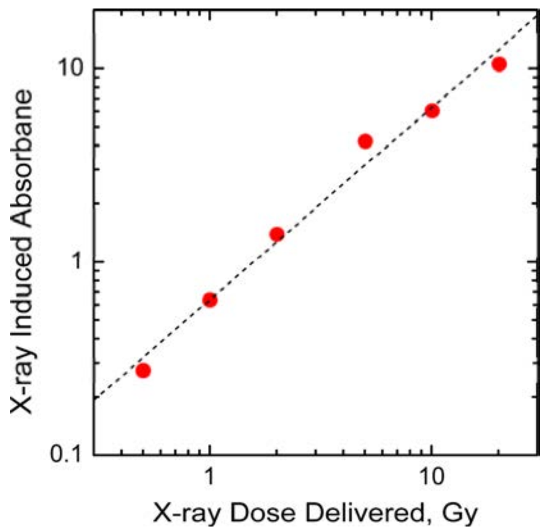

Fig. 2. X-ray induced absorbance of non-doped and 0.5% Sm-doped ZnO-P_2O_5 glasses.

Fig. 5 shows the TSL glow curve of 0.5% Sm-doped ZnO-P_2O_5 glass. Prior to the measurement, the sample was irradiated by X-rays with a dose of 10 Gy. As illustrated in the graph, the glow curve can be fitted with three Gaussians with the peak temperatures of 110, 160 and 256 °C. This fact implies that the charges generated by the X-ray irradiation were trapped by three different groups of trapping centers which have different trapping depths. Apparently, the trapped charges at the shallower centers are supposed to degrade over time due to thermal stimulation even at room temperature, the charges captured at deeper trapping centers should store the dose information for a sufficiently long time to be useful as a storage phosphor. Fig 6 plots the integrated TSL signal as a function of X-ray dose delivered. A linear response was confirmed over a wide range of irradiation dose from 1 mGy to 10 Gy.

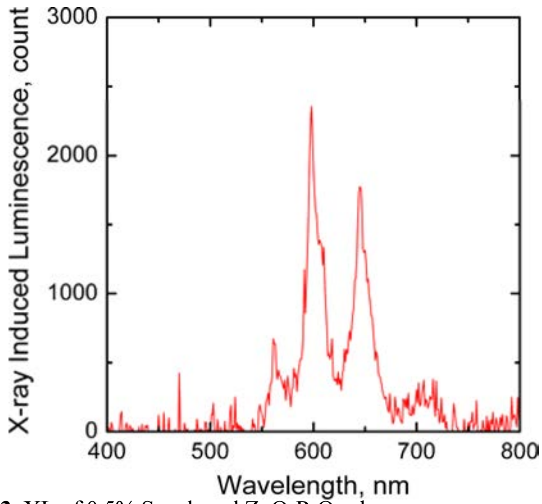

Fig. 3. XL of 0.5% Sm-doped ZnO-P_2O_5 glass.

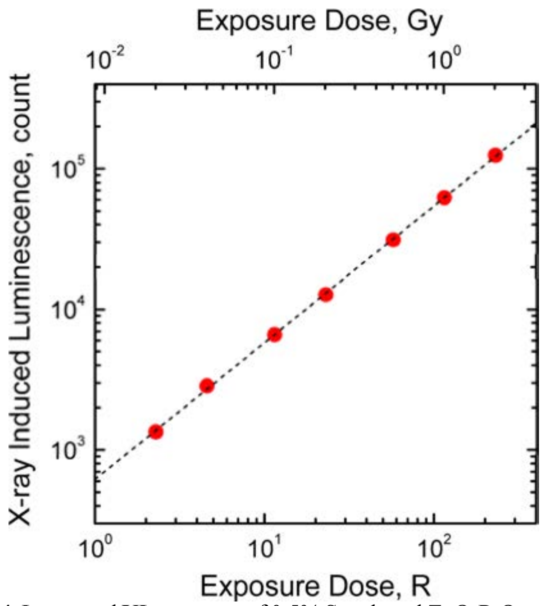

Fig. 4. Integrated XL response of 0.5% Sm-doped ZnO-P_2O_5 glass against the incident X-ray dose.

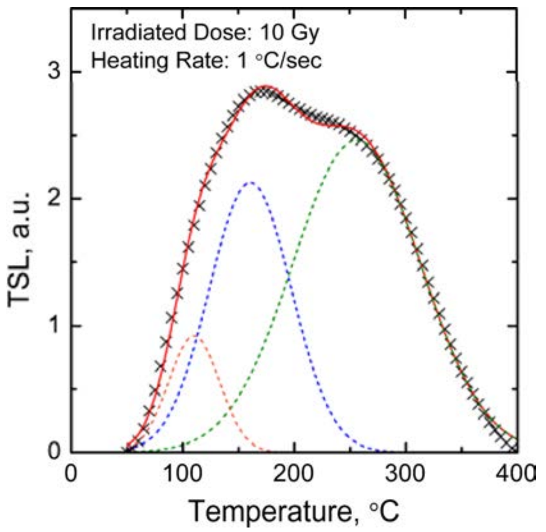

Fig. 5. TSL glow curve of 0.5% Sm-doped ZnO-P_2O_5 glasses. The X-ray irradiation dose prior to the measurement was 10 Gy.

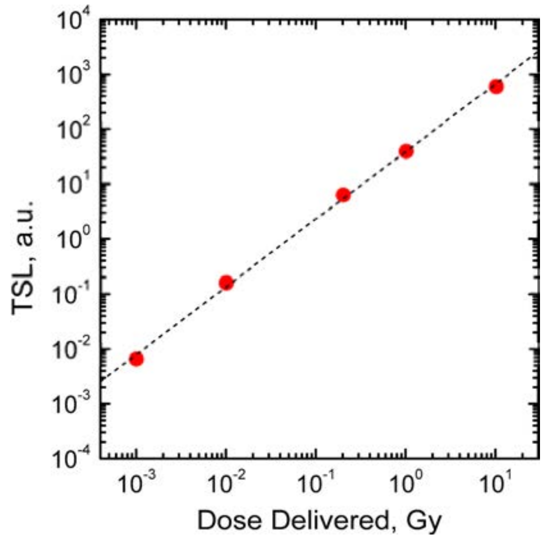

Fig. 6. Integrated TSL response of 0.5% Sm-doped ZnO-P_2O_5 glass against the incident X-ray dose.

Conclusions

We have investigated the X-ray induced effects in Sm^{3+}-doped ZnO-P_2O_5 glass in terms of the optical absorbance, XL, and TSL. With the use of these effects in our glass sample, we have confirmed that X-rays of a wide degree of doses are measurable. Owing to the red emission by Sm^{3+} ion as an activator, Si-based photodetectors can be used for higher quantum efficiency and compact integrated system design.

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