Radiation hardness of the storage phosphor europium doped potassium chloride for radiation therapy dosimetry

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(Received 26 January 2011; revised 21 June 2011; accepted for publication 23 June 2011; published 27 July 2011)

Purpose: An important property of a reusable dosimeter is its radiation hardness, that is, its ability to retain its dosimetric merits after irradiation. The radiation hardness of europium doped potassium chloride (KCl:Eu²⁺), a storage phosphor material recently proposed for radiation therapy dosimetry, is examined in this study.

Methods: Pellet-style KCl:Eu²⁺ dosimeters, 6 mm in diameter, and 1 mm thick, were fabricated in-house for this study. The pellets were exposed by a 6 MV photon beam or in a high dose rate ¹³⁷Cs irradiator. Macroscopic properties, such as radiation sensitivity, dose response linearity, and signal stability, were studied with a laboratory photostimulated luminescence (PSL) readout system. Since phosphor performance is related to the state of the storage centers and the activator, Eu²⁺, in the host lattice, spectroscopic and temporal measurements were carried out in order to explore radiation-induced changes at the microscopic level.

Results: KCl:Eu²⁺ dosimeters retained approximately 90% of their initial signal strength after a 5000 Gy dose history. Dose response was initially supralinear over the dose range of 100–700 cGy but became linear after 60 Gy. Linearity did not change significantly in the 0–5000 Gy dose history spanned in this study. Annealing high dose history chips resulted in a return of supralinearity and a recovery of sensitivity. There were no significant changes in the PSL stimulation spectra, PSL emission spectra, photoluminescence spectra, or luminescence lifetime, indicating that the PSL signal process remains intact after irradiation but at a reduced efficiency due to reparable radiation-induced perturbations in the crystal lattice.

Conclusions: Systematic studies of KCl:Eu²⁺ material are important for understanding how the material can be optimized for radiation therapy dosimetry purposes. The data presented here indicate that KCl:Eu²⁺ exhibits strong radiation hardness and lends support for further investigations of this novel material. © 2011 American Association of Physicists in Medicine.

[DOI: 10.1118/1.3611043]

Key words: storage phosphor, dosimeter, IMRT

I. INTRODUCTION

Multidimensional reusable dosimeters are extremely important for characterizing the complex dose distributions associated with modern radiation therapy techniques, such as intensity modulated radiation therapy (IMRT).^{1,2} In particular, reusability provides medical physicists a degree of confidence in dosimetric measurements through the acquisition of benchmarking datasets and through long-term, repeated use and performance monitoring. Yet the unique combination of desirable features of a radiation therapy dosimeter, such as water equivalence, high spatial resolution, capability of

phantom integration, temporal stability, and so forth, makes designing and developing a multidimensional reusable dosimeter challenging.

Recently, europium doped potassium chloride, KCl:Eu²⁺, was introduced as a material with the potential to make a significant advancement in megavoltage dosimetry as the physical foundation of a novel, reusable, high spatial resolution, water-equivalent, phantom insertable planar dosimeter.³ Functionally, KCl:Eu²⁺ dosimeters rely on a mechanism of photostimulated luminescence (PSL),^{4–10} similar to BaFBr_{0.85}I_{0.15}:Eu²⁺, a phosphor material used in diagnostic computed radiography (CR). Irradiation of the

material produces electron-hole pairs that are stored in metastable energy traps. The spatial distribution of these trapped charge carriers forms a “latent image,” whose information can be read out by stimulating the trapped charge carriers to recombine and release PSL photons proportional to the locally deposited dose. Charges remaining trapped after readout can be “erased” with a bright, broadband light and the material can be used again in the same manner.

One way to maximize the waterlike response of the KCl:Eu²⁺ dosimeter is to reduce the effective thickness of the sensitive volume. By reducing the effective thickness the absorbed dose in the sensitive volume may be deposited almost exclusively by electrons generated in the surrounding waterlike media which cross the volume, i.e., “crossers.”¹¹ Thus, Han *et al.* suggested that a two-dimensional (2D) KCl:Eu²⁺ storage phosphor panel on the order of a few microns thick, fabricated by, for example, physical vapor deposition, would minimize the energy dependence and maximize the water-equivalent response of the dosimeter. Zheng *et al.* confirmed this suggestion through Monte Carlo simulations, showing that micron-thick panels respond accurately in both depth dose and cross plane profiles.¹² Reducing the effective thickness of the commercial phosphor, BaFBr_{0.85}I_{0.15}:Eu²⁺, would not provide the same benefits, however, because low energy photon interactions in the sensitive volume remain significant in this material due to its high effective atomic number (49 vs 19 for KCl:Eu²⁺).¹² Zheng *et al.* also performed calculations to show that KCl:Eu²⁺ panels, although very thin, would still provide an excellent signal for delivered doses down to a few cGy for 0.5 × 0.5 mm² pixel size. Taken together, these reports are a strong indication that KCl:Eu²⁺ material-based dosimeters have the potential to advance the state of the art in radiation therapy dosimetry, thereby providing the medical physics community with a unique resource for multidimensional dosimetry.

