Temporal signal stability of KCl:Eu²⁺ storage phosphor dosimeters

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Purpose: Current KCl:Eu²⁺ prototype dosimeters require a wait time of 12 h between irradiation and dosimetric readout. Although irradiating the dosimeters in the evening and reading on the following day works well in the clinical schedule, reducing the wait time to few hours is desirable. The purposes of this work are to determine the origin of the unstable charge-storage centers and to determine if these centers respond to optical or thermal excitation prior to dosimetric readout.

Methods: Pellet-style KCl:Eu²⁺ dosimeters were fabricated in-house for this study. A 6 MV photon beam was used to irradiate the dosimeters. After x ray irradiation, dosimeters were subjected to external excitation with near-infrared (NIR) light, ultraviolet (UV) light, or thermal treatment. Photostimulated luminescence (PSL) signal's temporal stability was subsequently measured at room temperature over a few hours using a laboratory PSL readout system. The dosimeters were also placed in a cryostat to measure the temperature dependence of the temporal stability down to 10 K.

Results: Strong F-band was present in the PSL stimulation spectrum, indicating that F-centers were the electron-storage centers in KCl:Eu²⁺ where an electron was stored at a chlorine anion vacancy. Due to deep energy-depth (2.2 eV), F-centers were probably not responsible for the fast fading in the first a few hours post x ray irradiation. In addition, weak NIR bands were present. However, there was no change in PSL stabilization rate with intense NIR excitation, suggesting that the NIR bands played no role in the PSL fading. At temperatures lower than 77 K there was almost no signal fading with time. Noticeable PSL was observed for undoped KCl samples at room temperature, suggesting that Cl₂⁻ V_k centers served as hole-storage centers for both undoped and doped KCl where a hole was trapped by a chlorine molecular ion. V_k centers were stable at low temperature and became mobile at room temperature, probably causing the observed PSL fading with time. On the other hand, V_k center could be stabilized by Eu²⁺ activator or oxygen in the lattice, leading to the stable component in the PSL. A thermal process at elevated temperatures (60 °C or higher) was able to significantly accelerate the migration process resulting in a fast stabilization of PSL. However, this could not be accomplished using intense UV excitation.

Conclusions: Thermal treatment enables KCl:Eu²⁺ prototypes to be ready for readout in 1 h without the need of applying a large time-dependent correction factor. However, this cannot be achieved using optical preexcitation. © 2013 American Association of Physicists in Medicine. [<http://dx.doi.org/10.1118/1.4789482>]

Key words: storage phosphor, dosimeter, IMRT

I. INTRODUCTION

High spatial-resolution multidimensional dosimetry is crucial for experimental verification of highly conformal IMRT (intensity modulated radiation therapy) dose distributions with steep gradients.¹⁻³ In particular, the dosimeter should be reusable so that the user is able to *characterize* the dosimeter response and subsequently use that characterization to monitor the detector's suitability for commissioning, verification and other critical measurements. In addition, a dosimeter that is used for commissioning measurements should be usable in a wide variety of phantoms. Recent research⁴⁻⁶ has demonstrated that europium doped potassium chloride (KCl:Eu²⁺) storage phosphor material has the potential to become the physical foundation of a novel, reusable, high spatial-resolution, nearly water-equivalent planar dosimeter with film-like physical makeup.

KCl:Eu²⁺ dosimeters function using photostimulated luminescence (PSL) mechanism, also known as optically stimulated luminescence.⁷⁻⁹ During x ray irradiation, electron-hole pairs are generated in numbers proportional to the local x ray dose. Electron-storage centers, for instance, chlorine vacancies, so-called F⁺ centers, store the electrons producing F(Cl⁻) color centers, and hole-storage centers, for instance, Cl₂⁻ V_k centers store the holes where a hole is trapped by a chlorine molecular ion. During the subsequent photostimulation, the electrons are energetically excited and released from the F-centers, and then recombine with the trapped holes. The energy released in this process excites Eu²⁺ to the 4f⁶5d¹ state with subsequent radiative decay to the 4f⁷ ground state, resulting in characteristic Eu²⁺ luminescence. Charge carriers remaining trapped after readout can be removed with a bright, broadband light and the dosimeter can be used again in the same manner.

