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Inorganic scintillation detectors based on Eu-activated phosphors for ^{192}Ir brachytherapy

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Abstract

The availability of real-time treatment verification during high-dose-rate (HDR) brachytherapy is currently limited. Therefore, we studied the luminescence properties of the widely commercially available scintillators using the inorganic materials Eu-activated phosphors $\text{Y}_2\text{O}_3\text{:Eu}$, $\text{YVO}_4\text{:Eu}$, $\text{Y}_2\text{O}_2\text{S:Eu}$, and $\text{Gd}_2\text{O}_2\text{S:Eu}$ to determine whether they could be used to accurately and precisely verify HDR brachytherapy doses in real time. The suitability for HDR brachytherapy of inorganic scintillation detectors (ISDs) based on the 4 Eu-activated phosphors in powder form was determined based on experiments with a ^{192}Ir HDR brachytherapy source. The scintillation intensities of the phosphors were 16 to 134 times greater than that of the commonly used organic plastic scintillator BCF-12. High signal intensities were achieved with an optimized packing density of the phosphor mixture and with a shortened fiber-optic cable. The influence of contaminating Cerenkov and fluorescence light induced in the fiber-optic cable (stem signal) was adequately suppressed by inserting between the fiber-optic cable and the photodetector a 25-nm band-pass filter centered at the emission peak. The spurious photoluminescence signal induced by the stem signal was suppressed by placing a long-pass filter between the scintillation detector volume and the fiber-optic cable. The time-dependent luminescence properties of the phosphors were quantified by measuring the non-constant scintillation during irradiation and the afterglow after the brachytherapy source had retracted. We demonstrated that a mixture of $\text{Y}_2\text{O}_3\text{:Eu}$ and $\text{YVO}_4\text{:Eu}$ suppressed the time-dependence of the ISDs and that the time-dependence of $\text{Y}_2\text{O}_2\text{S:Eu}$ and $\text{Gd}_2\text{O}_2\text{S:Eu}$ introduced large measurement inaccuracies. We conclude that ISDs based on a mixture of $\text{Y}_2\text{O}_3\text{:Eu}$ and $\text{YVO}_4\text{:Eu}$ are promising candidates for accurate and precise real-time verification technology for HDR BT that is cost effective and straightforward to manufacture. Widespread dissemination of this technology could lead to an improved understanding of error types and frequencies during BT and to improved patient safety during treatment.

1. Introduction

The availability of real-time treatment verification during high-dose-rate (HDR) brachytherapy (BT) is currently limited. As of 2010, only about 20% of all BT clinics in

Europe had access to *in vivo* dosimetry equipment (Guedea *et al* 2010). As a consequence, errors that can affect BT (Anon 2000, Lopez *et al* 2000, Ashton *et al* 2004) can go unnoticed. Given the steep dose gradients near BT sources and the high-dose regimens of HDR BT, systematic source misplacement during treatments can result in harmful consequences for patients.

A scintillation detector consists of a scintillator volume optically coupled with a fiber-optic cable, which transmits the radiation-induced light emitted by the scintillator to a photodetector for readout. Scintillation detectors have shown a strong potential to provide real-time treatment verification during BT (Tanderup *et al* 2013, Kertzscher *et al* 2014). For example, detectors based on the blue-emitting $\text{Al}_2\text{O}_3\text{:C}$ crystal (Andersen *et al* 2011, Kertzscher *et al* 2011a, Beierholm *et al* 2008) exhibit the high signal intensities that are required for monitoring the absorbed dose rate with small statistical uncertainties over the wide dynamic range that is common with BT sources (Tanderup *et al* 2013, Kertzscher *et al* 2011b). However, $\text{Al}_2\text{O}_3\text{:C}$ crystals must be pre-irradiated with 20 Gy to generate a stable scintillation signal (Andersen *et al* 2011), which requires added time investments and therefore limits the practical incorporation of the technology into clinical workflows. Several other inorganic scintillators also exhibit large light yields (Derenzo *et al* n.d.) and may not require pre-irradiation before *in vivo* dosimetry. Some inorganic scintillators, therefore, may be useful in monitoring BT doses without substantially interrupting clinical workflows.

Inorganic scintillation detectors (ISDs) are scintillation detectors that incorporate an inorganic material as the scintillator detector volume. Several inorganic scintillators with large light yields are commercially available. In addition, ISDs are promising for BT applications because many inorganic scintillators emit in the red wavelength region, in which contaminating Cerenkov and fluorescence light induced in the fiber-optic cable (Beddar *et al* 1992a, Theriault-Proulx *et al* 2013) is less prominent than with blue-emitting scintillators. If the light yield is sufficiently large, a band-pass filter centered at the red emission peak can be used to suppress the Cerenkov and fluorescence light contamination in 1 optical channel (Kertzscher and Beddar 2016). In contrast, organic plastic scintillation detectors (PSDs) (Beddar *et al* 1992b, Beddar *et al* 1992c) require background signal suppression methods that add complexity to the readout (Fontbonne *et al* 2002, Theriault-Proulx *et al* 2011a, Kertzscher *et al* 2011a, Beierholm *et al* 2011, Boer *et al* 1999, Archambault *et al* 2006). Our research is presently focused on developing cost-effective red-emitting ISDs that can provide accurate and precise real-time treatment verification during BT.

However, inorganic scintillators can exhibit non-constant scintillation and afterglow effects that alter the proportionality of the scintillator response to the absorbed dose and therefore compromise measurement accuracy. For instance, ruby crystals can, depending on the admixture of impurities, exhibit significant non-constant effects (Kertzscher and Beddar 2016). Similar effects have occurred with ZnSe:Te scintillators (Ramírez *et al* 2016). Therefore, to ensure accurate absorbed-dose measurements, the absence of non-constant effects must be established before an inorganic scintillator can be used in BT. If the absence of non-constant effects can be established, inorganic scintillators with large light yields could be used in simplified detector systems that can be easily distributed to BT clinics. If

such ISDs become widely used, real-time dose verification during HDR BT can advance our understanding of error types and frequencies and improve patient safety.

The widely commercially available Eu-activated inorganic scintillators $\text{Y}_2\text{O}_3:\text{Eu}^{3+}$, $\text{YVO}_4 2:\text{Eu}^{3+}$, $\text{Y}_2\text{O}_2\text{S}:\text{Eu}^{3+}$ and $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}^{3+}$ in powder form (micrometer-sized crystals) emit narrow emission spectra in the red wavelength region and could therefore potentially exhibit suitable characteristics in detector systems for BT. These Eu-activated scintillators have been studied for use with various external-beam radiation sources (Martinez *et al* 2015, Molina *et al* 2013, 2012, Souris *et al* 2014, McCarthy *et al* 2014), but their suitability for use with ^{192}Ir BT sources has yet to be established. Investigators have studied detectors using nanometer-sized Y_2O_3 crystals co-doped with Eu and Li (Stanton *et al* 2014) for external-beam sources (Belley *et al* 2015b) and for ^{192}Ir HDR BT (Belley *et al* 2015a). To our knowledge, the light yield and other luminescence properties of crystals co-doped with Eu and Li have not been compared with those of commercial Eu-activated phosphors.

The luminescence properties of Eu^{3+} activated phosphors have been extensively described in literature (Blasse and Grabmaier 1994, Yen *et al* 2007, Binnemans 2015). Their prominent emission lines originate from energy transitions with a forbidden character and have lifetimes of about 10^{-3} s. However, the phosphors also undergo other mechanisms that introduce additional lifetimes that are even longer. For instance, luminescence centers can, during irradiation, populate quasi-stable excited states with longer lifetimes before being transferred to an emitting state by thermal activation. These mechanisms cause delayed light emission, or afterglow, which generates spurious signals when the irradiation dose rate decreases drastically (e.g., when the BT source moves away from the detector) or when the irradiation is terminated. Significant afterglow signals with greater than 0.1-s lifetimes compromise the accuracy of real-time radiation detectors during BT because they generate overestimated measurements after the source is retracted or moved to a new dwell position farther from the detector. Furthermore, the thermal activation of the mechanisms is time-dependent. If substantial, these mechanisms may contribute to the non-constant emission of light and thereby potentially alter the real-time measurements significantly during the period of time required for the activation process to stabilize. As a result, each time the BT source is moved to a new dwell position, the dose measurement is inaccurate for a duration that is characteristic of each phosphor.

The Eu^{3+} activated phosphors exhibit photoluminescence, which corresponds to light emission in response to the absorption of ultraviolet light. For ISDs, this ultraviolet light can originate from the fiber-optic cable to which the phosphor is attached, i.e. from the ultraviolet portion of the Cerenkov and fluorescence light emitted in fiber-optic cables when exposed to ^{192}Ir BT sources (Beddar *et al* 1992a, Therriault-Proulx *et al* 2013). The photoluminescence and the Cerenkov and fluorescence light are both proportional to the absorbed dose in the fiber-optic cable rather than in the detector and are therefore considered a background signal. Both sources of background can generate significant inaccuracies in the measured dose when the BT source is positioned close to the fiber-optic cable and far from the scintillator detector volume (Andersen *et al* 2009, Therriault-Proulx *et al* 2011a, Kertzscher *et al* 2011a, Kertzscher and Beddar 2016).

Measurements by Molina *et al* (2013), Pittet *et al* (2013) and Martinez *et al* (2015) show that the sensitivity to low-energy radiation is higher for ISDs with high Z_{eff} than for ionization chambers. Their measurements demonstrate that the ISD response increases with respect to that of the ion chamber as the lower energy secondary radiation component becomes more dominant compared with the higher energy primary radiation, which results in overestimated absorbed doses to water. Since the photon energy spectrum in water shifts towards lower values as the distance from a ^{192}Ir source increases (Meigooni *et al* 1988), absorbed dose measurements using ISDs are expected to be overestimated as the BT source moves towards positions farther away from the ISD. This energy dependence of ISDs needs to be characterized and corrected for, e.g. using a correction curve as described in Reniers *et al* (2012).