While KCl:Eu²⁺ has significant potential for radiation therapy dosimetry, its radiation hardness, that is, its ability to retain its dosimetric merits with cumulated dose, is an important property and in need of systematic study. Recent investigations have demonstrated the complex and material-dependent effects of cumulative dose on various solid-state dosimeters that are designed for multiple uses. For example, a decrease in sensitivity for accumulated doses from 20 to 60 Gy has been reported^{13,14} for Al₂O₃:C, which is used in optically stimulated luminescence dosimetry (OSLD) radiation badges. For a 6 MV beam, dose response for this material was reported to be linear at low doses and supralinear at higher absorbed doses.^{13,14} Recently, Jursinic reported further changes in OSLD sensitivity and dose response with dose history, attributing the changes to the balance of electron and hole trap states in the material lattice.¹⁵ At high dose histories, e.g., greater than 1000 Gy, OSLDs exhibited higher sensitivity and a linear response.¹⁵ Jursinic suggested that Al₂O₃:C dosimeters could be used for accurate radiation therapy dosimetry if the sensitivity and extent of supralinearity were established for each dosimeter in use.¹⁵ Recent research^{16–19} into an emerging diagnostic CR material, CsBr:Eu²⁺, however, has found a rapid loss of signal with cumulated dose,^{20–22} prob-

ably resulting from radiation-mediated agglomeration of activator ions (Eu²⁺) in this material, which leads to luminescence quenching.²³ It has been reported that CsBr:Eu²⁺ maintains only 10% of its initial sensitivity after 100 Gy.²⁰ While co-doping this material with lithium (Li) has been explored as a way to slow the agglomeration of activator ions, the sensitivity loss remains significant (50% after 25 Gy) by radiation therapy standards.^{21,22}

The present work offers the first systematic investigation into the radiation hardness of KCl:Eu²⁺. The prototype dosimeters used in this study were manufactured by using the solid state reaction methods. Macroscopic properties, such as sensitivity, dose response linearity, and signal stability, were explored using a laboratory-scale PSL readout system. Since phosphor performance is linked to changes in the charge storage centers and the activator environment within the host lattice,⁹ spectroscopic and temporal measurements were carried out to explore potential changes at the microscopic level.

II. MATERIALS AND METHODS

II. A. Dosimeters and phantom

High purity KCl (99.9975%) particles (45–63 μm) and reagent grade EuCl₃•6H₂O particles (< 25 μm) were added at 0.05% mole and mixed. Powders were pressed at 2200 lbs force in a hydraulic press (Carver Inc., Wabash, IN) using an evacuable pressing die to 6 mm in diameter and 1 mm thick. The pellets were sintered at 710 °C for 3 h in air and allowed to cool naturally to 300 °C followed by a rapid cooling to room temperature. Dosimeters were either coated with a moisture-protective coating²⁴ or wrapped in plastic wrap for most measurements to mitigate the adverse effects of moisture on the material. The plastic was removed for PL measurements in order to eliminate blue luminescence due to stimulation of the plastic material by UV light.

Forty-by-forty cm² water-equivalent sheets (SW-457, Gammex RMI, Middleton, WI) were placed on top of another to provide 11 cm of backscatter and desired depth for irradiation. The dosimeters were held in place in a 0.5 cm thick slab machined with a linear array of holes across the center. Optical annealing with a 500 W tungsten-halogen lamp prior to measurement removed background signal created through environmental irradiation.