An important property of a KCl:Eu²⁺ storage phosphor is the stability of the information stored in the phosphor after irradiation. Previous results have shown that PSL intensity decreases with storage time between irradiation and PSL readout, typically consisting of a fast component after irradiation and a much slower component afterward.⁴ The signal fading with time is likely due to unstable electron or hole-storage centers, or both in the material. Temporal variation has also been reported for other dosimetry materials, for example, radiation sensitive diacetylene monomer in radiochromic film¹⁰ (Type EBT, International Specialty Products, Wayne, NJ) and leucodye/initiator for 3D PRESAGE plastic dosimeter.¹¹ When evaluating BaFBr:Eu²⁺ material-based storage phosphor dosimeters for radiation therapy dosimetry,¹² in order to avoid having to apply a large time-dependent correction, Olch suggested that dosimeters should be readout the day after irradiation so that the delay time difference between calibration and measurement dosimeters will be negligible. For example, a 30 min delay time difference will lead to a small correction factor of approximately 0.2%. A long enough delay time is especially important for IMRT composite measurements where the treatment time is long. Although irradiating the dosimeters in the evening and reading on the following day works well in the clinical schedule, reducing the wait time to few hours is desirable.

The purposes of this work are to determine the origin of the unstable charge-storage centers and to determine if these centers respond to optical or thermal excitation prior to dosimetric readout. Our data demonstrate that a 15 min thermal treatment at 60 °C or higher can effectively eliminate the problematic fast fading-rate component of the PSL signal. As a result, the delay time can be reduced to 1 h. A model has been proposed to explain the signal fading phenomenon associated with KCl:Eu²⁺ material.

II. MATERIALS AND METHODS

II.A. Dosimeters and irradiation

The prototype KCl:Eu²⁺ dosimeters used in this study were manufactured according to standard materials processes and had a physical makeup similar to thermoluminescent dosimeter chips. High purity KCl (99.995%, Sigma-Aldrich) particles and reagent grade EuCl₃ · 6H₂O particles were mixed at a 500 ppm Eu²⁺ concentration. Powders were pressed at 2200 lbs force in a hydraulic press (Carver Inc., Wabash, IN) using an evaluable 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. Undoped KCl samples were also prepared. Great care was taken to avoid cross-contamination. X-ray diffraction (XRD) analysis was used to determine the phase composition of the dosimeters. Optical annealing with a 500 W tungsten-halogen lamp prior to irradiation removed background signal created through environmental irradiation. Dosimeters were irradiated to 2 Gy using a 6 MV beam from a Varian medical linear accelerator.

II.B. Dosimetric readout

The readout light source consisted of either a 2 mW, 594 nm He–Ne laser (Melles Griot, Covina, CA) or a 150 W, ultraviolet (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. Readout 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 signal (420 nm wavelength with 10 nm FWHM, full-width-half-maximum) was separated from readout light using a narrow bandpass filter (FB420-10, Thorlabs, Newton, NJ) and detected by a photomultiplier tube (PMT) (R1924A, Hamamatsu, Bridgewater, NJ) connected to a lock-in amplifier (Model SR830, Stanford Research Systems). For laser source, the readout power was attenuated by neutral density filters to allow for repeated readings. The signal temporal stability of dosimeters was monitored using the procedure explained in Han *et al.*⁴ This procedure calls for probing the signal at various times postirradiation and correcting the data for readout signal loss. The dosimeters were also placed in a cryostat (Model CCS-450, JANIS, Wilmington, MA) to measure temporal stability's temperature dependence down to 10 K.

