

Implantable real-time dosimetric probe using GaN as scintillation material

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ABSTRACT

This paper presents an implantable real-time dosimetric probe using near-tissue-equivalent GaN as a scintillator and an optical fiber for radioluminescence (RL) collection and transmission. Heavily-doped GaN was preferred because of enhanced light output with dominant near-band-edge UV emission and minimized yellow luminescence (YL) contribution. For encapsulating the small-volume GaN bulk at the end of the coupled fiber, a polymer cladding fiber was chosen and a core cavity was formed by a chemical-etching and tip-cleavage process. For testing the realized dosimetric probe, a probe-readout system was designed with UV-narrow-band selection and photodetection. Real-time measurements showed detected radiation pulses with fluctuations, but integrating the detected pulses exhibited a dose response curve steadily increasing, with less than 2% reproducibility errors between runs for a dose of more than 50 cGy. The GaN and fiber contributions to the averaged output signal were evaluated by different field size measurements. The fiber contribution was significant because of its much larger irradiated volume compared to that of GaN in the probe. On the other hand, the GaN contribution per volume was much larger than the fiber: 186 times for 6 MV photons and 89 times for 18 MV photons. The probe was also irradiated by receiving a 18 MeV 300 Gy electron beam dose. The GaN contribution was unchanged, while the fiber contribution was increased by 20%. This further demonstrates the need to subtract the fiber contribution for accurate measurements.

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1. Introduction

Modern radiotherapy makes use of sophisticated treatments to optimize outcome with minimizing complications, such as 3D treatment planning and conformal or intensity-modulated dose delivery, electron arc therapy, etc. This increases difficulties in assessing with certainty the delivered dose of the targeted region by using conventional indirect dosimetric measurements. One solution to deal with this technical problem is the direct and real-time dosimetric monitoring with implantable probes. There are suggested dosimetric probes. Some are based on radioelectric effects employing either semiconductor devices such as diodes [1] and MOSFETs [2,3], or diamond [4], and others utilize scintillation materials such as Al₂O₃ with optical or thermal stimulation schemes for luminescence output [5]. Unfortunately, semiconductor devices have directional dependence, and many organic and

inorganic scintillators even with activators do not produce sufficient light output to allow design of special-revolved miniature probes. Moreover, most such dosimetric systems are accumulation-type ones, and thus are not suitable for real-time readout of the dose.

In this paper, we present a new and simple implantable real-time dosimetric probe, basically consisting of a small-volume Si-doped GaN bulk used as scintillation material and a coupled optical fiber for collecting and transmitting radioluminescence (RL) output. GaN is a wide direct-gap semiconductor material with high light yields ($\sim 10^5$ Photons/MeV) and very short rising and decay times (<1 ns for its near-band-edge transitions). Its near-band-edge emission in UV range is convenient for the design of real-time RL readout systems using sensitive photodetectors such as photomultiplier tube (PMT) and avalanche photodiode (APD). Its narrow-band UV emission enables background rejection by simple spectral filtering. Furthermore, GaN has an atomic number close to that of water. It is thus nearly tissue-equivalent, and particularly suitable for implantable applications because there will be minimized scattering power variations between the scintillator material and its environmental tissue.

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Fig. 1. Photograph of a Si-doped GaN sample under 6 MV photon beam radiation with integration time of 10 s.

Our studies include tests of materials (GaN and coupled fiber) for their proper choices, realization and testing of the dosimetric probe.

2. Material tests

2.1. Verification of radioluminescence output of GaN

To verify whether GaN has significant RL output under routine radiotherapy conditions, we placed a Si-doped *n*-type GaN sample with 10^{18} cm^{-3} dopant concentration (from LUMILOG, France) under pulsed 6 MV photon beam radiation from a radiotherapy linear accelerator (VARIAN CLINAC 600), at a dose rate of 3 Gy/min. A picture on the exposed GaN sample was taken (Fig. 1) with a cooled CCD camera (LEICA, DFC340-FX). Despite poor UV sensitivity of the camera, the image shows clearly the luminescent sample.

