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Fluorescent nuclear track images of Ag-activated phosphate glass irradiated with photons and heavy charged particles

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ABSTRACT

In this paper we report about the demonstration of the nuclear track imaging capabilities of Ag-activated phosphate glass. A 375 nm laser and confocal laser scanning microscopy (CLSM) were respectively used for track excitation and detection. Specifically, the blue and orange radiophotoluminescent (RPL) tracks and dose distributions observed after irradiation with soft X-rays, gamma rays and heavy charged particles (HCPs) are examined. In addition, the origins of the reductions in RPL efficiency for high-dose X-ray irradiation and for irradiation with HCPs with high linear energy transfer (LET) values are investigated via a CLSM and a conventional fluorescent reader and discussed.

1. Introduction

Silver-activated phosphate glass, the most widely known radiophotoluminescent (RPL) material [1–4], can be used not only in personal, environmental and clinical dosimeters but also in two-dimensional (2D) and three-dimensional (3D) dose imaging detectors [5–7]. These accumulating passive detectors are based on radiation-induced, optically active Ag-related atomic-scale defects; therefore, the ultimate intrinsic spatial resolution of these detectors is several nanometres [7]. All materials that exhibit RPL phenomena, such as the F_2^+(2Mg) centres in Al_2O_3:C,Mg [8] and the Ag^0 and Ag^{2+} centres in Ag-activated phosphate glass (hereafter, Ag-glass) [3,9], have certain prominent features: wide dynamic ranges, high sensitivity, low energy responses and the capability of multiple non-destructive readouts. Other materials that are known to exhibit RPL phenomena include lithium fluoride (LiF) [10] and CsBr:Sm crystals [11].

Fluorescent nuclear track detectors (FNTDs) based on Al_2O_3:C,Mg crystals, developed by Landauer, Inc [12], represent one of the most attractive technologies for passive solid-state dosimetry as a novel tool of replacing CR-39 plastic nuclear track detectors (PNTDs) [13]. New technology combining confocal laser scanning microscopy (CLSM) with this novel type of FNTD has been developed for use in neutron detection and dosimetry, proton and heavy ion radiobiology, ion beam cancer therapy and space radiation dosimetry as well as nuclear and particle physics research [14–18]. Recently Kodaira et al. [19] demonstrated the utilisation of Ag-glass as a PNTD. The study opened a new stage to the material for RPL dosimeter as for a nuclear track etch detector. Here we have challenged the utilisation of RPL Ag-glass for a FNTD for the first time to reconstruct 2D and 3D track images of the material irradiated with X-rays, gamma-rays and HCPs.

In this paper, the 2D and 3D images determined based on the distributions of radiation-induced Ag^0 and Ag^{2+} defects throughout the entire sample volume in Ag-glass exposed to different X-ray irradiation doses (1, 10 and 100 Gy) are presented. 3D track imaging results for Ag-glass after irradiation with gamma ray (^{137}Cs) and heavy charged particles (HCPs) (protons and Fe ions) are examined via CLSM. In addition, the origins of the reductions in RPL efficiency for high-dose X-ray irradiation and for high-LET HCP irradiation are investigated via CLSM and a conventional fluorescent reader and discussed.

2. Experimental procedures

2.1. Samples

Commercially available Ag-glass plates with dimensions of approximately 8.5×8.5×1.5 mm^3 were used for all the measurements, including absorption, excitation, emission and fluorescent nuclear track imaging. All facets of the samples were optically polished. The composition of the material by weight was the same as that of FD-7 (AGC Techno Glass Co., Ltd., Japan), i.e., 31.55% P, 51.16% O, 6.12%

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Al, 11.00% Na and 0.17% Ag. The mass density, photon effective atomic number and refractive index of this material are 2.61 g/cm^3, 12.57 and 1.52 [6], respectively. After irradiation, all of the samples were preheated to 100 °C for 10 min to promote and complete the 'build-up' kinetics [9,20].

2.2. X-rays, gamma rays and heavy charged particles

Soft X-ray irradiations were performed using an X-ray unit with a copper target operating at a voltage of 30 kV and a current of 20 mA. The X-ray radiation covered the narrow spectral interval of the part of the soft X-ray region by adding filtration, i.e., quasi-monochromatic radiation, strongly peaked at approximately 8.04 keV ($K\alpha_1$ and $K\alpha_2$ lines) with a minor contribution at 8.9 keV ($K\beta_1$ line). The absorbed doses delivered to the samples were 1, 10 and 100 Gy. The samples were placed at a distance from the tube of approximately 10 cm.

Gamma irradiations were performed at a distance of 50 cm from a ^{137}Cs (662 keV) source that delivered a dose of 1 Gy at the Oarai Research Center, Chiyoda Technol Corporation, Japan.

5 Gy irradiations with a variety of high-energy heavy ions (detailed in Section 3.) were performed at the Heavy Ion Medical Accelerator (HIMAC) of the National Institute of Radiological Sciences (NIRS) in Chiba, Japan. X-ray beams were incident perpendicular to the sample surface (8.5×8.5 mm^2 plane).

2.3. Steady-state optical properties of the samples

The steady-state optical absorption spectra were determined at room temperature (RT) using a Hitachi U-3900H spectrophotometer with a 1 nm step. All excitation (EXC) and emission (RPL) spectra were recorded at RT using a Hitachi F-2500 fluorescence spectrophotometer. The ionoluminescent spectra were collected with a resolution of 2.5 nm and detected by a R928 photomultiplier (PMT) (Hamamatsu Photonics, Japan). All the spectra were corrected for the instrumental calibration. In this work, the blue and orange RPL signals were acquired through a long-pass filter passing all wavelengths longer than 400 nm (#84-754, Edmund Optics, USA).

