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Radio-Photoluminescence of Silver-Doped Phosphate Glass

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Commercial silver-doped phosphate glass is a promising material for a solid-state radio-photoluminescent (RPL) dosimeter. It has several promising characteristics, such as RPL intensity linearity with the ionizing irradiation dose, data accumulation, no fading, and measurement repeatability. It can be used as personal, environmental, and clinical dosimeter as well. The objective of this study was to synthesize silver-doped phosphate glass in the laboratory and compare its RPL properties with those of a commercial GD-352M glass. The emission and the excitation properties of the synthesized and commercial glasses were measured before and after X-ray irradiation. An intense orange emission peak at 600 nm and an excitation peak at 320 nm were measured for the synthesized glass after X-ray irradiation. The radiative lifetime of the synthesized glass had two decay components for the orange Radio-Photoluminescence.

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I. INTRODUCTIONS

A silver (Ag^+)-doped phosphate glass shows Radio-photoluminescence (RPL) and it is being used for RPL dosimeters [1]. It exhibits an intense photoluminescence under ultraviolet (UV) excitation when the glass is exposed to ionizing radiation. Electron-hole pairs will be created inside the glass by the exposure to ionizing radiation. The created electrons are captured into Ag^+ and converted to Ag^0. Whereas, at the beginning the holes are captured to the PO_4 tetrahedron and later make interaction with Ag^+ to create Ag^{++} . The Ag^0 emits blue light at 450 nm region by 345 nm excitation and the Ag^{++} emits yellow light at 560 nm region by 308 nm excitation. However, the emission and excitation wavelengths may slightly vary with glass matrix composition The RPL intensity linearity with exposed dose, luminescence centers stability with time, RPL readout by heat treatment and reuse are the main advantageous probes to use the glass as individual and environmental radiation dosimeter. Therefore, we need to study the silver (Ag^+)-doped phosphate glass. So, we synthesized a silver-doped phosphate glass and compared it with a commercial GD-352M in order to understand the glass properties and RPL mechanism.

II. EXPERIMENT

A glass with the composition of $74P_2O_5$:$18.2Li_2CO_3$:$2.6Na_2CO_3$:$2.5Al_2O_3$:$2.7AgF$ (mol%) was prepared using P_2O_5, Li_2CO_3, Na_2CO_3, Al_2O_3 and AgF powders of commercial reagent grade. All the chemicals except Al_2O_3 were first dissolved in deionized water and mixed using a magnetic stirring in their respective compositions. However, Al_2O_3 was mixed to the solution without dissolving in deionized water. The mixture was evaporated until viscous fluid was obtained. Then the fluid was poured into an alumina crucible and heated at 1100°C

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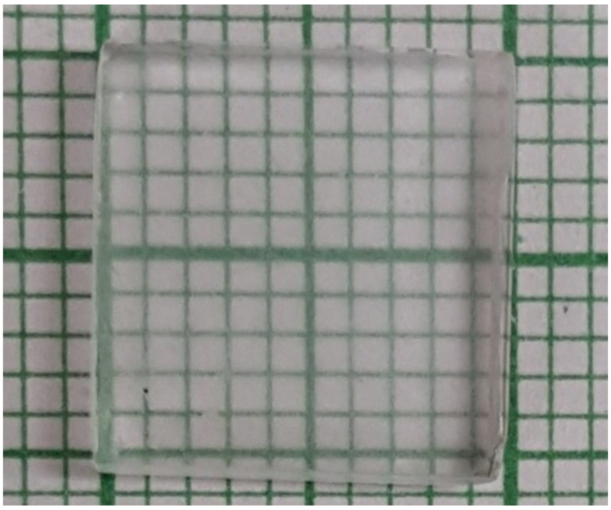

Fig. 1. (Color online) Synthesized glass after cutting and polishing.

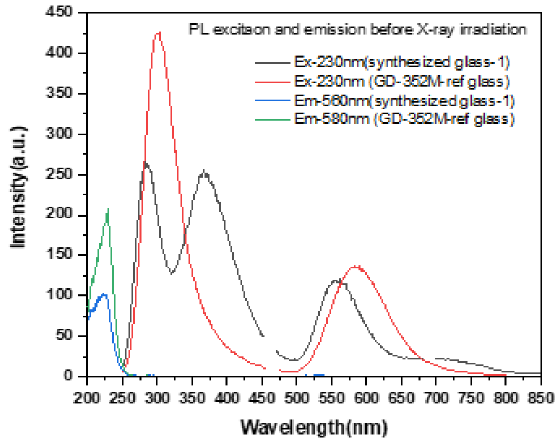

Fig. 2. (Color online) PL excitation and emission spectra of the synthesized and reference GD-352M glasses before X-ray irradiation.

for several hours. The aerogel obtained from the heat treatment was melt at 1200°C for 2 hours in the same alumina crucible and quenched in 300°C preheated brass block. The prepared glass was annealed for 10 hours at 300°C in order to release thermal stress.