The purpose of this study was to assess the luminescence properties of commercial $\text{Y}_2\text{O}_3:\text{Eu}^{3+}$, $\text{YVO}_4:\text{Eu}^{3+}$, $\text{Y}_2\text{O}_2\text{S}:\text{Eu}^{3+}$ and $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}^{3+}$ scintillators in powder form (micrometer-sized crystals) to determine whether they can be used in ISDs that accurately and precisely measure absorbed dose in real time during HDR BT. We assessed the luminescence properties based on measurements using an ^{192}Ir HDR BT source to determine signal intensities of the scintillators, the influence of the photoluminescence background and the Cerenkov and fluorescence light background on the measurement accuracy, and the influence of non-constant scintillation and afterglow effects. The scintillation strength was compared with that of commonly used organic scintillators to provide with a reference. We also tested strategies to optimize the characteristics of the ISDs.

2. Materials and methods

The potential suitability of the Eu-activated inorganic scintillators $\text{Y}_2\text{O}_3:\text{Eu}^{3+}$, $\text{YVO}_4:\text{Eu}^{3+}$, $\text{Y}_2\text{O}_2\text{S}:\text{Eu}^{3+}$, and $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}^{3+}$ in powder form for ^{192}Ir BT applications was determined based on experiments using an ^{192}Ir HDR BT source. The influence of the non-constant scintillation and afterglow effects, the signal intensity, the photoluminescence, and the Cerenkov and fluorescence light background on the measurement accuracy was assessed. Two commonly used organic scintillators, BCF-12 and BCF-60, were used as the references for measurements of the scintillation intensities of the Eu-activated phosphors. Characterizing the energy and temperature dependence of the scintillators was not within the scope of this study.

Throughout the remainder of this article, the superscript “3+” will be omitted from the names of the Eu-activated phosphors, radioluminescence and scintillation will be referred to as scintillation, and the Cerenkov and fluorescence signal will be referred to as the stem signal.

2.1. Detector probe designs and signal acquisition

Each ISD and PSD consisted of a 1-mm-long and 1-mm-diameter cylindrical scintillator optically coupled to a 1-mm-diameter fiber-optic cable (ESKA GH-4001; Mitsubishi Rayon Co. Ltd., Japan) made of poly(methyl methacrylate) (PMMA). The fiber-optic cable transmitted the light to a charge-coupled device (CCD) camera (Luca S 658M; Andor Technology Ltd., UK) or a spectrometer spectrograph (Shamrock SR-163i; Andor

Technology Ltd.). The detector volume of the ISDs consisted of the red-emitting Eu-activated phosphors $\text{Y}_2\text{O}_3\text{:Eu}$, $\text{YVO}_4\text{:Eu}$, $\text{Y}_2\text{O}_2\text{S:Eu}$, or $\text{Gd}_2\text{O}_2\text{S:Eu}$ in powder form (product numbers QK63/N-C1, QHK63/N-U1, QKL63/N-C1, and UKL63/N-R1, respectively; Phosphor Technology Ltd., UK). The PSDs used the blue-emitting BCF-12 and green-emitting BCF-60 organic scintillators (Saint-Gobain Crystals, France) (table 1). The organic scintillators BCF-12 and BCF-60 were made of polystyrene (C_8H_8).

Three detector probe types were manufactured (figure 1). To manufacture the PSD based probe type A, a scintillator was coupled directly with the fiber-optic cable using transparent glue (NOA68; Norland Products Inc., USA) that was cured with ultraviolet light. The scintillator volumes of probe types A, B and C that were based on the Eu-activated phosphors, were prepared by mixing 2 mass units of phosphor powder with 1 mass unit of transparent glue. The masses of the phosphor powder and the glue were measured with a scale (GeminiPRO Digital Milligram Scale, American Weigh Scales Inc., USA). The mixture was placed inside a 1-mm-long and 1-mm-diameter piece of transparent tubing (103-0292; Vention Medical Inc., USA). The mixture was cured directly onto the fiber-optic cable for probe type A and B. For probe type C, the mixture was cured onto a thin 1-mm-diameter circular plastic long-pass optic filter that was coupled with the fiber-optic cable using ultraviolet-cured glue.

The scintillation detector volumes of probe types B and C were covered with reflective paint (EJ-510; Eljen Technology, USA) to enhance the signal intensity. As described by Jordan (1996) and Kertzscher and Beddar (2016), the long-pass optic filter placed between the fiber-optic cable and the scintillator detector volume suppressed the generation of photoluminescence in the Eu-activated phosphor by preventing the ultraviolet component of the stem signal from reaching the phosphor. A long-pass filter (E-Colour+ 105; Rosco Laboratories Inc., USA) that transmitted about 84% for wavelengths greater than 600 nm, 50% at 563 nm, and less than 10% at under 511 nm was used for the $\text{Y}_2\text{O}_3\text{:Eu}$ - and $\text{YVO}_4\text{:Eu}$ -based ISDs. A Supergel 23 filter (Rosco Laboratories Inc.), which transmitted about 85% for wavelengths greater than 610 nm, 50% at 582 nm, and less than 10% at under 539 nm, was used for the $\text{Y}_2\text{O}_2\text{S:Eu}$ - and $\text{Gd}_2\text{O}_2\text{S:Eu}$ -based ISDs. The filters blocked wavelengths that matched the broadband photoabsorption spectra of the $\text{Y}_2\text{O}_3\text{:Eu}^{3+}$, $\text{YVO}_4\text{:Eu}^{3+}$, $\text{Y}_2\text{O}_2\text{S:Eu}^{3+}$ and $\text{Gd}_2\text{O}_2\text{S:Eu}^{3+}$ that, according to the manufacturers, peak at 255, 325, 345, and 355 nm and range up to 330, 400, 400, and 400 nm, respectively.

The PMMA core of the fiber-optic cable was surrounded by cladding with a lower refractive index than that of the core to facilitate internal reflection, and the cladding was surrounded by a jacket by polyethylene to protect the core and cladding from mechanical stress and contamination by ambient light. The probe tip was cast in black epoxy to protect the coupling from mechanical stress and ambient light.

With an aim to suppress the time dependent luminescence properties of the ISDs, we manufactured detectors where the scintillation detector volumes consisted of a mixture between the $\text{Y}_2\text{O}_3\text{:Eu}$ - and $\text{YVO}_4\text{:Eu}$ -phosphors (it will become apparent why those two phosphors were chosen in the Results section). We manufactured 2 to 3 detectors for 4 different ratios between the $\text{Y}_2\text{O}_3\text{:Eu}$ - and $\text{YVO}_4\text{:Eu}$ -phosphors to determine which ratio

generated the smallest time-dependence. Subsequently, we manufactured 6 ISDs based on the $\text{Y}_2\text{O}_3\text{:Eu-YVO}_4\text{:Eu}$ -ratio that generated the smallest time-dependence. Five of the 6 ISDs incorporated reflective paint (type B ISDs) and were manufactured with different ratios between the glue and the phosphor, to determine which ratio generated the largest signal intensity. Only one of the 6 ISDs was a type C detector which incorporated reflective paint and an optic filter (E-Colour+ 105; Rosco Laboratories Inc.) which allowed us to determine the emission spectrum of the type C ISD.

Table 2 lists the 48 detector probes that were manufactured for this study. All PSDs and most ISDs incorporated 15-m-long fiber-optic cables that, during experiments, were directly connected with the photodetector for readout. Three probe type A ISDs with 2-m-long fiber-optic cables were also manufactured for each Eu-activated phosphor. The 12 type A probes were optically coupled with 12-m-long fiber-optic cables that was connected with the photodetector during experiments. Transparent optical gel (Optical Gel 0607, product number 24230; Cargille Laboratories, USA) was used to improve the optical coupling between the 2-m ISDs and the 12-m-long fiber-optic cable. Similarly, 9 probe type A ISDs with 20-cm-long fiber-optic cables were manufactured with different ratios between the $\text{Y}_2\text{O}_3\text{:Eu}$ - and $\text{YVO}_4\text{:Eu}$ -phosphors. One 2-m and two 15-m probes composed of a fiber-optic cable without a scintillator, referred to as bare fibers, were used to measure the influence of the stem effect.

2.2. Measurements

All experiments were performed using a Nucletron MicroSelectron v2 HDR afterloader (Elekta, Sweden) that contained an ^{192}Ir BT source with an air-kerma strength of 17.6–40.0 $\text{mGy m}^2 \text{h}^{-1}$. The cylindrical ^{192}Ir core of that source is 0.6 mm in diameter and 3.5 mm long and it is housed inside a steel capsule with a 0.9 mm outer diameter (Granero *et al* 2011).

2.2.1. Signal intensities—To determine the relative signal intensities of the Eu-activated phosphors and BCF-12 and BCF-60 scintillators, the signal intensities were measured for all ISDs and PSDs. The measurements were performed with the ISDs, PSDs, and bare fibers placed one by one 2 cm from the BT source in a rigid polymer-based phantom (Solid Water; Gammex, USA). The measurements taken with the bare fibers were subtracted from those taken with the ISDs and PSDs to remove the contribution of the stem signal to the total signal. The relative signal intensities of the Eu-activated phosphors were determined based on signal measurements using the 2-m-long probe type A ISDs coupled with the 12-m fiber-optic cable. Measurements of the probe type B ISDs based on $\text{Y}_2\text{O}_3\text{:Eu}$ and the BCF-12- and BCF-60-based PSDs were performed to compare the signal intensities of the ISDs with those of the PSDs.

The detector signals were collected with the CCD camera and recorded per unit time. The signal of each detector was determined from 5 consecutive images acquired under identical irradiation conditions (i.e., without retracting the source). The exposure times were set to 10 s for the PSDs and 3 s for the ISDs, and the variability of the detector response was less than 0.3% for the PSDs and ISDs. The region of interest on the CCD chip was defined as a $140 \times$

140-pixel region that captured the entire projected signal spot (Klein *et al* 2011). The median signal value of each pixel in the 5 images was used to eliminate the impact of potential outlier pixel values caused by transient noise from, for example, cosmic rays incident on the CCD camera (Archambault *et al* 2008). The sum of all the median pixel values was the measured signal strength. The total measurement uncertainty attributable to the positional uncertainty of the detectors and the BT source, repeated connections between the detectors and CCD camera, and variability of the detector response was less than 5%. The measurements were corrected to compare the signal intensities between the PSDs and ISDs, for uniform quantum efficiency of the CCD camera and for 0- to 15-m attenuation in the PMMA fiber-optic cable.

