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A comparative study of optical and radiative characteristics of X-ray-induced luminescent defects in Ag-doped glass and LiF thin films and their applications in 2-D imaging



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ABSTRACT

We report novel disk-type X-ray two-dimensional (2-D) imaging detectors utilising Ag-doped phosphate glass and lithium fluoride (LiF) thin films based on the radiophotoluminescence (RPL) and photoluminescence (PL) phenomena, respectively. The accumulated X-ray doses written in the form of atomic-scale Ag-related luminescent centres in Ag-doped glass and F-aggregated centres in LiF thin films were rapidly reconstructed as a dose distribution using a homemade readout system. The 2-D images reconstructed from the RPL and PL detectors are compared with that from the optically stimulated luminescence (OSL) detector. In addition, the optical and dosimetric characteristics of LiF thin films are investigated and evaluated. The possibilities of dose distributions with a high spatial resolution on the order of microns over large areas, a wide dynamic range covering 11 orders of magnitude and a non-destructive readout are successfully demonstrated by combining the Ag-doped glass with LiF thin films.

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1. Introduction

Radiophotoluminescent (RPL) glass dosimeters [1] using silveractivated phosphate glass have been widely used for personal, environmental and clinical dosimetry, along with optically stimulated luminescence (OSL) dosimeters [2] and thermoluminescent (TL) dosimeters [3,4]. In particular, the RPL dosimeter has been recognised as possessing desirable characteristics such as high spatial resolution, non-destructive readout capabilities, a long-term stability against fading, a wide dynamic range and uniformity/ batch homogeneity [5]. Although these three passive types of luminescent dosimeters, based on the RPL, OSL and TL (referred to by their well-known abbreviations for convenience) phenomena, have advantages and disadvantages [3,6], there have been few reports of two-dimensional (2-D) dose distributions achieved over large areas with the aforementioned features.

Several reports have demonstrated stored micro-images written by extreme ultraviolet (EUV) rays, soft and hard X-rays, neutrons, high-energy heavy charged particles and alpha-particles using radiation-induced centres or stable aggregate colour centres (CCs) in transparent materials.

Firstly, although lithium fluoride (LiF) crystals doped with magnesium and titanium (LiF:Mg, Ti) are well known to function as TL detectors, it is difficult to adopt such TL phenomena to 2-D imaging due to a lack of repeatable readouts. However, stable F_2 and F_3^+ CCs, whose centres consist of two electrons bound to three or two adjacent anion vacancies, respectively, embedded in LiF crystals and films based on the photoluminescence (PL) phenomenon can be used for high-performance detectors in micro-radiography, X-ray microscopy and phase contrast imaging [7-9]. Secondly, stable F_2^{2+} (2 Mg) CCs in aluminium oxide doped with carbon and magnesium (Al₂O₃:C, Mg) crystals based on the OSL phenomenon have been used as a fluorescent nuclear track detector [10]. Thirdly, radiation-induced silver species such as stable Ag²⁺ or Ag⁰ CCs in Ag-doped glass based on the RPL phenomenon can be used in microscopic dose measurements [11]. However, all of the aforementioned image types are optically read by a confocal laser scanning microscope (CLSM). Consequently, these methods are less suitable for measuring large-area images, particularly, in medical dosimetry applications or in outdoor environments, e.g., in structural health monitoring for buildings, tunnels and bridges.

In this paper, novel disk-type X-ray 2-D imaging detectors utilising Ag-doped phosphate glass and LiF thin films deposited on glass based on the RPL and PL phenomena, respectively, are proposed and demonstrated for applications in diagnostic dosimetry and radiation therapy. The capabilities of the reconstructed dose distributions with a wide dynamic range covering 11 orders of

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magnitude, a high spatial resolution on the order of microns over a large area and a non-destructive readout are successfully demonstrated for the first time by combining the Ag-doped glass with LiF thin films.

2. Experimental details

2.1. Sample preparation and characterisation

A commercially available silver-doped phosphate glass dosimeter, GD-450 (Asahi Techno Glass Corporation), was used as a 2-D RPL detector. Although the weight composition of the 2-D detector was the same as that of the GD-450, i.e., 31.55% P, 51.16% O, 6.12% Al, 11.00% Na and 0.17% Ag, the size and shape were different; the 2-D detector was a disk-type plate with a diameter of 80 mm and a thickness of 1 mm [12].

As a PL detector, LiF thin films deposited by a thermal evaporation technique on a borosilicate glass (BK-7) substrate were used. In addition, a commercially available X-ray imaging plate, BaFBr:Eu²⁺ (Fuji Photo Film), referred to as BAS-SR, was used for comparison as an OSL (or PSL) detector, which was attached to a disk-type BK-7 glass plate. The three kinds of disk-type detectors had the same diameter and thickness, with a 15-mm-diameter hole at the centre for rotation.

LiF thin films with a thickness of 1 μ m were prepared as follows: LiF sintering pellets were used as the starting material. The substrate temperature was held constant during deposition at 200 and 300 °C. The present work did not aim to determine optimal conditions for the deposition rate, thickness or substrate temperature with respect to the structure and morphology; therefore, the evaporation parameters were selected based on a previous investigation [13] of LiF thin films deposited on a glass substrate. The evaporation rate and vapour pressure in the chamber were 0.5 nm/s and 4.0 $\times 10^{-4}$ Pa, respectively.

To analyse the polycrystalline structure of the deposited LiF thin films, X-ray diffraction (XRD) patterns were recorded. In addition, the properties of the top surface (\sim 10 nm thick), such as the chemical composition and the element bonding of the LiF films, were investigated by means of X-ray photoelectron spectroscopy (XPS).



Fig. 2. EXC and RPL spectra of Ag-doped glass (a) after X-ray irradiation with an absorbed dose of 1 Gy. EXC and PL spectra of X-ray-irradiated LiF thin films (b) after X-ray irradiation with an absorbed dose of 126 Gy.

In this work, the samples were coloured by irradiation from an Xray unit (8.05 keV) with a copper target operating at 30 kV and 20 mA. The absorbed doses on the samples ranged from 1 to 126 Gy. Excitation (EXC), PL and RPL spectra were obtained at room temperature using a Hitachi F-4500 fluorescence spectrophotometer.

2.2. Imaging readout system

A schematic view of the experimental setup employed to measure the 2-D dose distributions is shown in Fig. 1. To read out the



Fig. 1. Schematic diagram of a disk-type RPL, PL and PSL readout system (not to scale).

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