II.C. External excitation prior to dosimetric readout

Immediately after x ray irradiation and prior to dosimetric readout, dosimeters underwent uniform external excitations, either optical or thermal. Both near-infrared (NIR) excitation light (a 75 W tungsten-halogen light source filtered by a 780 nm long pass filter) and UV excitation light (a 150 W Xe-lamp filtered by either 340 or 390 nm band pass filters with 10 nm FWHM) were used for optical excitation. For thermal excitation, a laboratory oven (Binder, Bohemia, NY) was used to heat the dosimeters for 15 min at various temperatures up to 120 °C.

III. RESULTS AND DISCUSSION

Figure 1 displays PSL stimulation spectra (i.e., PSL emission intensity as a function of stimulation wavelength) for KCl:Eu²⁺ at room temperature. The inset shows the magnified stimulation spectrum in the NIR region. Besides the commonly observed F band centered at 560 nm (2.2 eV),⁷ the spectra show two stimulation bands in the NIR range at 822 nm (1.5 eV) and 880 nm (1.4 eV), respectively; however, PSL intensity was 2 orders of magnitude less. These two NIR bands suggest that another type of electron-storage center, other than the well-known, deep-seated F-center (an electron stored at a chlorine anion vacancy), may exist in the forbidden band with shallow energy level. As hypothesized by other luminescence dosimeter studies,^{4,13} these low-level electron-storage centers could be removed by incorporating a pre-read cycle that optically bleaches the low-level traps while not depleting the stable, higher energy dosimetric traps, i.e.,

FIG. 1. PSL stimulation spectra for KCl:Eu²⁺ showing the F band centered at 560 nm. The PSL emission was monitored at 420 nm. Inset: PSL stimulation spectrum for NIR region with two low-intensity bands centered at 822 and 880 nm.

F-centers. However, as shown in Fig. 2, there was almost no change in PSL stabilization rate under intense NIR excitation, suggesting that the two NIR bands played no role in PSL fading. F-centers have a depth of 2.2 eV, which is too deep for thermal stimulation at room temperature. Therefore, the fast fading component is probably not due to the thermal release of the stored electrons in the F-centers.

Figure 3 shows KCl:Eu²⁺ temporal stability's temperature dependence from 297 K down to 10 K. Dosimeters were irradiated by a Linac at room temperature, immediately transferred to the optical cryostat, and subsequently cooled down to the prescribed temperature. The cryostat requires approximately 2 h to reach 10 K and less time for other higher temperatures. Almost no signal fading in PSL was observed at 10 K from 2 h and later postirradiation. Fading was observed at

FIG. 2. PSL fading curves after NIR optical excitation for various time periods. The stabilization rate remains the same regardless of NIR excitation time.

FIG. 3. PSL fading curve at various temperatures. These dosimeters received no optical or thermal excitation. Almost no signal fading in PSL was observed at 10 K 2 h postirradiation and beyond. Noticeable fading was observed at temperatures greater than 77 K. However, the PSL remained relatively stable until 260 K compared to room temperature. Inset: PSL intensity as a function of temperature. The line is added to guide the eye.

temperatures higher than liquid-nitrogen temperature (77 K); however, even at 260 K, the PSL intensity appeared quite stable compared to that at room temperature. These data suggest that one type of charge-storage center is stable at relatively low temperatures but becomes mobile with increasing temperature. Since deep-seated F-centers can be excluded as previously discussed, we tentatively attribute these centers to hole-storage centers, i.e., $\text{Cl}_2^- \text{V}_k$ centers where a radiation-generated hole is trapped by a chlorine molecular ion.^{14,15} It has been reported that V_k centers serve as hole-storage centers at low temperature. However, V_k centers become mobile with increasing temperatures.¹⁴⁻¹⁷ A mobile V_k center can be trapped by an Eu²⁺ activator or other impurities and become stable at room temperature, which contributes to the stable PSL signal. On the other hand, V_k center can also encounter an electron-storage center (F-center) to recombine, resulting in a decrease of the number of available charge-storage centers and consequently a drop in PSL sensitivity upon photostimulation with increasing temperature (Fig. 3, inset). The life time of this thermally mediated process could be long, several hours in our case, which may account for the fading in the first a few hours postirradiation. In order to determine if V_k centers can serve as hole-storage centers at room temperature, the response of undoped KCl to x ray irradiation was measured. Figure 4 shows the PSL emission spectrum stimulated by an intense He-Ne laser for undoped KCl after irradiation to 200 Gy. Clearly, there is a luminescence emission peak centered at 430 nm, which could be attributed to the recombination of F-center electrons with holes stored at the $\text{Cl}_2^- \text{V}_k$ centers, or e- V_k recombination.¹⁶⁻¹⁹ The electron configuration of chlorine in KCl is $3s^2 3p^6$. During x ray irradiation, one p-electron is liberated, leaving behind a hole that could subsequently be trapped by a chlorine molecular ion forming a $\text{Cl}_2^- \text{V}_k$ center. Appleby *et al.*¹⁹ reported similar e- V_k emission in pure CsBr, another important