2.2. PL spectral characterization of GaN

To optimize near-band-edge emission of GaN by properly choosing a doping level, we carried out PL measurements on two Si-doped GaN substrates with doping concentrations at 10^{18} cm^{-3} and $1.5 \times 10^{19} \text{ cm}^{-3}$, respectively. The experimental set-up consisted of UV excitation using a frequency-doubled 244-nm Ar laser, and spectral measurements via a grating monochromator (Jobin–Yvon HR 640) equipped with a GaAs PMT (HAMAMATSU 5701). Synchronous detection technique was implemented by chopping the laser beam and employing a lock-in amplifier (EG&G 5209) to process the electrical output of the PMT. Via a set of optical devices, the excitation beam was guided and focused on to the sample under test.

Fig. 2 shows room temperature PL spectra of the two Si-doped GaN samples, with 10^{18} cm^{-3} and $1.5 \times 10^{19} \text{ cm}^{-3}$, respectively. Both exhibiting a dominant emission peak at $E_p = 3.41 \text{ eV}$, which is attributed to band-to-band emission. This band-to-band transition in high-quality GaN at room temperature has preponderant emission over excitonic transitions [6]. The 10^{18} cm^{-3} sample has lower near-band-edge emission intensity compared to that of the heavily-doped one ($1.5 \times 10^{19} \text{ cm}^{-3}$). This is predictable because the radiative recombination rate due to band-to-band transition is directly related to the population of majority free carriers. For the 10^{18} cm^{-3} sample, there are visible contributions of DAP (donor–acceptor-pair) or e–A (conduction-band-to-acceptor) band at 3.28 eV and a broad YL (yellow luminescence) band extending to near IR (infra red). The observed broad YL band is

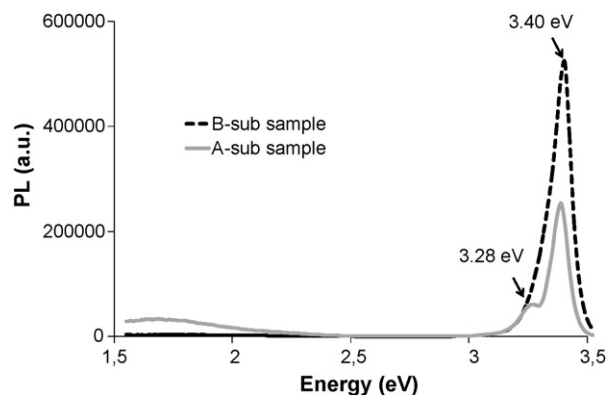


Fig. 2. Room temperature front-side PL spectra of two Si-doped GaN samples with 10^{18} cm^{-3} and $1.5 \times 10^{19} \text{ cm}^{-3}$, respectively.

defect-related, probably related to Ga vacancies [7], and may be attributed to donor-to-deep-acceptor and/or conduction-band-to-deep-acceptor transitions [8]. The heavily-doped sample, on the other hand, has only an intense band-to-band emission peak, without significant contributions of other bands.

It is therefore evident to choose heavily-doped GaN as a transducer for our dosimetric probe. First the band-to-band emission peak is the most suitable signal to be exploited. The high-intensity of the emission peak means a good sensitivity of RL conversion, and the narrow-band emission feature offers possibilities to improve signal-to-noise ratio by optical filtering for signal selection and background rejection. Second the exploited RL signal should have minimized contributions from defect-related emissions, which are ill-controlled.

3. Implantable dosimetric probe

3.1. Realization

The basic structure of the dosimetric probe is a small-volume GaN bulk encapsulated at the end of the coupled fiber. There are different configurations of encapsulation, one of which is to form a cavity at the end of the fiber for GaN incorporation. This configuration of encapsulation has an advantage in small size, because the probe diameter will be the outer diameter of the coupled fiber.