II.B. Optical setup

Photostimulated luminescence was read on an experimental optical setup described in Han *et al.*³ In brief, the stimulation source consisted of either a 2 mW, 594 nm He–Ne laser (Melles Griot, Covina, CA) or a 150 W, UV enhanced Xe arc lamp (Newport, Stratford, CT). Wavelength selection for the Xe source was provided by a motorized monochromator (Cornerstone 130, Newport) with a grating of 1200 lines/mm and a combination of optical filters. Stimulation light was directed to an integrating sphere (LabSphere, North Sutton, NH) using either glass or UV silica lenses. A custom mount held dosimeters in place on the integrating sphere

during measurement. PSL and PL were selected from stimulation light by means of optical filters and detected with a photomultiplier tube (Hamamatsu, Bridgewater, NJ) connected to a lock-in amplifier (Model SR830, Stanford Research Systems). Stimulation power was attenuated to allow for repeated readings. The standard error of repeated measurements was taken as the experimental error.²⁵

II.C. Dosimetric properties

II.C.1. Irradiations

A 6 MV (nominal) x-ray beam, generated by a Varian Trilogy (Varian Medical Systems, Palo Alto, CA) calibrated according to the AAPM TG-51 protocol, was used for irradiation to doses up to 300 Gy. The nominal dose rate was 600 MU/min. The dosimeter plane was perpendicular to the beam central axis for all irradiations.

For doses greater than 300 Gy, a high dose rate ¹³⁷Cs irradiator (J. L. Shepherd and Associates, Mark I Model 25, San Fernando, CA) was utilized. Based on factory calibrations and isodose curves, the dose rate to water was 10.7 Gy/min, corrected for decay to the time of irradiation. Dosimeters were mounted on a foam block that was cut to place the dosimeters near the center of the irradiation chamber. During irradiation, the block was rotated to mitigate any inverse-square distance dependence of dose delivery to individual chips. The delivered dose was verified using a commercial radiochromic film (EBT2, International Specialty Products, Wayne, NJ).

With the exception of escalating doses, irradiations were performed during evenings and irradiated dosimeters were kept in a laboratory desiccator in the dark, at the room temperature, and read on the next day. Relative chip sensitivities, or “chip factors,” were determined for each dosimeter prior to irradiation. Fluctuation of laser power was measured and found to be 2% during a period of 2–24 h after switching on the laser.

II.C.2. Dosimetric measurements

The following tests were conducted to examine the radiation hardness of the KCl:Eu²⁺ material:

1. *Sensitivity with cumulated dose.* A KCl:Eu²⁺ dosimeter was irradiated to 200 cGy in a 6 MV beam at 100 cm SSD, 1.5 cm depth, and a 10 × 10 cm² field to establish its zero-dose history response. The dosimeter was then bleached and given doses of 500, 1000, 2000, 3000, 4000, and 5000 Gy. After each escalating dose, the dosimeter was optically annealed to zero, and given another 200 cGy and the sensitivity was compared to the baseline value.
2. *Dose response linearity with cumulated dose up to 200 Gy.* Dosimeters were irradiated individually up to 700 cGy with an SSD of 100 cm, depth of 4 cm, and a field size of 10 × 10 cm² using a 6 MV beam. After reading the dosimetric information, the dosimeters were bleached, given escalating doses, and bleached again. The sensitometric data were then reacquired. This process was repeated for several dose levels up to 200 Gy.
3. *Dose response with cumulated dose up to 5000 Gy.* To check whether there were any changes in linearity at high

dose histories, sensitometric curves with points at 100, 300, and 500 cGy were taken with histories from 2 to 5000 Gy. For doses greater than 300 Gy, the Cs-137 irradiator was used to give the escalating doses.

4. *Response reset with annealing procedure.* Three chips with greater than 5000 Gy dose history were annealed at 710 °C in air for 3 h. A sensitometric curve was obtained and this curve compared to dosimeters with 2 and 5000 Gy dose history without an annealing procedure.
5. *Signal stability with cumulated dose.* The signal stability of dosimeters with various dose histories was monitored during the course of the experiments using the procedure explained in Han *et al.*³ This procedure calls for probing the signal at various times after irradiation, in this case every hour, and correcting the data for readout signal loss. Simulation power was reduced through the use of neutral density filters with a total optical density of 3 and applied for 1.5 s in order to reduce the readout loss to 0.15% per reading. A linear fit was made to data points collected 13 h or more after irradiation. The slope of the fitted curve provided a good estimate of the signal stability. Results are reported along with the uncertainty of the slope estimate, given that there was 1%–2% measurement point uncertainty due to electronic noise and laser power fluctuation. Note that adding a reference detector could partially mitigate this uncertainty, but it was not necessary for the purpose of this study.