FIG. 4. Emission spectrum for undoped KCl after x ray irradiation. The sample was stimulated with an intense 594 nm He-Ne laser. A significant luminescence emission peak centered at 430 nm was observed. This emission could be due to the recombination of F-center electrons and holes stored at the Cl_2^- V_k centers, or e - V_k recombination.

alkali-halide storage phosphor for computed radiography application. The data suggest that even undoped KCl is capable of storing radiation-generated electrons and holes giving off PSL via a photostimulation process. Note that the PSL intensity was several orders of magnitude lower than that of KCl:Eu²⁺ for the same given dose due to the absence of Eu²⁺ activator. The lack of the Eu²⁺ activator results in the majority of the energy released in the e - V_k recombination in pure KCl being transferred nonradiatively to the lattice. Figure 5 shows similar fading behaviors for doped and undoped KCl. Although no europium cations exist in the undoped KCl, other impurities in the lattice, such as oxygen incorporated into the lattice during fabrication process, could stabilize a portion of the V_k centers,²⁰ which may explain the stabilized PSL after 12 h. Furthermore, XRD data (Fig. 6) revealed that europium

FIG. 5. PSL fading curves for undoped KCl and KCl:Eu²⁺. Although no europium cations exist in undoped KCl, other impurities in the lattice, such as oxygen incorporated into the lattice during fabrication process, could stabilize a portion of the V_k centers.

FIG. 6. X-ray diffraction spectra of undoped KCl and KCl:Eu²⁺ showing that europium was completely incorporated into the KCl parent-matrix without the formation of a noticeable secondary phase.

was completely incorporated into the KCl parent-matrix without the formation of a noticeable secondary phase. Therefore, the impact of possible secondary phases can be excluded.^{21,22} Collectively, the data support that similar to undoped KCl, V_k centers can serve as hole-storage centers in KCl:Eu²⁺ at room temperature; thus could be responsible for the fading due to increased mobility at room temperature. Moreover, the V_k centers' temperature-dependent mobility (Fig. 3) suggests that a thermal process at elevated temperatures may accelerate V_k centers' migration process, thus eliminating the fast fading in the first a few hours postirradiation.

A proposed model for signal fading in KCl:Eu²⁺ is depicted in Fig. 7. The V_k centers (Cl_2^- molecular ion with a

FIG. 7. Proposed model for signal fading in KCl:Eu²⁺. Hole-storage Cl_2^- V_k centers are likely responsible for fast PSL fading in the first a few hours postirradiation due to high mobility at room temperature until they are stabilized by either Eu²⁺ activators forming a V_k -Eu²⁺ complex or other impurities, for instance, oxygen.

TABLE I. PSL signal stability and PSL sensitivity at various thermal treatment conditions after irradiation. Note that the reported fading rate per hour is an average over 0.5–24 h postirradiation for 60 °C and higher.