For the configuration, we chose a coupled optical fiber which was a step-index Hard Clad Silica Fiber, HCP-MO400T from SEDI Fibres, France. It was made of a 400 μm -diameter fused silica core, a 15 μm -thick polymer hard cladding, and a 730 μm -external-diameter TEFZEL. This type of fiber utilizes a polymer material as cladding, allowing us to realize a cavity using a chemical-etching and tip-cleavage process. Moreover, it has a large numerical aperture (≈ 0.37) for collecting more efficiently RL output of the GaN bulk, and the TEFZEL buffer is biocompatible.

The cavity of the fiber was realized through a process described as follows. The first step was to form a chemically etched fiber tip, as suggested by [9]. By dipping the fiber distal end in hydrofluoric acid (HF 50%) for 135 min, a silica tip was formed within the optical clad (see Fig. 3a). The tip formation was due to faster etching rates at and near the silica core–polymer cladding interface [10]. This tip was then mechanically cleaved off with a capillary to obtain a cavity (Fig. 3b). A polished silica rod with some diamond paste was introduced in the cavity for polishing the cleaved core surface. Finally the fiber was cleaned in an ultrasound bath and dried. This process preserves the continuity of the optical clad and the TEFZEL buffer (ETFE) for optimal optical and mechanical performances.

Small-volume GaN bulks were obtained by cutting a 1.4-mm-thick GaN wafer from LUMILOG, France with a precision diamond

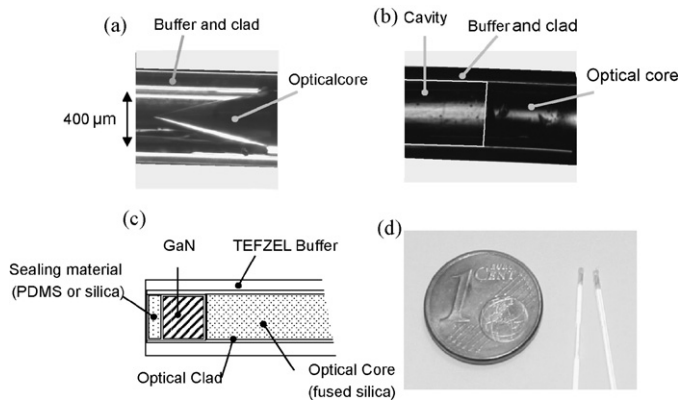


Fig. 3. (a) Etched tip at the end of the polymer cladding fiber; (b) core cavity after tip-cleavage; (c) structure of the dosimetric probe; (d) realized 730 μm -diameter dosimetric probe.

wire saw (Well Diamond Wire Saws, Inc., USA). This cutting tool has a 120 μm stainless steel wire with diamond embedded at the surface of the wire. The obtained GaN bulks had a square section of about 270 $\mu\text{m} \times 270 \mu\text{m}$ and a length equal to the wafer thickness (ranging from 300 μm to 1400 μm).

After introducing the GaN bulk in the fiber cavity, it was enclosed with a sealing polymer material (Fig. 3c). All this microfabrication steps are performed at NanoLyon technological facility.

Fig. 3d shows the realized dosimetric probe.

3.2. Testing

For testing the dosimetric probe, a probe-readout system was designed. It basically consisted of a 385/10 nm interferential band-pass filter and a photomultiplier tube (PMT) H6780-20 from Hamamatsu with a transimpedance amplifier. The band-pass filter served to select near-band-edge RL emission of the GaN signal, so as to minimize wide-band background contributions. The amplifier's output was connected to a PC via an acquisition board (Ch-3160, Acquiretek) for data recording, monitoring and processing. The experimental set-up is shown in Fig. 4.

The probe under test was placed in a 30 cm \times 30 cm \times 8 cm, tissue-equivalent PMMA (Plexiglas[®]) Phantom at the characteristic depth (depth corresponding to maximum absorbed dose: ~15 mm for 6 MeV and 35 mm for 18 MeV photon/electron beams). The probe was positioned for the GaN to be located at the field center with the coupled fiber at normal incidence and extending to the square-field limit at a shortest distance. Thus, for a field size of 2L \times 2L, the exposed segment of the fiber will have a length of L.