II.C.3. Spectroscopic measurements

In order to investigate whether high dose alters the properties of the charge storage center and Eu²⁺ activator in the KCl host lattice, the following measurements were carried out:

1. *PSL stimulation and emission spectra with cumulated dose.* The experimental setup for measuring PSL stimulation and emission spectra has also been described in a previous publication.³ For stimulation spectra measurements, the monochromator was positioned in the light path between the Xe source and the sample and scanned while PSL was collected through a bandpass filter. For emission spectra measurements, the monochromator was positioned between the sample and the PMT and scanned while PSL was collected at different wavelengths. These spectra were obtained for dosimeters with dose histories up to 5000 Gy in order to examine whether any microstructural changes were occurring with dose history.
2. *PL emission spectra with dose.* Broadband UV stimulation light was selected from Xe arc lamp emission by a combination of two 3 mm U340 (Schott) and two 3 mm OG550 (Edmunds Optics) short pass filters. UV optics directed the stimulation light to an input on the integrating sphere directly opposite sample. The monochromator was again placed between the integrating sphere and the PMT, which was fitted with two BG-3 filters and a BG-39 filter (3 mm each) and scanned between 400 and 500 nm in 1 nm increments. Background leakage through optical filters was collected and subtracted from measurements.

3. *Luminescence lifetime with dose.* The luminescence lifetime is defined as the time it takes to reduce PL intensity by a factor of e and is determined by the lifetime of the excited state of the Eu²⁺ activator ion in the surrounding host lattice.⁶ This figure of merit can be an indicator of microstructural changes occurring around the activator site. A 308 nm pulsed (30 ns) excimer laser was used as an excitation source.²⁶ PL intensity was collected at 420 nm following excitation and plotted on a log scale for dosimeters irradiated from 0 (i.e., a fresh dosimeter) up to 5000 Gy history.

III. RESULTS

III.A. Dosimetric measurements

1. *Sensitivity with cumulated dose.* Figure 1 shows that the sensitivity of a KCl:Eu²⁺ dosimeter at various dose levels relative to its baseline (zero dose history) value. An increase in sensitivity up to 3000 Gy was observed, followed by a decline in sensitivity to 5000 Gy. The material retained approximately 90% of its original sensitivity after delivery of 5000 Gy.
2. *Dose response linearity.* Figure 2 shows that the sensitometric curve changes with dose history (additional dose histories similar and not shown). At low dose history, a supralinear response over the range of 100–700 cGy is evident. As dose accrues, the magnitude of the supralinearity decreases and the curve becomes more linear. After 64 Gy history, measured data fit a linear model with $R = 0.999$ and an average deviation of 1.3% with a maximum of 3.5% over the full dose range, shown in the inset to Fig. 2, which is within the experimental uncertainty of the measurements. While there is residual supralinearity present over a dose range as large as 600 cGy, the effect can be minimized if the dosimeter is calibrated over a shorter range of interest. For example, the maximum deviation is less than 2% over a range of 100–400 cGy

FIG. 1. Sensitivity with cumulated dose. A KCl:Eu²⁺ dosimeter was given 200 cGy to establish its zero dose history sensitivity, given escalating doses, bleached, and checked for sensitivity at 200 cGy. An initial increase in sensitivity up to 3000 Gy is followed by a gradual decrease. 90% of initial sensitivity remains after 5000 Gy history. Error bars represent the standard error of repeated measurements.

FIG. 2. Response with cumulated dose up to 200 Gy. Dose response curves at various dose histories were obtained and plotted, normalized to 200 cGy. The dose response initially shows supralinear behavior but becomes more linear with dose history. The inset displays the percent deviation of the measured data from a linear model. Error bars represent the standard error of repeated measurements.

after 64 Gy accumulated dose. As shown in Fig. 3, the shape of the normalized sensitivity curves changes very little with cumulated dose up to 5000 Gy (additional dose histories not shown).

3. *Dose response reset.* Chips irradiated to greater than 5000 Gy were annealed at 710 °C for 3 h. The supralinear response returns, nearly to its original, zero dose history shape, which is shown in Fig. 4. It was found that after annealing the average chip sensitivity returned to value expected for fresh, nonirradiated dosimeters.
4. *Signal stability.* An important property of a storage phosphor is the stability of the information stored in the phosphor. It is known that the PSL signal decreases with time, typically consisting of a fast component immediately after irradiation and a slower component afterward.⁶ Signal stability was monitored in this study with accumulated

FIG. 3. Response with cumulated dose up to 5000 Gy. Dose response curves at various dose histories were obtained and plotted, normalized to 500 cGy. A linear dose response holds up to 5000 Gy. Error bars represent the standard error of repeated measurements.