2.4. Fluorescent nuclear track image acquisition

Preliminary investigations of fluorescent nuclear track imaging were performed after the exposure of the samples to various types of radiation, including X-rays, gamma-rays and HCPs, at RT. A Nikon C2+ CLSM instrument (along with its software, NIS Elements version 4.40) was used to acquire and plot fluorescent 2D and 3D images of the Ag-glass samples after irradiation with photons and HCPs. In this work, instead of the 405 nm line which is the shortest available excitation wavelength for a normal C2+ CLSM instrument [7], 375 nm light emitted from a continuous wave (CW) laser diode with a nominal output power of 16 mW (less than 1 mW at the sample) (CUBE 375–16C, Coherent, Inc., USA) was used to excite the Ag^{2+} and Ag^0 absorption bands in the Ag-glass.

For the C2+ CLSM instrument, three objective lenses (Nikon Plan Apo) with different magnifications, numerical apertures (NA) and working distances (WD in mm) as well as different immersion liquids were used: the applied configurations were 10×/0.45NA/4.0WD, 40×/1.25NA/0.16WD/water immersion (refractive index n=1.33), and 60×/1.40NA/0.13WD/oil immersion (n=1.52). The microscope resolution used in this work are defined by the system's point spread function (PSF) and the lateral size of the PSF (FWHM) d_{xy} and the axial size d_z are estimated for an excitation wavelength given 375 nm: d_{xy}=508 nm, d_z=1850 nm for the 10×/0.45NA objective lens, d_{xy}=183 nm, d_z=319 nm for the 40×/1.25NA lens and d_{xy}=163 nm, d_z=126 nm for the 60×/1.40NA lens, respectively. The blue RPL signals attributed to Ag^0 centres and the orange RPL signals attributed to Ag^{2+} centres were acquired using a band-pass filter (450 ± 25 nm) and a long-pass filter passing all wavelengths longer than 561 nm, respectively. Moreover, a pinhole of 40 μm in diameter was installed in front of the detector. Such a confocal detection scheme allowed for high spatial and depth resolution under the condition of one-photon absorption at 375 nm, even when areas of the sample adjacent to the focal spot of the excitation light produce fluorescence.

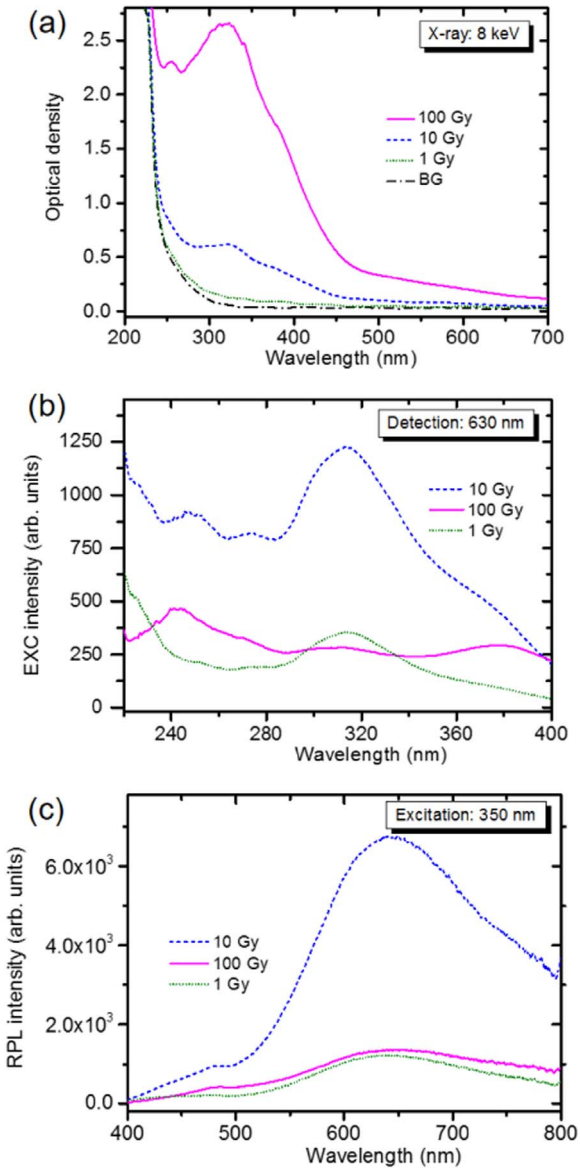

Fig. 1. Steady-state absorption spectra including BG spectra (a) as well as excitation spectra (b) and emission spectra (c) of Ag-glass after X-ray irradiation at various doses.

3. Results and discussion

Fig. 1 presents the steady-state optical absorption spectra, including background (BG, non-irradiated) spectra (a) as well as excitation (EXC) (b) and emission (RPL) (c) spectra of Ag-glass samples after X-ray irradiation at an energy of 8 keV. The samples were exposed to doses of 1, 10 and 100 Gy to examine and compare the effects of the dose on the optical curves. The observed absorption features are attributed to the superposition of a number of individual absorption bands in the range from 200 to 700 nm, corresponding to single Ag^+ ions, neutral Ag atoms, charged Ag_n^{m+} clusters, neutral Ag_n clusters, metal Ag nanoparticles and phosphorus-oxygen-hole centres, as previously reported [9,20,21].