4. Results and discussion

4.1. Real-time dose readout

Fig. 5a shows the PMT output signal corresponding to the pulsed radiation detected by the probe. There are random fluctuations

on radiation pulses, as is shown in Fig. 5b. However, integrating the measured pulses over time indicates the absorbed dose, which exhibits a linear increase with the number of radiation pulses (see Fig. 5c). The slope of this dose response curve corresponds to the dose rate. Due to random fluctuations of pulses, the dose response curve may vary slightly for each radiation run, but the evaluated reproducibility errors for a 50 cGy dose, which correspond to about 800 pulses at 6 MV, remain below 2%.

4.2. Dose rate dependence

It is noted that dose rate control of the Linac is carried out by modulating the pulsed radiation frequency instead of changing radiation intensity, typically from 50 Hz to 300 Hz. As the dosimetric probe has a response time in the range of nanoseconds, it is predicted that it has linear dose rate dependence. To confirm this linear response, we measured the integrated RL readout with dose rate variation (in cGy/min). Fig. 6 shows the dose rate response of the dosimetric probe for a 6 MV photon beam from a Varian Clinac 600.

4.3. GaN and fiber contributions

Unavoidably, when the dosimetric probe is placed in a radiation field, one segment of the GaN-coupled optical fiber is also irradiated and thus produces a radiation-induced light superimposed with that of GaN. The fiber contribution is primarily due to Cerenkov radiation, as well as fluorescence or luminescence emission [11]. This optical fiber contribution to the probe output represents background compared to GaN RL signal. As Cerenkov emission has a wide-band distribution [12], narrow-band selection of GaN signal was implemented in the RL readout system to minimize the background contribution, and thus to improve the signal-to-noise ratio (SNR). However, the fiber contribution to the filtered output light may still be very significant, simply because the irradiated volume of the fiber may be a few hundred times that of the GaN bulk.

To estimate GaN and fiber contributions, we may perform series of measurements with different field sizes. As already mentioned, the probe in the radiation field was positioned for this fiber-irradiated length to correspond to the field size 2L \times 2L. Increasing the field size means to increase the irradiated length of the fiber, or its irradiated volume.

Fig. 7 shows the averaged PMT output (in V, arbitrary unit) over 200 detected pulses as a function of irradiated fiber length L (in cm), in 6 MV and 18 MV photon beams, respectively. For a given beam energy, the plotted results exhibit a linear increase versus L. This linear relationship is predictable because the fiber contribution is proportional to its irradiated volume. It also means that the effect of the phantom scattering factor remains negligible for a field size down to 3 cm \times 3 cm. Extrapolation of the curve to Y-axis extracts the GaN contribution, and the slope of the curve gives the fiber contribution per length. Then their respective contributions per volume can easily be evaluated. The obtained values are presented in Table 1. One may estimate that GaN contribution per

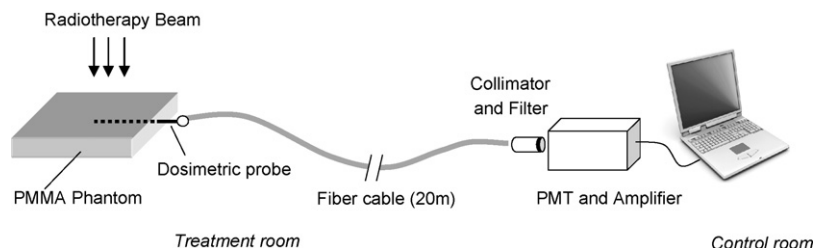


Fig. 4. Experimental set-up for testing the dosimetric probe.