The glass was cut and polished well (Fig. 1). However, a small piece of sample was used to check the optical properties before and after X-ray irradiation. A reference glass GD-352M (AGC TECHNO GLASS CO., LTD, LotNo. FD7131213-2) was purchased and used to compare the RPL property with that of the synthesized glass. The weight composition of the reference glass was 31.55% P, 51.16% O, 6.12% Al, 11.00% Na and 0.17% Ag [1]. The chemical content in synthesized glass had additional Li_2CO_3 than in reference glass which helps to sustain the RPL centers in higher temperature and will not affect the RPL property [2].

Measurement of photoluminescence (PL) emission spectra excitation by 230 nm and 247 nm, and radioluminesence emission spectra excitation by 320 nm were carried out at room temperature by using a Cary Eclipse Fluorescence Spectrophotometer (Agilent Technologies) as well as with 340 nm light emitting diode (LED).

Radiative life time was measured by adjusting 2.5×10^{-3} ms gate time and 1.0×10^{-3} ms delay time using the same spectrometer. X-ray irradiation was conducted using X-ray copper target operated at 80 kV and 2 mA.

III. RESULTS AND DISCUSSION

Emission spectra of the synthesized and reference glasses were measured before and after X-ray irradiation. The intense emission peaking at 287 nm, 367 nm and 560 nm was observed for the synthesized glass, whereas 300 nm and 580 nm was observed for the reference glass with 230 nm excitation before X-ray irradiation (Fig. 2). One additional emission band at 367 nm was observed in synthesized glass which was not found in the reference glass which might be related to the compositional difference between two glasses.

After PL measurement, the synthesized glass was irradiated with X-ray for 25 minutes and stored in dark for overnight to provide time for build-up process. The PL and RPL emission and excitation were measured and presented in Fig. 3.

The RPL excitation and emission peaks were recorded at 320 nm and 600 nm, respectively, from their respective excitation and emission wavelengths. It can be pointed out that the RPL emission at 600 nm is related to the Ag^{++} which were created by x-ray irradiation to the Ag^{+} in phosphate matrix [3].

The location of Ag^{+} represent defects in the lattice of the glass matrix. Irradiation with ionizing radiation releases electrons in the dosimeter material from phosphate. As a result, electron and hole are created which are captured by Ag^{+} (Fig. 4), creating stable luminescent centers (Ag^{0}, Ag^{++}) as follows [4,5]

$$Ag^{+} + hPO_4 \rightarrow Ag^{++} \tag{1}$$

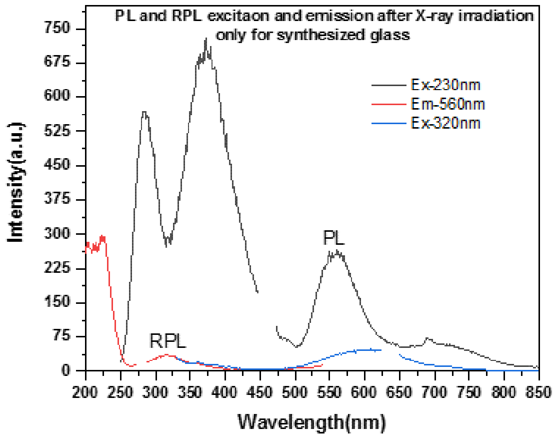

Fig. 3. (Color online) PL and RPL emission excitation spectra of the synthesized glass.

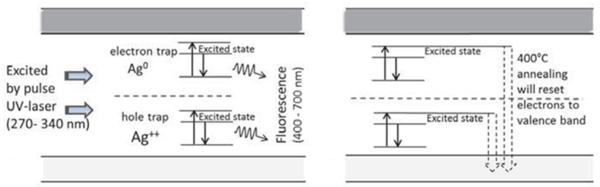

Fig. 4. Schematic representation of fluorescence emission after UV excitation for stable luminescent centers (Ag^0, Ag^{++}) created after X-ray irradiation and read out of RPL centers by annealing in 400 °C (referenced from [6,7][6, 7]).

$$Ag^+ + e^- \rightarrow Ag^0 \tag{2}$$

The created lumenesencent centers does not disappear until sufficient heat energy provided to the glass. Specifically, for the most of the dosimeter glasses are reported to be required 400 °C heat treatment for several minutes. However, it can be different with glass compositional constituents and level of dose in the glasses [6]. The 450 nm emission from the Ag^0 could not be observed for the synthesized as well as for commercial reference glasses for the present study.

The glass under UV light before and after X-ray irradiation is presented in Fig. 5. It was clearly observed that the intense orange emission was generated by 365 nm UV light and fade off original PL emission after X-ray irradiation.