2.2.2. Scintillation and background signal components—To quantify the photoluminescence and stem signal backgrounds for the ISDs, the scintillation, photoluminescence, and stem signal components of the total signals for the ISDs and PSDs were measured using the experimental setup we previously described in detail (Kertzscher and Beddar 2016). These measurements were used to determine the relative magnitudes of the signal components for BT source positions along the axis of the detector.

The emission spectrum of the scintillation signal was measured using the spectrometer for each ISD and PSD in phantom experiments in which the scintillation detector volume was positioned 0.5 cm from the BT source. The influence of the stem signal was removed from the ISD and PSD emission spectra by subtracting the spectrum measured using the bare fiber from the spectra measured using the ISDs and PSDs. The stem-corrected signal intensities, which were measured using the CCD camera, and the absorbed dose in the detector, which was calculated with the TG-43 protocol (Rivard *et al* 2004), were used to determine the signal intensity per absorbed dose for each ISD and PSD. The stem-corrected emission spectra of each ISD and PSD were normalized to the corresponding signal intensity per absorbed dose to obtain the wavelength-dependent scintillation signal intensity per absorbed dose.

The wavelength-dependent stem signal intensity per absorbed dose was determined using the same procedure that was used to obtain the intensity of the scintillation signal. However, the emission spectrum and intensity of the stem signal were measured using the bare fiber, and the absorbed dose in the fiber-optic cable was calculated as a line integral of point doses using the TG-43 protocol.

The emission spectrum of the photoluminescence signal was measured by irradiating the fiber-optic cable of the ISD while shielding the scintillator detector volume in a Pb container. The measurements were performed with probe type B ISDs to determine the influence of the photoluminescence signal on the total signal and with type C ISDs to assess the photoluminescence suppression in the filtered detectors. The measurements were repeated using the bare fiber to subtract the stem signal. The photoluminescence spectra were normalized to the signal intensity of the photoluminescence signal per absorbed dose in the fiber-optic cable.

The normalized emission spectra of the scintillation, photoluminescence, and stem signals were used to determine the relative contribution of each signal component for specific wavelength regions and for BT source positions along the axis of each detector. The spectra were corrected for the quantum efficiency of the CCD camera and the spectrometer system and calculated for 0-, 2-, 5-, 10-, and 15-m attenuation in the PMMA fiber-optic cable. Using the geometrical conventions of the TG-43 protocol (Rivard *et al* 2007) where the source is positioned at the origin, the signal components were determined for detector positions from -10 to 10 cm in the z-direction and at 0.5, 1.0, 1.5, 2.0, 3.0, 4.0, and 5.0 cm in the x-direction ($y = 0$ cm).

2.2.3. Time-dependent luminescence properties—The time-dependent luminescence properties of the Eu-activated phosphors were assessed using the probe type B ISDs and experimental procedures described in detail in (Kertzscher and Beddar 2016). Each ISD was positioned 1 cm from a BT source catheter in a rigid polymer-based phantom. The catheter was positioned between two 5-cm-thick Pb bricks which were positioned 2 cm from the final BT source position. The Pb bricks shielded the scintillation detector volume from the BT source while the source was moved toward and away from its final position facilitating measurement of the non-constant scintillation and afterglow, respectively, in absence of significant stray radiation. The CCD camera was used to monitor the ISD and, simultaneously, 3 additional scintillation detectors that were positioned on each side of the Pb bricks and at the exit point of the afterloader. The additional detectors served as triggers to indicate when the BT source was on the side of the Pb barrier opposite the ISD, at which point the BT source no longer induced significant stray radiation (Kertzscher and Beddar 2016), and when the source was located near the exit point of the afterloader.

The non-constant scintillation and afterglow were measured using the CCD camera with a 0.1-s exposure time. This experimental setup facilitated determining, with satisfactory temporal resolution, when the BT source was on the opposite side of the Pb barrier. Time-resolved emission spectra were acquired using the spectrometer with subsecond exposure time instead of the CCD camera to determine whether the time-dependent luminescence properties were more prominent in certain wavelength regions than in others. If only certain wavelength regions exhibited time dependence, the time dependence could be suppressed by omitting those wavelength regions with an optical filter.

The multiple-detector trigger system described above could not be used for the spectrometer measurements because our spectrometer had only 1 optical input channel; therefore, only 1 detector per measurement was used. However, those measurements were acquired for longer periods, for which subsecond temporal resolution of the BT source movement was not needed. The non-constant scintillation was characterized by monitoring the ISDs during about 520 s of irradiation using a BT source with an air-kerma strength of $40.0 \text{ mGy m}^2 \text{ h}^{-1}$ and with a 1-cm separation between the source and scintillation detector volume, yielding an absorbed dose of about 65 Gy. The measurement of the afterglow started about 10 s before the BT source was retracted and continued for about 500 s thereafter. The whole procedure was repeated once to determine whether the approximately 65-Gy irradiation of the Eu-activated phosphors would remove the time dependence of the scintillation, as it did for carbon-doped Al_2O_3 crystals exposed to at least 20 Gy (Andersen *et al* 2011). One ISD of

each Eu-activated phosphor was irradiated five times ($N=5$) with a 10-minute pause in between to determine if the scintillation intensity changed for the different irradiations. The detectors were irradiated 3 cm away from the BT source to reduce the positional uncertainty to less than 1% (0.7%, 2 standard deviations). The duration of each irradiation was 945 seconds which yielded 10 Gy using a BT source with an air-kerma strength of $30.7 \text{ mGy m}^2 \text{ h}^{-1}$. Using the same source, two different detectors of each Eu-activated phosphor were irradiated with 20 Gy at 0.5 cm away from the source (60 seconds) or with 10 Gy at 2 cm (425 seconds). The irradiations yielded dose rates 353, 24 and 11 mGy/s at 0.5, 2 and 3 cm away from the source, respectively. The scintillation intensities were normalized to the maximum intensity within the first 50 seconds of irradiations and within the first 10 Gy, to investigate the change in scintillation intensity over time and absorbed dose, respectively. The Eu-activated phosphors had not been irradiated for 3 months before the initial procedures.

2.3.4. Dose rate response of ISDs—The ISDs were exposed to a wide range of dose rates that are applicable to *in vivo* dosimetry during HDR brachytherapy treatments. During these experiments, the brachytherapy source moved in a catheter placed perpendicularly to the dosimeter probe. The source dwelled at 15 positions that were between 0.8 and 6.1 cm away from the position of the scintillator volume, which, using a source with an air-kerma strength of $30.7 \text{ mGy m}^2 \text{ h}^{-1}$, generated dose rates between 145 and 2 mGy/s. The source catheter and the dosimeter probe were positioned in a solid water phantom such that the scintillator volume was surrounded by solid water in all directions by at least 15 cm.

The ISD response was corrected for the energy dependence of the Eu-activated phosphors using the following procedure: a) The detector response was calibrated to reference calculations based on the TG-43 protocol (Rivard *et al* 2004) using separate measurements where the detector was placed 2 cm away from the brachytherapy source. The calibration coefficient converted the ISD response to uncorrected dose rates. b) The ratio between the uncorrected measured dose rate and the reference TG-43 dose rate was plotted against the total distance from the brachytherapy source for several source dwell positions. A first-order polynomial was fitted to this data. c) For the experiments, where the source moved inside the catheter, the parameters of the first-order polynomial and the total distance between the scintillator volume to the brachytherapy source were used to generate a correction factor for each dwell position. This correction factor suppressed the energy dependence of the ISDs.

3. Results

3.1. Emission spectra of detector probe types

The emission spectra of the scintillation signals and stem signals for the BCF-12- and BCF-60-based PSDs and the Eu-activated phosphor-based ISDs are shown in figure 2. As indicated in figure 2a, the emission spectra of the Eu-activated phosphors overlapped less with the stem signal than did the spectra of the BCF-12 and BCF-60 scintillators.

3.2 Signal intensities of scintillation detectors

The signal intensities of all scintillators are shown in table 3. The signal of the $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}$ -based ISDs was 2 to 3 times stronger than the signals of the $\text{Y}_2\text{O}_3:\text{Eu}$ - and $\text{Y}_2\text{O}_2\text{S}:\text{Eu}$ -based ISDs and about 8 times stronger than those of the $\text{YVO}_4:\text{Eu}$ -based ISDs. Even though the emission spectra of the 4 Eu-activated phosphors were similar to one another, the relative signal intensities depended on the attenuation length because the emission near 610 nm overlapped a steep gradient of the PMMA attenuation. As a consequence, the signal intensities of the $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}$ - and $\text{Y}_2\text{O}_2\text{S}:\text{Eu}$ -based ISDs benefitted more from shortening the fiber-optic cable than did those of the $\text{Y}_2\text{O}_3:\text{Eu}$ - and $\text{YVO}_4:\text{Eu}$ -based ISDs.

The signal intensity of the $\text{Y}_2\text{O}_3:\text{Eu}$ -based ISD was 32 times greater than that of the BCF-12-based PSD for 15-m PMMA attenuation. The intensity increased to 44 times greater than that of the BCF-12-based ISD when the attenuation length decreased to 0 m because the wavelengths near the $\text{Y}_2\text{O}_3:\text{Eu}$ emission peak were penalized by attenuation in PMMA more than were the wavelengths near the BCF-12 emission peak. The signal intensity of the $\text{Y}_2\text{O}_3:\text{Eu}$ -based ISD was 63 times greater than that of the BCF-60-based PSD for 15-m PMMA attenuation and 92 times greater for 0-m attenuation.

The attenuation spectra of the PMMA fiber-optic cable and the measured emission spectra of the scintillators were used to calculate the signal increase for shortened fiber-optic cable from 15 m to 0 m (see Table 3). The signal of the Eu-activated phosphors at wavelengths of 400 to 710 nm increased by factors between 2.2 and 3.0 and by factors between 2.4 and 4.1 when a 20-nm-wavelength band-pass region centered around the emission peak was considered.