FIG. 4. Reset of sensitivity curve with annealing procedure. Chips with 5000 Gy dose history were annealed in a laboratory furnace at 710 °C for 3 h in air. Dose response at points of 100, 300, and 500 cGy were obtained and compared with chips with no annealing procedure. After annealing, the dose response returns to the supralinear behavior. Error bars represent the standard error of repeated measurements.

dose and results are presented in Table I. Signal stability did not deviate from low dose values outside experimental uncertainty.

III.B. Spectroscopic measurements

1. *PSL stimulation spectra with cumulated dose.* Figure 5 shows the shape of the stimulation curve out to 5000 Gy dose history. The data up to 200 Gy are consistent with previous findings, in which the slight red shift at higher dose histories was attributed to the creation of larger trap centers.³ Importantly, however, Fig. 5 indicates that the detrapping process was not significantly altered by cumulated dose up to 5000 Gy.
2. *PSL emission spectra with cumulated dose.* It was observed that the peak of the PSL emission spectrum remained near its fresh dosimeter peak of 420 nm with dose histories up to 5000 Gy (Fig. 6).
3. *PL spectra and luminescence lifetime.* As shown in Fig. 7, there is a slight broadening (3.5 nm) and peak shift (2 nm) of the PL spectra at high doses; however, in general, the spectra for the different excitation processes (i.e., PL and PSL) agreed well. This data suggests that Eu²⁺ cation acts as the luminescence center in the PSL process via $4f^65d^1 \rightarrow 4f^7(^8S_{7/2})$ transition. The close agreement between the PSL spectra (Fig. 6) and the PL spectra suggests that Eu²⁺ is acting as the photostimulated luminescence center at all dose histories. The combined data also suggest that little changes in the vicinity of the activator have occurred with dose and that the energy transfer mechanism to Eu²⁺ is robust even after high accumulated

FIG. 5. PSL stimulation spectra with cumulated dose. Stimulation wavelength was varied while collecting PSL at 420 nm (see text). The maximum stimulation efficiency for photostimulated luminescence remained constant at 560 nm with cumulated dose.

dose. Figure 8 shows that the photoluminescence lifetime with dose history up to 5000 Gy did not deviate from its fresh value, approximately 1.2 μ s, outside the experimental uncertainty, consistent with the previous findings. According to Zheng *et al.*,¹² a luminescence time of the order of 1 μ s per pixel allows a readout time within a few seconds for a 20×20 cm² panel with 0.5×0.5 mm² pixels. Collectively, these findings, when considered with the recovery of sensitivity after high temperature annealing, suggested that the sensitivity loss with dose after 3000 Gy may be largely due to repairable radiation-induced perturbations in the lattice, such as local lattice distortions, that act as luminescence killers.

IV. DISCUSSION

Designing and developing a new radiation therapy dosimeter is a challenging task, due in part to the unique combination of properties desired of the device. At a basic level, the dosimeter must possess some measurable property that changes with absorbed dose.¹¹ In a clinical environment, however, other properties, such as reusability, accuracy and precision, sensitivity, high spatial resolution, water equivalence, ease of use, and cost are also important. Consequently, once a sensitive material is selected, several proof-of-concept and systematic studies must follow before the device is put into clinical use. Previous works have demonstrated that KCl:Eu²⁺ is an exciting alternative material for reusable, water-equivalent dosimetry. The current study supplements the literature in an important way: it offers the first systematic study of the radiation hardness of KCl:Eu²⁺.

PSL in KCl:Eu²⁺ is a complex process that can be influenced by a number of variables, including irradiation. It has

TABLE I. Signal stability with cumulated dose. Dosimeters were irradiated to several dose levels and the signal stability 13 h after irradiation was examined according to a procedure described in Ref. 3. Signal stability remained largely unchanged after irradiation to greater than 5000 Gy history.

