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X-ray storage performance of KCl:Eu^{2+} with high cumulated dose

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ABSTRACT

The effects of high cumulative radiation dose on the luminescence properties of KCl:Eu^{2+} are investigated. Pellet samples of KCl:Eu^{2+} were given doses of up to 200 kGy at the Louisiana State University Synchrotron facility. After synchrotron irradiation, samples were optically bleached and given a clinical dose of 2 Gy from a 6 MV medical linear accelerator. Optical properties were evaluated using photostimulated luminescence (PSL), photoluminescence (PL), and temperature-dependent PSL measurements. For a cumulated dose of up to 5–10 kGy, the PSL emission intensity increased by 15% compared to the PSL signal with no radiation history. For doses higher than 10 kGy, the PSL emission intensity retained at least 70% of the original intensity. Spatial correlation of the charge storage centers increased for doses up to 5 kGy and then decreased for higher cumulative doses. Emission band at 975 nm was attributed to transitions of Eu^{1+}. PL spectra showed an intense peak centered at 420 nm for all cumulative doses. The results of this work show that KCl:Eu^{2+} storage phosphors are excellent reusable materials for radiation therapy dosimetry.

1. Introduction

Two-dimensional radiation therapy dosimeters require sub-millimeter spatial-resolution due to high dose gradient associated with modern radiation therapy treatment modalities such as IMRT (intensity modulated radiation therapy). In particular, a dosimeter should be reusable so that response variation from pixel to pixel can be quantified and therefore corrected. Radiographic film has been the detector of choice for IMRT dose distribution verification. Films can be inserted in any orientation in a phantom mimicking patient's anatomy, and it has unparalleled high spatial-resolution that is essential for verification of steep dose gradients. However, film is not reusable, and quantitative use requires the acquisition of a sensitometric curve for each measurement with a questionable assumption that individual films from a single batch and individual pixels on the same sheet share a common response. Also, the implementation of digital imaging in diagnostic and radiation oncology departments is causing departments to systematically remove film processors. In 2005, Olch showed that the BaFBrI:Eu^{2+}-based computed radiography panels had the potential to be used for two-dimensional megavoltage radiation therapy dosimetry [1]. However, BaFBrI has a high Z number (Z_{eff} = 49) which leads to a strong photon energy dependence and consequently unacceptable measurement accuracy. Also, BaFBrI or CsBr-based detector panels were designed for diagnostic radiology where radiation doses are on the order of μGy-mGy. For radiation therapy, a typical fractionated dose is 2 Gy. Therefore, a reusable dosimeter with tissue-like response, high spatial-resolution, and excellent radiation hardness properties is desirable for quantitative radiation therapy dosimetry.

Recently, we have shown that KCl:Eu^{2+}-based dosimeters provide similar energy response as radiographic film [2,3]. The Z_{eff} is 18, which is much closer to tissue than BaFBrI or CsBr. Further, we have shown that the luminescence properties of KCl:Eu^{2+} remain optimal for cumulative doses of up to 3000 Gy which means that a KCl:Eu^{2+}-based dosimeter can be used at least 1500 times in the clinic before any signal degradation occurs [4]. A radiation resilient dosimeter allows for less frequent dosimeter calibration and, as a result, reduces measurement uncertainty. For comparison, it has been reported that the PSL signal of CsBr:Eu^{2+} starts to degrade after a few tens of Gy [5,6].

In this work, we present the first attempt to explain why KCl:Eu^{2+} exhibits superior radiation hardness properties, compared to other alkali halide phosphors. We will examine the electronic environment and microstructure of KCl:Eu^{2+} dosimeters with high cumulative dose using low temperature photostimulated luminescence (PSL) spectroscopy, X-ray diffraction, and photoluminescence (PL) emission spectroscopy.

2. Experimental

Pellets of KCl:Eu^{2+} (1 mm thick, 6 mm diameter) with 0.05 mol% Eu, were made using a hydraulic press using a procedure described

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earlier [4]. The XRLM4 beam line at the Center for Advanced Microstructures and Devices (CAMD) synchrotron facility at Louisiana State University was used for high dose irradiation of samples. The energy of the particles in the storage ring was 1.3 GeV with a 100 mA beam current. The energy spectrum emitted from the XRLM4 beamline was a Gaussian distribution with the peak energy of approximately 20 keV. A 175 μm Be window was used to filter out low energy wavelengths. Pellets were placed in an acrylic holder with circular holes with the spacing between each hole at 0.2–0.4 cm. The sample holder was moved in a vertical direction to ensure homogeneous irradiation along the sample surface.

After synchrotron irradiation, samples were shipped back to St. Louis, MO and were bleached of all radiation dose information using an intense halogen lamp. After bleaching, clinical doses of 2 Gy were given using a medical linear accelerator (Trilogy, Varian Medical System, Palo Alto, CA, USA). Solid-water phantoms (Gammex RMI, Middleton, WI) consisted of several 30×30 cm^2 slabs and were arranged to provide 10 cm of backscatter and the desired depth for irradiation. Dosimeter pellets were placed in a 0.5 cm thick slab machined with a linear array of holes across the center. PSL point measurements were obtained using a setup described earlier [4]. Low temperature measurements were obtained by placing dosimeters in a temperature-controlled cryostat (Model CCS-450, JANIS, Wilmington, MA). PL and PSL emission spectra were obtained using a Nanolog Spectrofluorometer (Horiba Jobin Yvon Kyoto, Japan). A 450 W Xe lamp was used for the excitation source and the excitation light passed through a 1200 lines/mm grating to the solid sample. Emission light was detected using a Synapse Open-electrode CCD detector (Horiba, Kyoto, Japan) sensitive between 300 and 1000 nm.