Thermal excitation temp.	20 °C	50 °C	60 °C	70 °C	80 °C	90 °C	120 °C
Fading rate per hour	13.6% (first 2 h)	1.7% (first 4 h)	0.44%	0.37%	0.41%	0.18%	0.08%
Sensitivity after 15 h	100%	93%	91%	83%	72%	60%	18%

trapped hole, shown in the lower right corner) are stationary at low temperatures. Due to the lack of thermal energy, recombination of the V_k centers with the F-centers (chlorine vacancy with a trapped electron shown in the upper left corner) is unlikely. As the temperature increases, V_k centers become mobile and are more likely to encounter F-centers. The interaction of mobile V_k centers and F-centers results in recombination and a loss of overall PSL signal. Thus we tentatively attribute the fast fading component of the PSL immediately after irradiation to the interaction of mobile V_k centers and F-centers. However, a portion of the V_k centers can be stabilized by forming complexes with Eu^{2+} activators ($V_k\text{-Eu}^{2+}$) or impurities, such as oxygen. These complexes will not recombine with F-centers until optically stimulated and are attributed to the stable component of the fading curve 12 h postirradiation.

As shown in Fig. 8, a 15-min thermal excitation immediately after x ray irradiation resulted in a remarkable improvement in signal stabilization rate. A temperature as low as 50 °C lowered the fading rate to 1.7%/h for the first 4 h after irradiation, and reached almost the same fading rate thereafter as a dosimeter would have after 12 h postirradiation without thermal intervention. Thermal treatments at 60 °C stabilized the PSL (Table I) to a slow fading rate of approximately 0.4%/h over a time period of 24 h. Moreover, 90% of the PSL sensitivity can be retained. Greater improvements in fading rate (0.18 and 0.08%/h fading rate for 90 and 120 °C ther-

mal annealing, respectively) can be achieved at the expense of PSL sensitivity (60% and 18%, respectively). A calculation was performed by Zheng *et al.*⁵ in order to estimate if a 1 μm thick KCl:Eu^{2+} panel (under development) in a 1 cGy dose-to-water irradiation would generate a sufficient number of photoelectrons to be detectable by a readout PMT. Even a large reduction, for example, a 60% reduction in the number of PSL photons generated by a $0.5 \times 0.5 \text{ mm}^2$ pixel would nevertheless produce over 5000 photoelectrons at the PMT photocathode under similar assumptions. Also, the fabrication process of the dosimetry material could be further optimized to improve intrinsic sensitivity.

Templer *et al.*^{23,24} have measured the thermal trap depths for BaFBr:Eu^{2+} which are filled during room-temperature irradiation, the shallowest being $0.89 \pm 0.02 \text{ eV}$. According to the Franck-Condon principle, equivalent trap depth for photostimulation is greater, and could be as large as 2.9 eV, which is about in the UV range.¹⁴ Therefore, we further studied if intense UV light excitation could lead to the same result as thermal preannealing for KCl:Eu^{2+} . In a separate experiment, the same batch of dosimeters were bleached, irradiated, optically excited using intense UV light, and then read. Similar results were found as those for the NIR excitation; that is, there was almost no change in stabilization rate (Fig. 9). This suggests that the accelerated thermal-release at elevated temperatures in KCl:Eu^{2+} cannot be accomplished using photons in the UV range.

FIG. 8. PSL fading curves after 15 min thermal treatments at various temperatures. A short thermal treatment at 60 °C can remarkably improve the signal stabilization rate.

FIG. 9. PSL fading curves after UV optical bleaching for various time periods. Optical bleaching with UV light did not improve the PSL stabilization rate.

IV. CONCLUSIONS

This work studied the effects of different treatments on the KCl:Eu²⁺ dosimeter's signal fading rate prior to dosimetric readout. Optical excitation with NIR or UV light did not reduce the time needed to reach the slow PSL fading rate. A short thermal treatment at 60 °C reduced the postirradiation fading rate to 0.4%/h while maintaining 90% of the PSL intensity if without intervention. Our results suggest that incorporating a thermal preread cycle can reduce the time to readout from 12 h to just 1 h. Hole-storage V_k centers were probably responsible for fast PSL fading in the first a few hours postirradiation due to high mobility at room temperature until they were stabilized by either Eu²⁺ activators forming a V_k-Eu²⁺ complex or other impurities, for instance, oxygen.

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