Dose history (Gy)	0	144	630	1770	3000	5000
Signal stability (%decrease/h)	0.24 ± 0.04	0.17 ± 0.06	0.13 ± 0.06	0.13 ± 0.03	0.14 ± 0.05	0.16 ± 0.08

FIG. 6. PSL emission spectra with cumulated dose. KCl:Eu²⁺ dosimeters were stimulated while varying the collection wavelength (see text). The dosimeters emit intense photostimulated luminescence centered at 420 nm, which remained constant with dose history.

been suggested²¹ that a major cause of radiation damage in alkali halide storage phosphors is the creation of large defect centers, for example, M_{Eu}-centers (i.e., two adjacent anion vacancies each occupied by an electron in the neighborhood of an Eu²⁺). Within such a structure, Eu²⁺ ions can easily change sites causing an agglomeration of europium ions and concentration quenching. The results of this study, however, suggest that agglomeration is controlled in a KCl:Eu²⁺ system to at least 5000 Gy dose history. Furthermore, the observed level of radiation hardness of KCl:Eu²⁺ allows the physicist to establish and monitor dosimeter performance characteristics and represents a factor of 25 increase in reusability compared to previous data.³ The difference between the results in Fig. 1 and the previously reported sensitivity loss after even 50 Gy (Ref. 3) could mainly be attributed to improved processing techniques designed to mitigate KCl deliquescence. Water adsorption and incorporation into the microstructure of the as-fabricated dosimeters lowers the electrical impedance of the insulator, which in turn decreases the charge separation capability of the dosimeter. Advanced

FIG. 7. PL spectra with cumulated dose. Broad-band UV light was used to stimulate the KCl:Eu²⁺ dosimeters to emit photoluminescence. The peak of the photoluminescence spectrum remained nearly constant at 420 nm, indicating that little changes in the vicinity of the activator have occurred with dose.

protective coatings, however, provide adequate protection against extraneous moisture. For example, Nakazawa *et al.* reported that using modern coating technologies, similar to the one employed in this study, the moisture-susceptible thallium activated rubidium bromide (RbBr:Tl) storage phosphor maintained initial signal stability characteristics for over 570 days.²⁷

A gradual increase in sensitivity for KCl:Eu²⁺ with dose history to 3000 Gy was observed (Fig. 1), a phenomenon also reported for carbon doped aluminum oxide.¹⁵ In this case, however, it is hypothesized that the increase in sensitivity is due to a combination of an increase in trapping efficiency in photosimulable energy bands and migration of trapped charge species into photostimulable complexes after optical annealing. In addition to preexisting vacancies,^{4,28} according to Itoh irradiation produces vacancies occupied by electrons (F-centers) and interstitial halogen atoms bound to lattice halogens by holes.²⁹ Photostimulation at F-center bands releases stored electrons to produce PSL, but radiation-induced lattice changes remain. During very high escalating doses and subsequent optical annealing, more centers could be created and emptied, resulting in an increase in the number of storage centers for electrons and holes. Charge carriers generated during later irradiations may then have a higher probability of being trapped in photostimulable centers, and thus increasing sensitivity. Additionally, charges or defects remaining after optical annealing to background levels could migrate in the dark at room temperature to form photostimulable complexes, leading to an observed increase in sensitivity.^{15,30} Since the literature related to KCl:Eu²⁺ dosimetry is limited, however, further investigations into the nature of sensitivity change with dose history are needed, as well as a detailed understanding of the optical annealing process.

A decline in sensitivity was observed after 3000 Gy, however, which indicates that competing processes are also at work. It is likely that radiation damage in the crystal lattice becomes significant at this dose level. Consequently, radiation-induced local lattice perturbations could act as luminescence killers, decreasing sensitivity. In this interpretation, irradiation still produces charge carriers that are trapped in photostimulable centers, but radiation damage affects the luminescence process. It is anticipated that sensitivity changes with dose history could be manageable by giving a calibration dose prior to beginning the dose measurement session. In Zheng *et al.* a calculation was performed in order to estimate if a 1 μ m thick panel of KCl:Eu²⁺ in a 1 cGy dose-to-water irradiation would generate a sufficient number of photoelectrons to be detectable by a readout PMT. A 10% reduction in the number of PSL photons generated by a 0.5×0.5 mm² pixel would nevertheless produce over 10 000 photoelectrons at the PMT photocathode under similar assumptions,¹² a number sufficient for detection and amplification.