3.3. Scintillation and background signal components

The relative intensities of the scintillation, photoluminescence, and stem signal components were determined within 20-nm-wide wavelength regions for the $\text{Y}_2\text{O}_3:\text{Eu}$ -, $\text{YVO}_4:\text{Eu}$ -, $\text{Y}_2\text{O}_2\text{S}:\text{Eu}$ -, and $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}$ -based probe type C ISDs (600–620 nm, 605–625 nm, 612–632 nm, and 611–631 nm, respectively) and for the BCF-12- and BCF-60-based probe type A PSDs (410–430 nm and 515–535 nm, respectively). The photoluminescence signal was negligible for the BCF-12- and BCF-60-based scintillators. The relative intensity of the signal components did not change considerably within the 20-nm-wavelength regions at fiber-optic cable lengths ranging from 0 to 15 m. The results presented in this section assumed no attenuation in the fiber-optic cable.

Table 4 shows the portions of the signal components out of the total signal for certain BT source positions with respect to the detector axis. The proportion of the stem signal in the total signal increased as the separation between the BT source and detector longitudinal axis decreased. When the BT source dwelled 0.5 cm from the detector axis and alongside it up to ± 8.0 cm from the scintillation detector volume, the stem signal accounted for up to 0.4% – 4.9%, depending on the Eu-activated phosphor. In contrast, the stem signal component of the PSDs was up to 82% of the total signal because of the strong overlap of the emission spectra of the stem signal with those of BCF-12 and BCF-60 and because of the relatively weak scintillation intensities of BCF-12 and BCF-60.

The photoluminescence signal was suppressed by sandwiching the optic filter between the Eu-activated phosphor and the fiber-optic cable. For BT source dwell positions 0.5 cm from the detector axis and up to ± 8.0 cm from the scintillation detector volume, the photoluminescence signal accounted for up to 0.2% – 2.2% (see Table 4) for the probe type B ISDs (the ISDs that did not incorporate an optic filter). The photoluminescence signal was reduced to 0.1% – 0.6% when the optic filter was placed between the Eu-activated phosphor and the fiber-optic cable (i.e., for the probe type C ISDs).

Figure 3 shows the composition of the total signal for the BCF-12- and BCF-60-based PSDs and the $\text{Y}_2\text{O}_3\text{:Eu}$ -based probe type C ISD for BT source dwell positions 1 cm from the detector axis. Figures 3a and b shows the signal components stacked onto each other for a 5-cm longitudinal distance between the BT source and the scintillation detector volume. The vertical dashed lines indicate the 20-nm-wide wavelength regions of interest. The vertical lines labeled with numbers 2 and 3 in figure 3b indicate 2 wavelength regions (590–630 nm and 580–710 nm) that we used in the analysis to compare with the 20 nm wavelength region (600–620 nm). Figures 3c and d show the relative intensities of the signal components for up to ± 8.0 cm longitudinal BT source distances from the scintillation detector volume. The stem signal of the PSDs (figure 3c) equaled the scintillation signal at about $z = 5.5$ cm and exceeded the scintillation signal beyond that point. The results highlight that the chromatic stem-suppression technique by Fontbonne *et al* (2002) is crucial for BCF-12- and BCF-60-based detectors. In contrast, using a 20-nm band-pass filter suppressed the combined background effect of the stem and photoluminescence signals to values less than 1% of the total signal for the $\text{Y}_2\text{O}_3\text{:Eu}$ -based probe type C ISD (figure 3d). The total background signal (stem + photoluminescence) increased to 1.3% and 2.8% of the total signal for the wider wavelength regions, underlining that an approximately 20-nm-wide bandpass filter centered at the emission peak should be incorporated into ISDs based on Eu-activated phosphors.

3.4. Time-dependent luminescence properties

All the Eu-activated phosphors exhibited time-dependent luminescence properties during and after ^{192}Ir BT source exposure. Figure 4 shows the non-constant scintillation in the wavelength region between 400 and 710 nm for all phosphors during the 2 consecutive approximately 65-Gy irradiations (a–d) and during the five consecutive 10 Gy-irradiations (e–h). The scintillation intensities of the $\text{Y}_2\text{O}_3\text{:Eu}$ - and $\text{YVO}_4\text{:Eu}$ -based ISDs reached near-maximum values within 1 s of irradiation and that it took more than 5 s for the $\text{Y}_2\text{O}_2\text{S:Eu}$ - and $\text{Gd}_2\text{O}_2\text{S:Eu}$ -based ISDs to reach 98% of their maximum response. As shown in table 5, within the first 10 s of irradiation, the responses of the $\text{Y}_2\text{O}_2\text{S:Eu}$ - and $\text{Gd}_2\text{O}_2\text{S:Eu}$ -based ISDs changed by 4% to 6% from the baseline response measured 1 s after the start of the irradiation. During the same period, the responses of the $\text{Y}_2\text{O}_3\text{:Eu}$ - and $\text{YVO}_4\text{:Eu}$ -based ISDs changed by no more than 0.6%.

The ISD responses to repeated irradiation (figure 4a–d) showed that the strong non-constant scintillation for the $\text{Y}_2\text{O}_2\text{S:Eu}$ and $\text{Gd}_2\text{O}_2\text{S:Eu}$ phosphors persisted even after an only 500-s pause after the initial irradiation, and that approximately 65-Gy irradiation did not remove the time dependence of the scintillation for the phosphors. The non-constant scintillation

was weaker for the $\text{Y}_2\text{O}_3\text{:Eu}$ and $\text{YVO}_4\text{:Eu}$ phosphors than for the $\text{Y}_2\text{O}_2\text{S:Eu}$ and $\text{Gd}_2\text{O}_2\text{S:Eu}$ phosphors. The time dependence of the scintillation persisted for $\text{YVO}_4\text{:Eu}$ and was reduced for $\text{Y}_2\text{O}_3\text{:Eu}$. The signal intensities of the $\text{Y}_2\text{O}_3\text{:Eu}$ - and $\text{YVO}_4\text{:Eu}$ -based ISDs decreased by 0.1% to 1.0% and increased by 0.9% to 1.3%, respectively, for the first 6-Gy irradiation and decreased by 1.0% to 3.0% and increased by 1.5% to 1.7%, respectively, for the first 24-Gy irradiation. The corresponding response changes for the $\text{Y}_2\text{O}_2\text{S:Eu}$ - and $\text{Gd}_2\text{O}_2\text{S:Eu}$ -based ISDs ranged from 5% to 7%. The 6-Gy and 24-Gy doses correspond to the doses the detectors could be expected to absorb if placed in the rectum or urethra during HDR BT (Therriault-Proulx *et al* 2011b).

Afterglow was present for at least 100 s for all Eu-activated phosphors as shown in the log-linear plots in figure 5. However, the intensity of the afterglow decreased to no more than 1% of the total signal within 1 s after the source had retracted for the $\text{Y}_2\text{O}_3\text{:Eu}$ - and $\text{YVO}_4\text{:Eu}$ -based ISDs (figure 5a inset) and did not, therefore, generate significantly inaccurate dose measurements. The afterglow of the $\text{Y}_2\text{O}_2\text{S:Eu}$ - and $\text{Gd}_2\text{O}_2\text{S:Eu}$ -based ISDs exhibited higher intensities (figure 5a) and shorter half-lives (b-e) than did that of the other Eu-activated phosphors. These properties of the $\text{Y}_2\text{O}_2\text{S:Eu}$ and $\text{Gd}_2\text{O}_2\text{S:Eu}$ afterglow generated spurious signals that made up about 2% of the total signal 2 s after the BT source had retracted from its dwell position. The afterglow of the $\text{Y}_2\text{O}_2\text{S:Eu}$ and $\text{Gd}_2\text{O}_2\text{S:Eu}$ phosphors would therefore generate significant measurement inaccuracy during BT because it would be superimposed on the scintillation signals, causing overestimation of the dose each time the source was moved to a new dwell position farther from the ISD.

The repeated irradiations (labeled “2nd irradi.” in figure 4a-d and “Irrad. #2”, “#3”, “#4”, “#5” in figure 4e-h) were normalized to the maximum scintillation intensity of the first irradiation (labeled “1st irradi.”). The initial seconds of the second 65 Gy-irradiation (figure 4a-d) differed from the last seconds of the first irradiation by + 3% and + 2.4% for the $\text{Y}_2\text{O}_3\text{:Eu}$ - and $\text{YVO}_4\text{:Eu}$ -based ISDs, respectively. The responses of the $\text{Y}_2\text{O}_2\text{S:Eu}$ - and $\text{Gd}_2\text{O}_2\text{S:Eu}$ -based ISDs shifted toward lower values. Although the measured positional uncertainty of the 65 Gy-irradiations was 1.2% (2 standard deviations), indicated by the blue-shaded regions in figure 4 (a-d), the changes in scintillation intensity may have been caused by lateral shifts of the BT source inside the needle, which, for a 1-cm distance between the scintillation detector volume and the BT source, can cause a $\pm 4\%$ variability in the detector response, as indicated by the yellow-shaded regions in figure 4 (a-d). The positional uncertainty and the variability were reduced to 0.7% and 1.4%, respectively, for the irradiations 3 cm away from the BT source (figure 4e-h). The ISDs exhibited changes in the scintillation intensities that were consistent with those observed for the 65 Gy-irradiations, i.e. that the overall intensity increased for the $\text{YVO}_4\text{:Eu}$ -based ISD and decreased for the others. Furthermore, the intensity-increase exceeded the positional uncertainties. Therefore, both the 65 Gy- and 10 Gy-irradiations indicate that the population of trapped charges change during the time interval between irradiations.

The time-resolved emission spectra of the non-constant scintillation and afterglow were acquired to determine the wavelength dependence of the time-dependent luminescence properties. Figure 6 shows the emission spectra for the $\text{Gd}_2\text{O}_2\text{S:Eu}$ -based ISD as an example. The non-constant scintillation and the afterglow were calculated for five 10-nm-

wide wavelength regions centered at 505, 536, 589, 620, and 702 nm. The scintillation for shorter wavelengths reached the maximum value more rapidly than did that of the longer wavelengths (figure 6b). The half-lives of the shorter wavelengths were shorter than those of the longer wavelengths (figure 6c). The time dependence of the most intense emission line, the one centered at 620 nm, was pronounced and therefore similar to those shown in figures 4 and 5. The time dependence for the $\text{Y}_2\text{O}_3\text{:Eu}$ and $\text{Gd}_2\text{O}_3\text{:Eu}$ phosphors was most pronounced for the longer wavelengths, whereas the opposite was true for $\text{Y}_2\text{O}_3\text{:Eu}$ and $\text{YVO}_4\text{:Eu}$. The most intense emission lines for all of the phosphors exhibited time-dependence, so the time dependence could not be suppressed significantly by omitting the other wavelength regions with an optic filter.