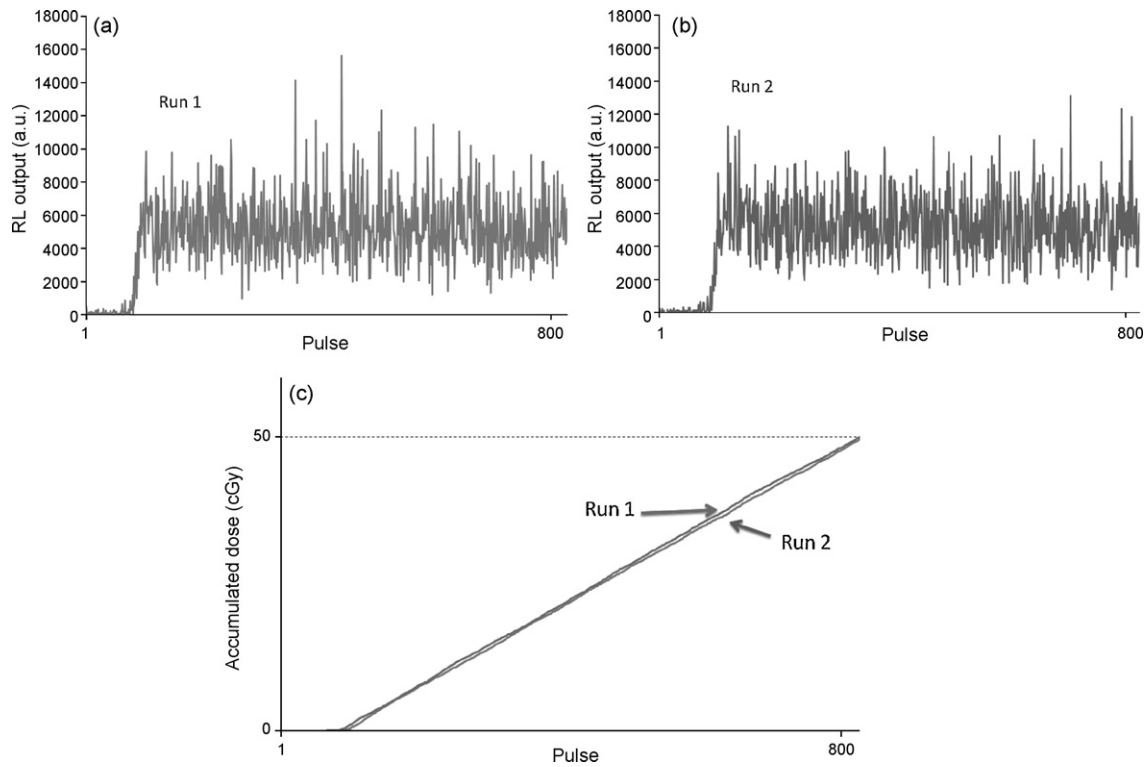


Fig. 5. (a) Real-time detection of radiation pulses; (b) two irradiation runs showing fluctuations of pulses; (c) two dose response curves (by integrating detected pulses), corresponding to the two runs, respectively.

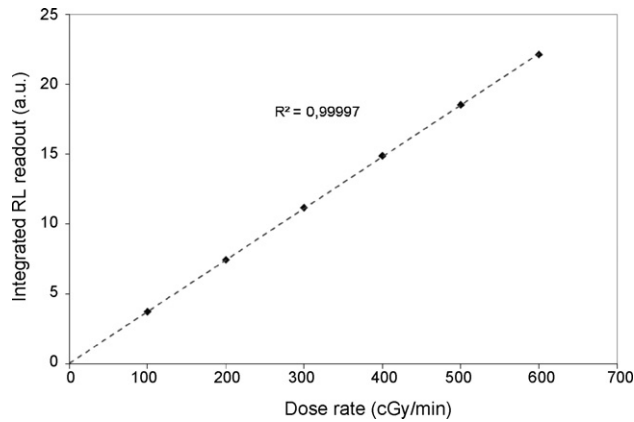


Fig. 6. Averaged RL PMT output versus dose rate.