A linear response to dose is expected of PSL systems.^{5,6} In theory, the number of PSL active centers is proportional to the locally deposited dose. While previous data showed a supralinear response up to 800 cGy,³ the current data shows that the response of KCl:Eu²⁺ dosimeters changes with dose

FIG. 8. Luminescence lifetime with cumulative dose. A 308 nm eximer laser was used to excite KCl:Eu²⁺ PL in dosimeters with dose histories up to 5000 Gy. The PL was sampled during its decay and plotted on a log scale. PL lifetime remained nearly constant at 1.2 μ s within experimental uncertainty.

history, becoming more linear with higher cumulated doses. It is hypothesized that the supralinear response of fresh or nearly fresh dosimeters is due to deep, nonphotostimulable traps present in the material, perhaps as a result of the crude manufacturing process. These deep traps compete with photostimulable traps for electrons generated during charge separation and result in a reduced signal. At greater cumulated doses, the deep electron traps become filled and more electrons are stored in the photostimulable traps, leading to enhanced photostimulated luminescence signal. Once all of the deep traps are filled, a linear response dominates. If this interpretation is correct, a preclinical dose loading protocol would be feasible for ease of calibration. For example, 100 Gy could be given to a fresh dosimeter, the dosimeter optically bleached, and then calibrated for clinical use. This pre-dose should have negligible impact on reusability.

One indication that nonphotostimulable traps are responsible for supralinearity may be the sensitivity curve reset after high temperature annealing. After some delocalization temperature, electrons stored in deep traps could be released. It would follow that these traps would once again be available to compete with PSL traps for radiation-generated electrons. High temperature annealing would also provide thermal energy for lattice atoms to move back to equilibrium positions, which would explain the observed recovery of sensitivity. Since sensitivity recovery is a desirable feature, further systematic studies are necessary to fully understand the effects of thermal annealing and optimize the procedure.

In high speed, two-dimensional computed radiography readout systems, a stimulation laser is focused on a single pixel, the information read out, the laser moved to the next pixel, and the process repeated so that the entire imaging panel is read in the matter of seconds. Since similar technology could be applied to KCl:Eu²⁺ dosimetry panels, it is interesting to note that high irradiation does not significantly alter the spectral or temporal properties of the activator in the KCl matrix. Thus, in clinical use, only the change of sensitivity with dose would need to be monitored. Importantly,

a change in luminescence lifetime was not detectable outside the uncertainty of the experiment. Thus the readout time, as well as the contribution of latent luminescence from neighboring pixels to any given pixel, would not change significantly with dose history.

As the complexity of radiation therapy delivery increases, the need for novel devices to measure and monitor the radiation output of medical devices also increases. As research into KCl:Eu²⁺ continues and technology advances, improvements on the above data are certainly to be expected. One significant area to explore for commercial application of KCl:Eu²⁺ is moisture control and protection. The fabrication process used in this study could easily be incorporated into modern industrial clean room and protected atmosphere systems, which would minimize degradation of sensitive powders during processing. Dosimeters produced in such a protected environment could be further protected with optimized commercial-grade conformal coating technologies.²⁴ These changes could minimize the effects of ambient humidity as well as provide resistance to abrasion during handling. Furthermore, as processing techniques continue to improve, signal stability may also be optimized. Future preclinical studies can then confidently assess in detail other properties of this novel dosimetry material.

V. CONCLUSIONS

This work has presented the first systematic study of how cumulated dose affects the response of KCl:Eu²⁺. A supralinear response to dose for fresh dosimeters becomes more linear with dose history. Approximately 90% of the initial sensitivity remained after doses up to 5000 Gy, which recovered after a high temperature annealing procedure. There were no significant changes in the signal stability, the PSL stimulation spectra, the PSL emission spectra, the PL emission spectra, or the luminescence lifetime with dose. While much work remains to be done, the data presented here support the strong radiation hardness of KCl:Eu²⁺ as well as further investigations of this novel material.

ACKNOWLEDGMENTS

This work was supported in part by NIH Grant Nos. R21CA131690 and R01CA148853. JPD was also supported in part by the Nuclear Science and Engineering Institute of the University of Missouri under a Graduate Assistance in Areas of National Need Fellowship funded by the Department of Education. The authors thank Buck Rogers, Girdhar Sharma, and Dinesh Thotala for coordinating the use of the ¹³⁷Cs irradiator, Baozhou Sun and Logan Ice for technical assistance, and Paul Leblans and Luc Struye (AGFA HealthCare, Belgium) for providing protective coatings.

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