A contour image was prepared by using scan data obtained in each 5 nm excitation wavelengths from 230 nm to 445 nm for X-ray irradiated synthesized glass in order to understand complete PL and RPL excitation and emission region. PL emissions ranged from 250-630 nm, whereas RPL emissions ranged from 500-700 nm were measured. Linear lines were background obtained from the second and third harmonics of the excitation wavelengths in contour plot (Fig. 6).

Fig. 5. (Color online) Synthesized glass under 255 nm and 365 nm UV light (a) before and (b) after X-ray irradiation.

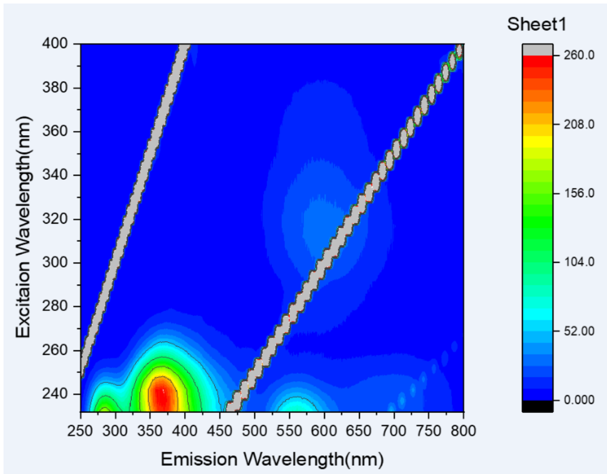

Fig. 6. (Color online) Contour plot of excitation-emission for PL and RPL properties of the synthesized glass after X-ray irradiation.

The RPL emission of the glasses obtained by exciting with 340 nm LED is presented in Fig. 7. The range (500-850 nm) of emission was similar for both glasses. However, the emission peak position shifted to the lower wavelength (600 nm) in the synthesized as compared to reference (632 nm) glass. Inconsistent peak positions were found for other published reports [5,8].

Radiative lifetime of the synthesized glass was recorded by using RPL excitation 320 nm and emission 600 nm. The lifetime was fitted with two exponential decay functions and presented in Fig. 8. Fast decay 2.29×10^{-3} ms component with 89.4% contribution and slower decay 9.96×10^{-3} ms component with 10.6% contribution were recorded and confirm with other report [3].

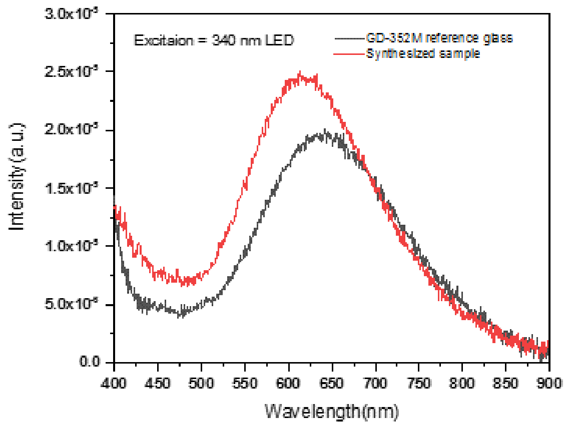

Fig. 7. (Color online) The RPL emission obtained by exciting the synthesized and the reference glasses after X-ray irradiation with 340 nm LED.

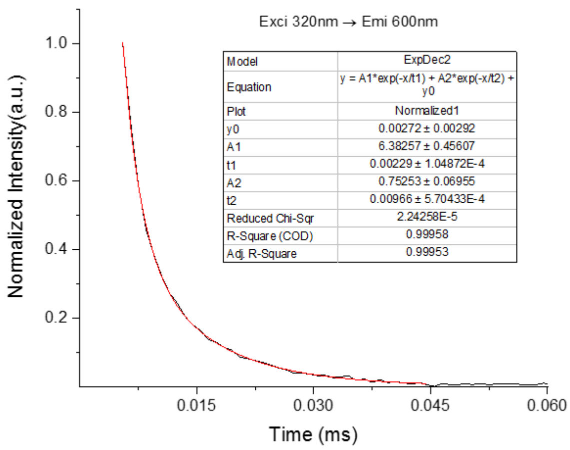

Fig. 8. (Color online) RPL radiative decay time fitting for synthesized glass.

IV. CONCLUSION

Ag^+ doped glass was synthesized in laboratory using commercial reagent grade powders. PL and RPL properties of the glass were measured and compared with reference GD-352M glass. Three PL emissions (287 nm, 367 nm and 560 nm) for synthesized glass and two emissions (300 nm and 580 nm) for reference glass were recorded by exciting 230 nm wavelength. Orange emission peak at 600 nm and excitation peak at 320 nm were measured for synthesized glass after X-ray irradiation. Similar emission range from 500-850 nm were measured for both glasses under 240 nm LED excitation. Radiative lifetime of synthesized glass had two decay components with 2.29 μs and 9.96 μs.

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