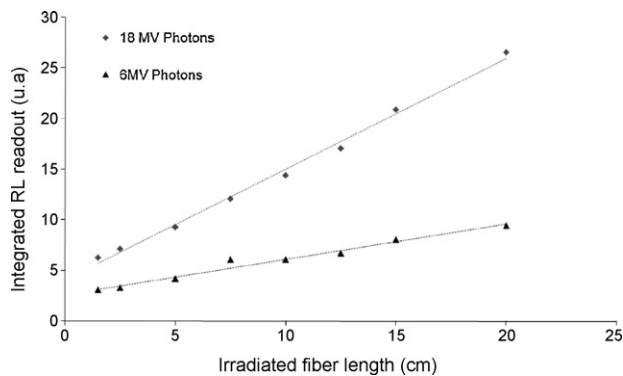


Fig. 7. Averaged PMT output in 6 MV and 18 MV Photon beams, respectively, as a function of irradiated fiber length (directly related to the square field size).

Table 1

Evaluated GaN and fiber contributions in 6 MV and 18 MV photon beams angular dependence.

	6 MV photons		18 MV photons	
GaN contribution	2.64 V	52.6 V/mm ³	4.10 V	79.9 V/mm ³
Fiber contribution	0.347 V	0.283 V/mm ³	1.09 V	0.894 V/mm ³

volume is, respectively, 186 times that of the fiber for 6 MV photons, and 89 times that of the fiber for 18 MV photons. The increase of fiber contribution with higher radiation energy may be accounted for by the Cerenkov effect, which has a strong energy dependence.

As the light output of the probe has fiber contribution which may be due to Cerenkov radiation and thus may have beam/fiber angular dependence, we performed an experiment for different field sizes with angular variation, as illustrated in Fig. 8. The probe under test was placed with GaN transducer at the isocenter of the field. The PMMA thickness was adjusted at each angle to maintain a constant depth (~1.5 cm) of irradiation for the probe.

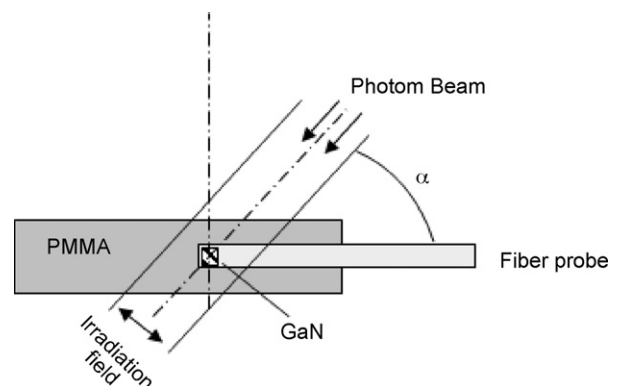


Fig. 8. Experimental configuration for angular dependence measurements.

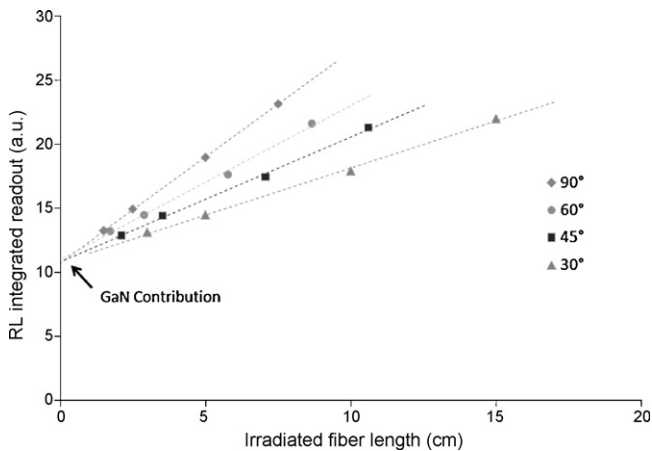


Fig. 9. Angular dependence for a 18 MeV photon beam.

Fig. 9 plots results of measurements using an 18 MeV photon beam. They are presented as a function of irradiated fiber length with beam/fiber angle as a parameter ranging from 30° to 90° . It can be seen that increasing the angle increases the slope of the curve, which corresponds to the fiber contribution per volume. On the other hand, different linear curves can be extrapolated to intercept with Y-axis at the same point, which indicates the GaN contribution. Obviously, the GaN contribution is angular independent.