3.5. ISDs based on mixture of $\text{Y}_2\text{O}_3\text{:Eu}$ and $\text{YVO}_4\text{:Eu}$ phosphors

Although correction factors can be used to reduce the time dependence of the Eu-activated phosphors, such factors introduce uncertainties and add complexity to the readout protocol. To avoid the need for complex algorithms, the $\text{Y}_2\text{O}_3\text{:Eu}$ and $\text{YVO}_4\text{:Eu}$ phosphors could be mixed in an ideal proportion that balances the negative rate of change in the non-constant scintillation of $\text{Y}_2\text{O}_3\text{:Eu}$ with the positive rate of change of $\text{YVO}_4\text{:Eu}$.

Figure 7 shows the non-constant scintillation for wavelengths of 600 to 625 nm that was measured for the $\text{Y}_2\text{O}_3\text{:Eu}$ based ISD (a) and the $\text{YVO}_4\text{:Eu}$ based ISD (c), and calculated as the linear combination of both phosphors (b). The linear combination corresponds to 1 part $\text{Y}_2\text{O}_3\text{:Eu}$ and 10 parts $\text{YVO}_4\text{:Eu}$. This linear combination of $\text{Y}_2\text{O}_3\text{:Eu}$ and $\text{YVO}_4\text{:Eu}$ reduced the non-constant scintillation to less than 1% for approximately 65-Gy irradiation, to be compared with 5% for the $\text{Y}_2\text{O}_3\text{:Eu}$ -based ISD, and 2% for the $\text{YVO}_4\text{:Eu}$ -based ISD.

The $\text{Y}_2\text{O}_3\text{:Eu}$ and $\text{YVO}_4\text{:Eu}$ phosphors were mixed in 1 part $\text{Y}_2\text{O}_3\text{:Eu}$ and 4, 5, 6, and 10 parts $\text{YVO}_4\text{:Eu}$ and used in ISDs (2–3 detectors for each ratio). The ISDs were exposed to approximately 65 Gy to examine which mass-ratio of the phosphors generated the smallest amount of time-dependence. The detector with 1 part $\text{Y}_2\text{O}_3\text{:Eu}$ and 4 parts $\text{YVO}_4\text{:Eu}$ exhibited the smallest amount of time-dependence (data not shown).

Six ISDs with detector volumes based on 1 part $\text{Y}_2\text{O}_3\text{:Eu}$ and 4 parts $\text{YVO}_4\text{:Eu}$ were manufactured. Five of the 6 ISDs (type B) incorporated detector volumes based on 1 part glue and 1.3, 2.0, 4.4, 5.0, or 6.4 parts phosphor. The 6th ISD (type C) incorporated 1 part glue and 6.4 parts phosphor. Figure 8 shows the characteristics of the 6 ISDs. The comparison of the measured emission spectrum (figure 8a, red solid line) and the calculated spectrum (black dashed line) shows that the measured scintillation intensity near 600 nm is smaller than the calculation, which suggests that the proportion of the $\text{YVO}_4\text{:Eu}$ phosphor was higher than that measured with the weight scale. The emission spectra of the 5 type B ISDs with different glue-phosphor ratios (figure 8b) shows that the ratios were similar for all the detectors, which suggests that the ratio between the $\text{Y}_2\text{O}_3\text{:Eu}$ and $\text{YVO}_4\text{:Eu}$ was homogenous in the mixture. The scintillation intensities of the 5 ISDs (figure 8c) shows that the highest intensity was achieved when there was 4.4 parts phosphor and 1 part glue, and that the intensity increased for higher proportions of phosphor. The components of the total signal was obtained according to the method described in section 2.4.2, incorporated with the emission spectrum of the type C ISD and the measured scintillation intensity of the ISD

with 4.4 parts phosphor and 1 part glue multiplied by the fraction of the type C and type B ISDs with 6.4 parts phosphor. Assuming that the photoluminescence is negligible, the measurements (figure 8d) demonstrated that the stem signal background was up to 1.6% of the total signal for BT source dwell positions 1.0 cm from the detector axis and up to ± 8.0 cm from the scintillation detector volume. The time dependent luminescence properties of the ISDs were measured for two 65-Gy irradiations, where the second irradiation took place after a 500-s pause after the initial irradiation (figure 8e). The scintillation intensity-increase for all 5 detectors was during the first irradiation between 0.2% to 0.9% after 24 Gy and 1.1% to 2.3% after 50 Gy. The scintillation intensity-increase during the second irradiation was between 0.9% to 1.6% after 24 Gy and 1.2% to 2.7% after 50 Gy. The intensity-increase was observed for the detector with 4.4 parts phosphor and 1 part glue, when it was irradiated five times with 10 Gy (figure 8f). The increase was greater than the 0.7% positional uncertainty (2 standard deviations) which indicates that the population of trapped charges change during the time interval between irradiations.

3.6 Dose rate response of the ISDs

Figure 9 shows 3 consecutive dose rate measurements, using the probe type C ISD based on the mixture between $\text{Y}_2\text{O}_3\text{:Eu}$ and $\text{YVO}_4\text{:Eu}$, for BT source positions between 0.8 and 6.1 cm. The dose rates not corrected for energy dependence (Figure 9a,b) differed substantially from the TG-43 reference calculations. However, the measurements were accurate to within the percent level from the reference calculations (Figure 9c,d) when the energy correction function (e) had been applied. The uncertainty of the dose rate calculation (blue-shaded regions in Figure 9) were due to the uncertainty of the BT source and scintillator volume positions, and were calculated as described in (Andersen *et al* 2009, Kertzscher *et al* 2011b). Similar results were obtained for the other Eu-activated phosphors.

Figure 10 shows the scintillation intensities of the ISDs during constant irradiation at the dose rates 353, 24 and 11 mGy/s. Figures 10a-e show the scintillation intensities normalized to the maximum intensity within the first 50 seconds. The results show that the time derivative of the scintillation intensity depends on the dose rate for the $\text{Y}_2\text{O}_2\text{S:Eu}$ and $\text{Gd}_2\text{O}_2\text{S:Eu}$ phosphors, in contrast to the $\text{Y}_2\text{O}_3\text{:Eu}$ and $\text{YVO}_4\text{:Eu}$ phosphors, and the mixture between those two phosphors for which the time-derivatives are similar. Figures 10f-j show the scintillation intensities normalized to the maximum intensity within the first 10 Gy. The time-dependence does not differ substantially for different absorbed dose rates for the $\text{Y}_2\text{O}_3\text{:Eu}$ and $\text{YVO}_4\text{:Eu}$ phosphors and the mixture. In contrast, the time-dependence is less pronounced for smaller dose rates for the $\text{Y}_2\text{O}_2\text{S:Eu}$ and $\text{Gd}_2\text{O}_2\text{S:Eu}$ phosphors.

4. Discussion

We investigated the luminescence properties of the Eu-activated phosphors $\text{Y}_2\text{O}_3\text{:Eu}$, $\text{YVO}_4\text{:Eu}$, $\text{Y}_2\text{O}_2\text{S:Eu}$, and $\text{Gd}_2\text{O}_2\text{S:Eu}$ to determine their suitability for use in real-time verification of HDR BT. Our results showed that the $\text{Y}_2\text{O}_3\text{:Eu}$ and $\text{YVO}_4\text{:Eu}$ phosphors are promising materials for the manufacturing of ISDs that accurately and precisely monitor the absorbed dose in real time during ^{192}Ir HDR BT, as long as corrections for the energy dependence are applied during measurements. Optimal performance was achieved by mixing

the $\text{Y}_2\text{O}_3\text{:Eu}$ and $\text{YVO}_4\text{:Eu}$ phosphors to reduce the time-dependent luminescence properties to 1% for HDR BT relevant doses and by optimizing the amount of phosphor powder in the scintillator detector volume to reduce the stem signal contamination and minimize the statistical uncertainty of time-resolved dosimetry. We found that the $\text{Y}_2\text{O}_2\text{S:Eu}$ and $\text{Gd}_2\text{O}_2\text{S:Eu}$ phosphors were not suitable for real-time verification during HDR BT because they exhibited time-dependent luminescence properties that introduced significant inaccuracies in absorbed dose measurements.

The scintillation intensities of the ISDs were 1 to 2 orders of magnitude greater than that of the BCF-12-based PSDs. By using the specific Eu-activated phosphor powders applied in this study, the scintillation intensity could be increased by up to a factor of 3 because one third of the scintillator detector volume in each ISD consisted of the phosphor. However, more densely packed phosphor powder could limit the effective scintillation detector volume to the layer closest to the fiber-optic cable because the topography of small phosphor crystals can cause poor light transmission (Abrams and Holloway 2004). We were able to increase the scintillation intensity by a factor of 1.6 by increasing the phosphor-glue mixture mass ratio from 2 to 1 to 4.4 to 1.

The signal intensities of the Eu-activated phosphors increased by a factor of 2 to 4 when the PMMA fiber-optic cable was shortened from 15 m to 0 m. The signal intensity could potentially be further increased if the ISDs incorporated phosphors with parameters optimized for maximal light yield, as the light yield depends on both the Eu ion concentration (Yen *et al* 2007) and the particle size (Chakradhar *et al* 2011, Nakamura *et al* 2014). Studies have shown that Eu-activated phosphor crystals with 10- to 100-nm particle sizes exhibit larger scintillation yields than do micrometer-sized crystals (Wakefield *et al* 2001, Osseni *et al* 2011). Furthermore, Y_2O_3 crystals activated with Eu and Li emit stronger scintillation (Shin *et al* 2005, Stanton *et al* 2014, Bae *et al* 2007) and photoluminescence (Sun *et al* 2001) than do crystals activated with Eu ions alone. In a study using external-beam radiation sources, Belley *et al* (2015b) employed 11- μm -thick pellets of about 50-nm-sized $\text{Y}_2\text{O}_3\text{:Eu,Li}$ crystals (Stanton *et al* 2014) coupled with a fiber-optic cable. The importance to compare the light yield and other luminescence properties of nanometer-sized crystals and crystals co-doped with Eu and Li with the properties of commercial Eu-activated phosphors, has been stressed by Kawahara *et al* (2006). Kawahara *et al* (2006) developed nanometer-sized $\text{Y}_2\text{O}_2\text{S}$ scintillators but found generating larger light yields than those exhibited by commercial micrometer-sized counterparts challenging. They pointed out that market competition has, over decades, led to the refinement of the production processes for commercial phosphors so as to achieve optimal luminescence properties.