3. Results and discussion

Fig. 1 shows the PSL sensitivity with various cumulated dose histories. Fig. 1 shows that the sensitivity increases by 15% with a cumulative dose of 5 kGy and then decreases with higher cumulated doses. These results correlate well with previous results [4]. Batentschuk et al. [6] also reported a slight increase in PSL intensity for CsBr:Eu^{2+}. Cumulative doses higher than 5 kGy result in a 20–30% loss of signal, relative to no dose history. In order to further investigate the changes in PSL with cumulative dose, we will focus on two questions.

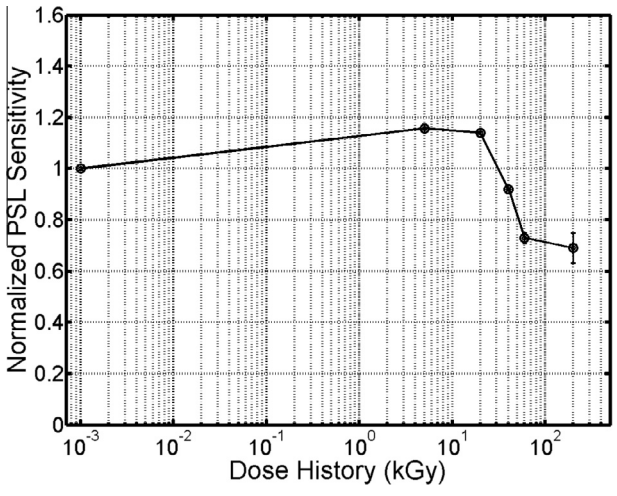

Fig. 1. PSL sensitivity with cumulated dose in KCl:Eu^{2+}. The PSL sensitivity was normalized to the signal from fresh KCl:Eu samples with no cumulative dose. Samples were irradiated using the synchrotron facility at LSU. After optical bleaching, samples were given 2 Gy using a 6 MV medical linear accelerator and the PSL signal was measured. Samples were excited with 560 nm and emission was detected at 420 nm.

3.1. First, why does the PSL increase with cumulative dose?

The Takahashi model assumes the existence of F$^+$-centers in the lattice prior to irradiation [7]. On the other hand, according to Itoh [8], the photostimulable centers in alkali halides can also be created by ionizing radiation by the process in which a self-trapped exciton undergoes an isomeric transformation to an F-H pair. Itoh's model suggests that electron and hole storage centers created by ionizing radiation are spatially correlated, which has been confirmed by von Seggern et al. [9]. After photostimulation, F-center electrons require thermal energy to be released from the relaxed excited state (RES) of the F-center into the conduction band. When the temperature is too low, an electron in the RES will not be able to thermally leave the F-center and, therefore, will either decay back into the F-center ground state or tunnel to a nearby V_k-center to recombine. However, in order for an electron to tunnel to a nearby V_k-center, the F-center must be physically close (spatially correlated) to the V_k-center. Thus, PSL yield at low temperatures corresponds to the concentration of spatially correlated traps [10,11].

Fig. 2 shows the spatial correlation of V_k and F-centers in KCl:Eu^{2+} for various cumulative doses. Samples were irradiated at 50 K in a cryostat chamber. Samples were left overnight in the dark, under vacuum, in order to avoid recuperation effects [12]. The next morning, PSL measurements were taken by stimulating with 560 nm light and collecting emission light at 420 nm. The PSL emission is only from the Eu^{2+} transition to the ground state. About 20% of the centers in the fresh sample were correlated, compared to 40% correlated for the 5 kGy sample, and 15% for the 200 kGy sample. The increase in spatially correlated centers for the 5 kGy sample coincides with the PSL intensity results shown in Fig. 1, suggesting additional photostimulable traps are created as a result of ionizing radiation in addition to preexisting traps.

3.2. Second, what radiation damage effects are created by high accumulated dose in KCl:Eu^{2+}?

Fig. 3 shows the PSL emission spectra as a function of cumulative dose in the IR region. Samples were stimulated with 560 nm or F-center light. All spectra were normalized to the peak intensity of the characteristic 420 nm PSL emission for KCl:Eu^{2+} (shown in

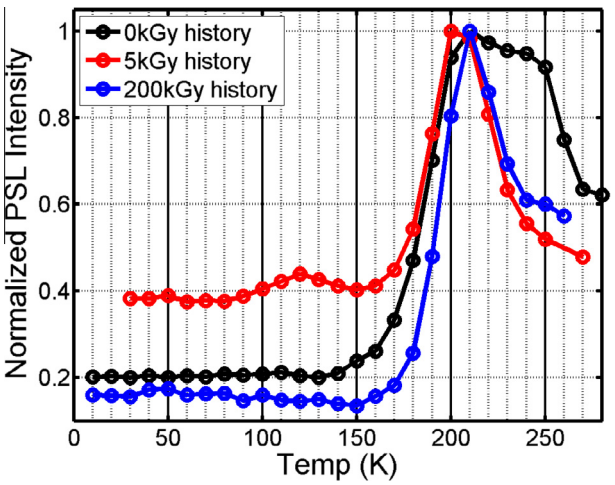

Fig. 2. Normalized PSL intensity at 420 nm as a function of temperature in KCl:Eu^{2+} after X-ray irradiation at 50 K. Samples were stimulated with 560 nm.