The angular dependence of the fiber contribution is undesirable for implantable applications because of uncertainty of angular evaluation. This effect can be minimized by reducing fiber contribution to the probe output light.

4.4. Resistance to electron irradiation

Fig. 10 compares results of the averaged PMT output versus irradiated fiber length L , before and after a 300 Gy dose with 18 MeV electron beam radiation. The GaN contribution remains unchanged at 11 V, while the slope of the curve indicating the fiber contribution is increased by 20%. These results demonstrate that GaN is much more resistant to electron beam radiation than the coupled fiber. This reinforces the argument that, for accurate dosimetric measurements, the unstable fiber contribution should be subtracted.

For cancelling fiber contribution, one solution is to employ a 2-fiber bundle for the probe with a blank fiber as reference for differential measurements [13], but this would increase the probe dimensions. There are also optical signal processing methods that can be implemented in the RL readout unit, such as that suggested by [14], to separate and subtract the fiber contribution.

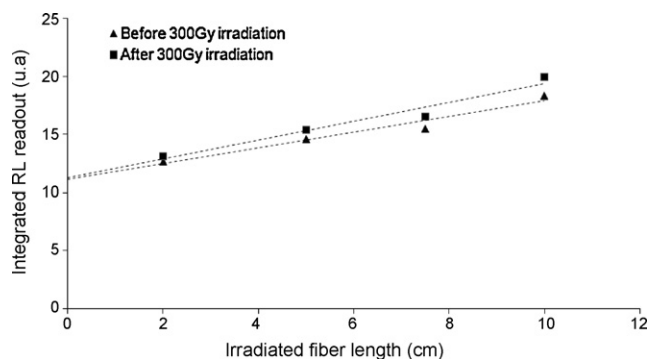


Fig. 10. Averaged PMT output as a function of irradiated fiber length, before and after a 300 Gy dose with an 18 MeV electron beam.

5. Conclusion

We have investigated the use of GaN as a scintillator and presented an implantable dosimetric probe for external radiotherapy application. Heavily-doped GaN was chosen because of its enhanced and dominant UV-narrow-band luminescence output. A polymer cladding fiber was employed and a chemical-etching and tip-cleavage process was proposed to form a core cavity for GaN encapsulation. The fabricated probe was tested with a probe-readout system performing UV-narrow-band selection and photodetection. Real-time measurement with integration of detected radiation pulses plotted a dose response curve, with reproducibility errors below 2% for a dose of more than 50 cGy. Different field size measurements allowed estimation of GaN and fiber contributions to the averaged output signal. After a 300 Gy dose with an 18 MeV electron beam, the GaN contribution remained the same, while the fiber contribution was increased by 20%. We have been working on implementation of a spectral processing technique for subtracting the unwanted fiber contribution. Other characterization work has been undertaken, including evaluation of inter-probe reproducibility and temperature influence.

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Biographies

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Jean-Yves Giraud received the M.S. and PhD degrees in medical physics from University Paul Sabatier Toulouse, respectively in 1987 and 1991. He worked as an R&D engineer for a medical device company from 1991 to 1993. From March 1993 to September 1997, he has held a Medical Physicists position at the university hospital of Grenoble. He has been a visiting scientist at the Harvard Medical School, Boston, USA from 1997 to 1998. Since October 1998, he returned to his position in Grenoble where his current research interests include image guided radiation therapy and in vivo dosimetry.

Jacques Balosso received his M.S. degree in 1989, the MD degree from University of Montpellier in 1990, and his PhD degree from university Paris VII in 1997. In 1991, he was an assistant professor at the Saint Antoine University, Paris. From 1991 to 1993, he spent two years at the Harvard school of Public Health, Boston, USA. Since 2003, he has been a full professor at university hospital of Grenoble. He is also the director of the national centre for light-ion hadrontherapy in France called ETOILE.