We found that the signal intensities of the $\text{Y}_2\text{O}_3\text{:Eu}$ - and $\text{Y}_2\text{O}_2\text{S:Eu}$ -based ISDs were about 3 times greater than that of the $\text{YVO}_4\text{:Eu}$ -based ISD and about 0.4 times that of the $\text{Gd}_2\text{O}_2\text{S:Eu}$ -based ISD. These findings are similar to those of previous studies by Molina *et al* (2012) and Nakamura *et al* (2014). Molina and colleagues investigated ISDs made with $\text{Y}_2\text{O}_3\text{:Eu}$, $\text{YVO}_4\text{:Eu}$, and $\text{Y}_2\text{O}_2\text{S:Eu}$ phosphors from the same manufacturer that produced the phosphors used in this study (Phosphor Technology Ltd.). The authors reported that the scintillation intensity of $\text{YVO}_4\text{:Eu}$ was about 1.8 times greater than that of $\text{Y}_2\text{O}_3\text{:Eu}$ and about 1.6 times greater than that of $\text{Y}_2\text{O}_2\text{S:Eu}$ under beta-irradiation from a ^{90}Sr source. The

differences between our results and those reported by Molina et al may arise from the dependency of the light yield on the radiation sources. Nakamura and colleagues used phosphors from a different manufacturer (Mitsubishi Chemical) and found that the scintillation intensity of $\text{Y}_2\text{O}_3\text{:Eu}$ was about 4 times greater than that of $\text{YVO}_4\text{:Eu}$ under exposure to 60–120 kVp x-rays, which is consistent with the results presented in this study.

The contribution of the stem signal background to the total signal was at the percent-level for the $\text{Y}_2\text{O}_3\text{:Eu}$ - and $\text{Y}_2\text{O}_2\text{S:Eu}$ -based ISDs and about 5% and less than 1% for the $\text{YVO}_4\text{:Eu}$ - and $\text{Gd}_2\text{O}_2\text{S:Eu}$ -based ISDs, respectively, when the BT source was located 0.5 cm from the fiber-optic cable and 8 cm from the scintillation detector volume, assuming no attenuation in the PMMA fiber-optic cable. Considering those relative positions of the BT source and the ISDs is necessary because they could occur during *in vivo* dosimetry (e.g., if the ISD were inserted in the urethra with the scintillation detector volume placed near the bladder wall and the BT source positioned near the urethra). Measures used to avoid large stem signals and ensure accurate dose measurements during *in vivo* BT dosimetry include maintaining a high signal intensity, for example, by shortening the fiber-optic cable to about 2 m, and, as mentioned in section 3.3, by using an approximately 20-nm optic band-pass filter to reduce the acceptance of the stem signal, as the stem signal has a wider wavelength spectrum than do the Eu-activated phosphors and as the Eu-activated phosphors exhibit narrow emission peaks.

The photoluminescence signal of the $\text{YVO}_4\text{:Eu}$ -based ISD was suppressed from 2.2% to 0.6% of the total signal when an optic filter was placed between the scintillator and the fiber-optic cable and from 0.3%, 0.5%, and 0.2% to 0.2%, 0.2%, and 0.1% for the $\text{Y}_2\text{O}_3\text{:Eu}$ -, $\text{Y}_2\text{O}_2\text{S:Eu}$ -, and $\text{Gd}_2\text{O}_2\text{S:Eu}$ -based ISDs, respectively. With the exception of that for $\text{YVO}_4\text{:Eu}$, these results misleadingly suggest that the optic filter that suppresses the photoluminescence is redundant. Around the emission peak (610–620 nm), the measured photoluminescence signal intensity per absorbed dose in the fiber-optic cable was about half of the intensity of the stem signal for the $\text{YVO}_4\text{:Eu}$ -, $\text{Y}_2\text{O}_2\text{S:Eu}$ -, and $\text{Gd}_2\text{O}_2\text{S:Eu}$ -based ISDs. However, the ultraviolet portion of the stem signal, which generates photoluminescence in the phosphors, traversed in the experimental setup about 40 cm through the PMMA fiber-optic cable before reaching the phosphor. Because PMMA strongly attenuates ultraviolet light, a large part of the portion of the stem signal that generates photoluminescence was lost during the experiments. A smaller part of the ultraviolet light is attenuated when the BT source dwells near the fiber-optic cable, for instance, 8 cm from the scintillation detector volume. Therefore, the experimentally obtained photoluminescence intensity was likely an underestimation. As a consequence, the photoluminescence intensity of the $\text{YVO}_4\text{:Eu}$ -, $\text{Y}_2\text{O}_2\text{S:Eu}$ -, and $\text{Gd}_2\text{O}_2\text{S:Eu}$ -based ISDs would likely exceed a factor 0.5 of the stem signal during *in vivo* dosimetry without an optic filter between the scintillation detector volume and the fiber-optic cable. Thus, our recommendation is to incorporate an optic filter into ISDs that use $\text{Y}_2\text{O}_3\text{:Eu}$, $\text{YVO}_4\text{:Eu}$, $\text{Y}_2\text{O}_2\text{S:Eu}$, and $\text{Gd}_2\text{O}_2\text{S:Eu}$ scintillators.

Although all Eu-activated phosphors exhibited time-dependent luminescence properties, the time dependencies of $\text{Y}_2\text{O}_3\text{:Eu}$ and $\text{YVO}_4\text{:Eu}$ were more easily compensated for than were those of $\text{Y}_2\text{O}_2\text{S:Eu}$ and $\text{Gd}_2\text{O}_2\text{S:Eu}$. The time dependence of the $\text{Y}_2\text{O}_2\text{S:Eu}$ - and

Gd₂O₂S:Eu-based ISDs generated a non-constant response to irradiation, which resulted in a large (greater than 4%) scintillation signal increase during the first 10 s of irradiation and a strong afterglow. It was not possible to suppress the time dependence to values less than 1% by optically filtering out the wavelength regions outside the most intense emission lines because those emission lines exhibited strong time dependence. In contrast, Molina *et al* (2012) found no significant time dependence for Y₂O₃:Eu, YVO₄:Eu, or Y₂O₂S:Eu phosphors that were exposed to 1.4 Gy from a ⁹⁰Sr source during a 3600 s time period, or for Y₂O₃:Eu exposed to about 0.35 Gy (about 60 s) from a ⁶⁰Co source. The scintillation signal of Y₂O₃:Eu, YVO₄:Eu, and Y₂O₂S:Eu in our experiments changed by less than 0.5%, up to 0.6%, and 4%, respectively, with exposure to a ¹⁹²Ir source at 1.2 Gy, which corresponded to 10 s. The study by Molina *et al* and our results showed that the time-dependent luminescence properties of Y₂O₃:Eu and YVO₄:Eu result in non-constant scintillation less than 1% during 1.4 Gy irradiation. When exposed to doses that exceed 6 Gy, however, we found that Y₂O₃:Eu and YVO₄:Eu can exhibit non-constant scintillation greater than 1%. We speculate that our different findings regarding the time-dependent luminescence properties of Y₂O₂S:Eu may have 2 explanations. First, the non-constant scintillation was less pronounced as the dose rate decreased from 353 to 11 mGy/s (figure 10f-j) which suggests that the non-constant scintillation may have been negligible for the 0.39 mGy/s dose rate used in the experiments by Molina *et al*. Second, Molina and colleagues may have used Y₂O₂S:Eu that did not exhibit non-constant scintillation. In contrast, Nakamura *et al* (2014) reported that the scintillation intensity of their micrometer-sized Y₂O₃:Eu and YVO₄:Eu crystals decreased by 10% and 6%, respectively, under exposure to 2 Gy 120-kVp x-rays at 2.1 mGy/s, which strongly differed from our results. These differences may be explained by the fact that Nakamura and colleagues (Nakamura *et al* 2014) used Eu-activated phosphors from a different manufacturer from those selected for our study and that, according to the authors, their Y₂O₃:Eu and YVO₄:Eu phosphors exhibited significant brightness deterioration caused by reversibly radiation-generated color centers that quenched the scintillation.

Repeated irradiations of 65 Gy or 10 Gy showed that the non-constant scintillation for the Eu-activated phosphors persisted even after a 500-s or 600-s pause after each irradiation. Consequently, the pre-irradiation protocol published by Andersen *et al* (2011) is not applicable for the phosphors. Thorough studies of the time-dependent luminescence properties of the Eu-activated phosphors could reveal that the non-constant scintillation and afterglow can be modeled based on, e.g., the accumulated dose, the dose rate and the time interval since the last irradiation. Such a model could adapt correction functions with non-linear dependence on irradiation time and/or accumulated dose to stabilize the dose-measurement readout similar to the algorithms that Andersen *et al* (2006) and Damkjær *et al* (2008) developed for Al₂O₃:C. As shown by Andersen *et al* (2011) using Al₂O₃:C crystals, repeatability can be investigated by using several approximately 10-Gy irradiations paused for randomly chosen periods ranging from 1 s to 24 h or longer under irradiation conditions with small dose gradients. If the time dependence can be reliably modeled, such that the unstable detector response can be accurately corrected, the Gd₂O₂S:Eu phosphor could be useful in ISDs because it generates scintillation intensities that are superior to those of the other Eu-activated phosphors.

However, models for correcting for the impact of the time-dependent luminescence properties could introduce uncertainties and added complexity in the readout protocol, which could make it more difficult to develop accurate BT detector systems that are straightforward to operate. Furthermore, the slow response of the $\text{Y}_2\text{O}_2\text{S}:\text{Eu}$ and $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}$ phosphors would complicate the calibration procedure because it is essential to ensure that the response has stabilized before acquiring the calibration coefficients.

Instead of correcting for time dependence, we pursued the use of mixtures of $\text{Y}_2\text{O}_3:\text{Eu}$ and $\text{YVO}_4:\text{Eu}$ phosphors, which, in contrast with $\text{Y}_2\text{O}_2\text{S}:\text{Eu}$ and $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}$, respond rapidly to absorbed dose and generate time dependence of no more than 0.6% within the first 10 s of irradiation. A disadvantage of the $\text{Y}_2\text{O}_3:\text{Eu}$ and $\text{YVO}_4:\text{Eu}$ phosphors, however, is that their scintillation intensity changes by -1% to -3% and by about 1.6%, respectively, while absorbing 24 Gy, the dose to which the scintillator could be exposed if placed in the urethra during HDR BT (Therriault-Proulx *et al* 2011b). To mitigate this effect, we mixed the phosphors in proportions that balanced their opposing time dependencies which reduced the non-constant scintillation to 1% for HDR BT relevant doses (see values after 24 Gy for 1st irradiations in Table 5 and Figure 8e). The 1%-level time-dependence of the mixture is negligible compared to other uncertainties inherent to BT dosimetry (e.g. large positional uncertainty for small source-to-detector distances or large statistical uncertainties for large distances (Kertzscher *et al* 2011b)), while the 3%-level uncertainty of $\text{Y}_2\text{O}_3:\text{Eu}$ is not. As seen in figure 8e (“2nd irradiation”), the scintillation intensity increases to up to 1.5% from the initial intensity within the first 20 Gy if the detector has been exposed to 65 Gy only 500 s prior to the measurement. As seen in figure 5a, it takes longer for the afterglow of $\text{Y}_2\text{O}_3:\text{Eu}$ to decay than does the afterglow of $\text{YVO}_4:\text{Eu}$. If more time passes after the previous irradiation, more of the afterglow of the $\text{Y}_2\text{O}_3:\text{Eu}$ will be emitted which would result in more electrons and holes (trapped in impurity centers or crystal defects) thermally activated into the conduction and valence bands, respectively. More empty traps may cause the $\text{Y}_2\text{O}_3:\text{Eu}$ crystals to regain the opposing time dependency with respect to $\text{YVO}_4:\text{Eu}$ and, therefore, also cause the mixture between the $\text{Y}_2\text{O}_3:\text{Eu}$ - and $\text{YVO}_4:\text{Eu}$ -phosphors to regain the less than 1% time dependency. The increasing scintillation intensity observed for the repeated 65 Gy- and 10 Gy-irradiations (figure 8e,f), which is likely caused by changing populations of trapped charges during the time interval between irradiations, is not fully suppressed when the $\text{Y}_2\text{O}_3:\text{Eu}$ - and $\text{YVO}_4:\text{Eu}$ -phosphors are mixed with a 1-to-4 ratio. However, further optimization of the ratio between the two phosphors may result in suppressed scintillation intensity-changes below 1%.

We found that the scintillation intensity of the ISDs reached a maximum if the 0.8 mm^3 scintillation detector volume consisted of 1 part glue and approximately 4 parts phosphor, and we observed that the scintillation intensity decreased if the phosphor was more densely packed. When developing ISDs based on Eu-activated phosphors, the scintillation intensity should be maximized by optimizing the phosphor-glue ratio to limit the influence of stem signal contamination and to minimize the statistical uncertainty of time-resolved measurements. In addition to shortening the fiber-optic cable to about 2 m, choosing a more sensitive photodetector than the one we used could further minimize the statistical uncertainty (Boivin *et al* 2015).

Before any phosphor-based ISD is used for *in vivo* HDR BT dosimetry, the energy, temperature, and time dependencies of the luminescence properties must be carefully characterized and corrections for these characteristics must be properly applied. Corrections for energy-dependence can be applied as suggested in this article and by (Reniers *et al* 2012, Pittet *et al* 2013), and corrections for temperature and time-dependence can be applied as suggested by Wootton and Beddar (2013), and the present study, respectively. Also, Martinez *et al* (2015) showed that the scintillation signal of YVO₄:Eu changes by 0.01%/°C under ⁶⁰Co irradiation. Although temperature dependence was not considered in our study, future research must characterize the non-constant scintillation and afterglow of the phosphors at body temperature because the thermal activation processes that govern time dependence can be different at body temperature during *in vivo* dosimetry regimen from the time dependence at room temperature during calibration.

5. Conclusion

In this study, we demonstrated that the Eu-activated phosphors Y₂O₃:Eu and YVO₄:Eu in powder form are promising materials for the manufacturing of inorganic scintillator detectors (ISDs) that accurately and precisely verify HDR BT doses in real time because they can be used to manufacture sub-mm detector volumes that generate large and stable signal intensities that are insignificantly contaminated by the stem signal. Of the Y₂O₃:Eu, YVO₄:Eu, Y₂O₂S:Eu, and Gd₂O₂S:Eu phosphors that we investigated in this study, high accuracy was most easily achieved by using a combination of the Y₂O₃:Eu and YVO₄:Eu phosphors as the scintillation detector volume, primarily because the combination suppressed the time-dependence of the detectors. We also demonstrated that high signal intensities can be achieved with an optimized packing density of the phosphor mixture in the scintillation detector volume, which limits stem signal contamination and minimizes the statistical uncertainty of time-resolved measurements. The signal intensity can be further increased by shortening the fiber-optic cable to about 2 m and by choosing a photodetector with high sensitivity in the wavelength region near the emission peak of the phosphors. The stem signal can be adequately suppressed by incorporating a band-pass filter that transmits wavelengths from 600 to 625 nm between the fiber-optic cable and the photodetector. Unwanted photoluminescence can be suppressed by placing a long-pass filter between the scintillation detector volume and the fiber-optic cable. We conclude that ISDs based on a mixture of Y₂O₃:Eu and YVO₄:Eu can be used to construct accurate and precise real-time verification technology for HDR BT that is cost effective and straightforward to manufacture and could improve the dissemination of HDR BT verification. Widespread dissemination of this technology could lead to an improved understanding of error types and frequencies during BT and to improved patient safety during treatment.

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Figure 1.

The 3 detector probe types used in the experiments: (A) a scintillator coupled with a fiber-optic cable, (B) a scintillator coated with reflective paint and coupled with a fiber-optic cable, and (C) an optic filter sandwiched between the scintillator and the fiber-optic cable.

Figure 2.

(a) the emission spectra of the scintillation signals for all scintillator materials used in this study and of the radiation-induced Cerenkov and fluorescence light in the PMMA fiber-optic cable. (b) the most prominent scintillation wavelength regions of the Eu-activated phosphors. The emission spectra were corrected for the quantum efficiency of the spectrometer and for the attenuation in the fiber-optic cable.

Figure 3.

Measured radioluminescence (RL), photoluminescence (PL), and stem signal (Cerenkov and fluorescence, S) components of the total signal, corrected by the attenuation in the PMMA fiber-optic cable and quantum efficiencies of the photodetectors. Values are shown for the BCF-12- and BCF-60-based PSDs (a,c) and the probe type C $Y_2O_3:Eu^{3+}$ -based ISD with a long-pass filter sandwiched between the scintillator and the fiber-optic cable (b,d). Figures a and b show the signal components stacked onto each other when the ^{192}Ir BT source was positioned 1 cm from the fiber-optic cable and 5 cm from the scintillation detector volume. Figures c and d show the portion of each signal component out of the total signal (RL + PL + S) when the BT source was positioned 1 cm from the detector axis and up to ± 8 cm from the detector volume. The signal portions were calculated within the wavelength regions

indicated by the vertical dashed lines in b: region 1 (600–620 nm), region 2 (590–630 nm), and region 3 (580–710 nm), respectively.

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Figure 4.

a-d: the temporally non-constant scintillation of the Y₂O₃:Eu-, YVO₄:Eu-, Y₂O₂S:Eu-, and Gd₂O₂S:Eu-based ISDs during 2 approximately 65-Gy irradiations with an ¹⁹²Ir BT source. The irradiations were separated by an approximately 500-s pause. The scintillation intensities from both irradiations were normalized to the maximum value acquired during the first irradiation for about the first 50 Gy. e-h: scintillation intensity measurements for the Eu-activated phosphors of five 10-Gy-irradiations.

Figure 5.

The afterglow of $\text{Y}_2\text{O}_3:\text{Eu}$ -, $\text{YVO}_4:\text{Eu}$ -, $\text{Y}_2\text{O}_2\text{S}:\text{Eu}$ - and $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}$ -based ISDs. a: all the Eu-activated phosphors exhibited afterglow for at least 100 s after the BT source was retracted from the dwell position (time = 0). b-e: the afterglow half-lives for each Eu-activated phosphor. The half-lives were acquired by fitting 1-term exponential functions ($y = Ae^{at}$) to the $\text{Y}_2\text{O}_3:\text{Eu}$ and $\text{YVO}_4:\text{Eu}$ afterglow data and 2-term exponential functions ($y = Ae^{at} + Be^{bt}$) to the $\text{Y}_2\text{O}_2\text{S}:\text{Eu}$ and $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}$ data.

Figure 6.

a: emission spectra for $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}$ acquired during and after irradiation with an ^{192}Ir BT source. The green dashed-dotted line represents the spectrum that was acquired 0.1 s before full scintillation (black solid line), and the red dashed lines represent spectra acquired 0.1, 1, and 10 s after the BT source was retracted from the dwell position. b,c: the time-resolved scintillation signals (b) and afterglow signals (c) integrated within the wavelength regions (indicated by numbers 1–5) shown in blue in a. The insets show the signals within the first 10 s of irradiation (b) and after source retraction (c).

Figure 7.

The measured non-constant scintillation from 600 to 625 nm for the $\text{Y}_2\text{O}_3:\text{Eu}$ -based ISD (a), $\text{YVO}_4:\text{Eu}$ -based ISD (c), and the calculated linear combination of the $\text{Y}_2\text{O}_3:\text{Eu}$ and $\text{YVO}_4:\text{Eu}$ phosphors (b).

Figure 8.

Characteristics of ISDs based on 1 mass-part $\text{Y}_2\text{O}_3:\text{Eu}$ and 4 mass-parts $\text{YVO}_4:\text{Eu}$. a: measured and calculated emission spectra of probe type B ISDs based on the Eu-phosphor mixture (1 mass-part $\text{Y}_2\text{O}_3:\text{Eu}$ and 4 mass-parts $\text{YVO}_4:\text{Eu}$), and measured emission spectra of individual Eu-phosphors $\text{Y}_2\text{O}_3:\text{Eu}$ and $\text{YVO}_4:\text{Eu}$. b: measured emission spectra of probe type B ISDs based on the Eu-phosphor mixture with different phosphor-glue proportions. c: scintillation intensities of probe type B (filled circles) and probe type C (filled square) ISDs containing different proportions of the Eu-phosphor mixture and the glue (for 0 m PMMA attenuation and emission wavelengths between 400 and 710 nm). d: signal components of the probe type C ISD based on the Eu-phosphor mixture (for source dwell positions 1.0 cm from the detector axis, for emission wavelengths between 600 and 625 nm, and based on the detector with 4.4 mass-parts phosphor). e: time-dependent luminescence properties of probe type B ISDs based on the Eu-phosphor mixture with different phosphor-glue proportions

(for a 1 cm source-to-detector distance and 96.4 mGy/s dose rate (32 mGy m²/h), and for emission wavelengths between 600 and 625 nm). f: scintillation intensity measurements for five 10 Gy-irradiations (for a 3 cm source-to-detector distance and 10.6 mGy/s dose rate (31 mGy m²/h), and for emission wavelengths between 600 and 625 nm).

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Figure 9.

Dose rate measurements using the probe type C ISD based on the mixture between $\text{Y}_2\text{O}_3:\text{Eu}$ and $\text{YVO}_4:\text{Eu}$. The BT source dwelled at 15 positions between 0.8 and 6.1 cm from the scintillator volume. The uncorrected dose rate measurements (a,b) were factored with the values retrieved using the linear energy correction function (e) to generate the energy corrected measurements (c,d).

Figure 10.

The scintillation intensities normalized to the maximum intensity within the first 50 seconds (a-e) or within the first 10 Gy (f-j).

Table 1

Physical characteristics of the scintillators.

Scintillator material	Emission peak ^a (nm)	Density ^a (g/cm ³)	Index of refraction ^b (n _d)	Electron density (electrons/cm ³)	Effective atomic number ^c (Z _{eff})	Form ^d	Volume ^e (mm ³)
BCF-12	435	1.05	1.60	3.4×10^{23}	5.7	Single piece	0.8
BCF-60	530	1.05	1.60	3.4×10^{23}	5.7	Single piece	0.8
Y ₂ O ₃ :Eu	611	5.0	1.54	1.4×10^{24}	36.1	Powder (8.0 μm)	0.8
YVO ₄ :Eu	619	5.3	1.54	1.5×10^{24}	31.1	Powder (6.5 μm)	0.8
Y ₂ O ₂ S:Eu	626	5.0	1.54	1.4×10^{24}	35.5	Powder (7.5 μm)	0.8
Gd ₂ O ₂ S:Eu	626	7.5	1.54	1.9×10^{24}	60.1	Powder (8.0 μm)	0.8

^aThe emission-peak wavelength and density values were provided by the manufacturers.^bIndices of refraction for BCF-12 and BCF-60 were provided by the manufacturers. The index of refraction values used for the Eu-activated phosphors corresponded to that of the glue with which the phosphor powders were mixed.^cEach effective atomic number (Z_{eff}) was calculated using equation 7.26 with $m = 3.5$, as published by Johns and Cunningham (1983). The Z_{eff} for water is 7.5.^dThe Eu-activated phosphors were acquired in powder form. Median particle sizes are shown in parentheses.^eThe volumes of the Y₂O₃:Eu, YVO₄:Eu, Y₂O₂S:Eu, and Gd₂O₂S:Eu crystals in the phosphor-glue mixture in the 0.8-mm³ cylinders were approximately 0.26, 0.25, 0.26, and 0.19 mm³, respectively, which is equivalent to 1.3–1.4 mg.

Table 2

Numbers of probes of each probe type that were manufactured for this study.

Scintillator material	Detector probe type		
	Type A ^a	Type B ^b	Type C ^c
BCF-12	3	0	0
BCF-60	3	0	0
Y ₂ O ₃ :Eu	6 ^d	3	3
YVO ₄ :Eu	3 ^d	1	1
Y ₂ O ₂ S:Eu	3 ^d	1	1
Gd ₂ O ₂ S:Eu	3 ^d	1	1
Mixture between Y ₂ O ₃ :Eu and YVO ₄ :Eu	9 ^d	5	1

^aThe scintillator volume was optically coupled with the fiber-optic cable.

^bReflective paint covered the scintillator volume which was optically coupled with the fiber-optic cable.

^cA long-pass optic filter was sandwiched between the scintillator volume (which was covered with reflective paint) and the fiber-optic cable.

^dThree type A probes that were 2 m long (and during experiments coupled to a 12-m-long fiber-optic cable for readout) were manufactured for the Y₂O₃:Eu, YVO₄:Eu, Y₂O₂S:Eu and Gd₂O₂S:Eu based ISDs. Nine type A probes that were 20 cm long were manufactured for the mixture between Y₂O₃:Eu and YVO₄:Eu. All other ISDs were 15 m long.

Table 3

The signal intensities of the ISDs based on Eu-activated phosphors and the PSDs based on organic scintillators. The intensities were normalized to that of the BCF-12-based PSDs. The uncertainties (1 standard deviation) correspond to the positional uncertainty of the detector volume and the BT source, the variation introduced when connecting and disconnecting the detector probe with the photodetector, and the statistical uncertainty.

Wavelength region and attenuation	Scintillator material							
	BCF-60	BCF-12	Y ₂ O ₃ :Eu	YVO ₄ :Eu	Y ₂ O ₂ S:Eu	Gd ₂ O ₂ S:Eu		
λ -region (nm)	400–710	400–710	400–710	400–710	400–710	400–710		
15 m PMMA ^a	0.51±0.04	1±0.07	32±2	9±1	29±2	72±5		
0 m PMMA ^b	0.48±0.03	1±0.07	44±3	16±1	54±4	134±9		
Signal increase ^c	1.5	1.6	2.2	2.7	3.0	3.0		
λ -region (nm)	515–535	410–430	600–620	605–625	612–632	611–631		
15 m PMMA ^a	0.59±0.04	1±0.07	74±5	20±1	53±4	134±9		
Part of spectrum ^d	26%	23%	52%	49%	41%	43%		
0 m PMMA ^b	0.45±0.03	1±0.07	102±7	35±2	123±9	307±21		
Part of spectrum ^d	24%	26%	59%	57%	58%	59%		
Signal increase ^c	1.3	1.8	2.4	3.2	4.1	4.1		

^aThe intensity measurements assumed uniform quantum efficiency of the CCD camera.

^bThe intensity measurements assumed uniform quantum efficiency of the CCD camera and 0-m attenuation in the PMMA fiber-optic cable.

^cThe signal-increase was due to the reduced attenuation when the fiber-optic cable was shortened from 15 to 0 m.

^dThe fraction of the spectrum from 400 to 710 nm corresponding to the 20-nm-wavelength region.

The portions of each signal component out of the total signal when the BT source is 0.5 cm from the fiber-optic cable and 8.0 cm from the scintillator volume.

Table 4

Signal component	Scintillator material					
	BCF-60	BCF-12	Probe type ^a	Y ₂ O ₃ :Eu	YVO ₄ :Eu	Y ₂ O ₂ S:Eu
Scintillation	18%	18%	B:	98.5%	93.9%	98.6%
			C:	98.3%	94.5%	98.7%
Stem signal	82%	82%	B:	1.2%	3.9%	1.0%
			C:	1.5%	4.9%	1.1%
Photoluminescence ^b	–	–	B:	0.3%	2.2%	0.5%
			C:	0.2%	0.6%	0.2%

^aProbe type C incorporated a long-pass filter between the scintillator volume and the fiber-optic cable to prevent the stem signal in the fiber-optic cable to induce photoluminescence background in the scintillator volume. Probe type B did not incorporate such a filter.

^bThe measured photoluminescence signals of the BCF-12/60 based PSDs were negligible.

The scintillation signals of $\text{Y}_2\text{O}_3\text{:Eu-}$, $\text{YVO}_4\text{:Eu-}$, $\text{Y}_2\text{O}_2\text{S:Eu-}$, and $\text{Gd}_2\text{O}_2\text{S:Eu-}$ during constant irradiation with an ^{192}Ir BT source. The signal intensities were measured during 2 approximately 65-Gy irradiations. The second irradiation was delivered consecutively with a 500 s period in between. The signal intensities at specific times (5, 10, 50, 100, and 200 s) are indicated as percentage differences with respect to the baseline signal intensity 1 s after the start of irradiation.

Table 5

Time after treatment start (dose)	Signal intensity ^a (arbitrary units)							
	$\text{Y}_2\text{O}_3\text{:Eu}$		$\text{YVO}_4\text{:Eu}$		$\text{Y}_2\text{O}_2\text{S:Eu}$		$\text{Gd}_2\text{O}_2\text{S:Eu}$	
	1 st irradi	2 nd irradi	1 st irradi	2 nd irradi	1 st irradi	2 nd irradi	1 st irradi	2 nd irradi
Baseline: 1 s (0.12 Gy)	1.16×10^7	1.12×10^7	2.49×10^6	2.61×10^6	9.56×10^6	9.65×10^6	3.09×10^7	2.96×10^7
5 s (0.6 Gy)	- 0.1 %	+ 0.1 %	+ 0.1 %	+ 0.4 %	+ 3 %	+ 3 %	+ 4 %	+ 3 %
10 s (1.2 Gy)	- 0.2 %	+ 0.1 %	+ 0.2 %	+ 0.6 %	+ 4 %	+ 4 %	+ 6 %	+ 5 %
50 s (6 Gy)	- 1.0 %	- 0.1 %	+ 0.9 %	+ 1.3 %	+ 7 %	+ 6 %	+ 6 %	+ 5 %
100 s (12 Gy)	- 1.8 %	- 0.4 %	+ 1.1 %	+ 1.6 %	+ 8 %	+ 6 %	+ 6 %	+ 5 %
200 s (24 Gy)	- 3.0 %	- 1.0 %	+ 1.5 %	+ 1.7 %	+ 7 %	+ 6 %	+ 5 %	+ 5 %

^aThe percentage values represent changes from the baseline values.