

Evaluation of low energy electron beam dose application by means of a portable optical device

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Abstract. We present our recent development concerning the evaluation of a low energy dose application to electron beam responding materials with a simple portable optical device. Electron beam irradiation is a promising option to sterilize sensitive and high performance products or surfaces at a low temperature and without moisture. Especially in the fields of the food industry and medicine, regulations regarding sterility are increasingly tightened. Because of this, a secure proof for electron-beam-assisted sterilization is required. However, no non-destructive and *in situ* method exists up until now. Our approach to provide a secure proof of sterilization is to place a suitable marker material based on rare-earth-doped phosphors inside or on the top of the packaging material of the respective product. Upon electron irradiation the marker material changes its luminescence properties as a function of the applied energy dose. We verified the energy dependence by means of time-resolved measurements of the luminescence decay of an upconversion phosphor with a portable optical device. In our experimental realization, short laser pulses in the near-infrared range are triggered by a microcontrol unit (MCU) and excite the marker material. The light emitted by the marker is collected in the range between 400 and 1100 nm via a silicon photodiode, processed by the MCU, and analyzed in a Labview program via a single-exponential fit. As a main result, we observe an increasing reduction of the luminescence lifetime with higher dose applications. © 2014 Society of Photo-Optical Instrumentation Engineers (SPIE) [DOI: 10.1117/1.OE.53.11.114102]

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1 Introduction

Recent application of sensitive materials as high-performance polymers and the integration of sophisticated functionalities such as electronics and ultrasonic sensors in medical products lead to an essential need for alternative sterilization processes in life sciences. An auspicious alternative to classical sterilization methods is electron beam treatment. Up until now, no reliable *in situ* proof of dose application exists. Currently applied microbiological methods or even dosimetric approaches are time consuming. The authors reported in previous proceedings on an optical technique which provided proof of electron beam sterilization with an optical active material.^{1,2} Up until now, measurements have been realized with a bulky laboratory setup with expensive components. Envisaging an application, a transfer of this optical technique to rather simple electronics would be desirable. Hence, the evaluation of sterilization can be realized with a portable optical device, as is presented by the authors in this contribution.

2 Background

2.1 Electron Beam Sterilization

Electron beam irradiation is a promising option for classical sterilization methods due to its advantages such as safety, high speed processing, and applicability to sensitive

materials. The technique relies on its effect on microbial germs on, e.g., surfaces of medical products or food packaging.³

The methodology for monitoring the application of an electron beam previously presented by the authors is based on the integration of an optical marker material in or on the packaging material, which changes its optical properties upon electron beam irradiation.^{1,2} In addition to the change of optical properties, qualified materials are required to be nontoxic, mechanically and chemically stable and integrable into packaging materials. One material class which fulfills these requirements are upconversion (UC) phosphors based on lanthanide-doped host lattices.

2.2 Upconversion Phosphors

In this paper, the authors present time-resolved luminescence results on $\text{NaYF}_4:\text{Yb}^{3+}, \text{Er}^{3+}$. The Er^{3+} -doped phosphor exhibits a strong green and red UC as depicted in the steady state spectra with the assigned emission bands in Fig. 1.

The UC process is a transformation of two or more low energy pump photons in the near-infrared through the population of long-lived intermediate states into one emitted higher-energy photon in the visible range. The processes of UC can be explained with different models. In general, two main mechanisms can be differentiated: excited state absorption (ESA) and energy transfer upconversion (ETU).⁴⁻⁶ In the case of ESA, at least two photons of the same energy are sequentially absorbed by the activator

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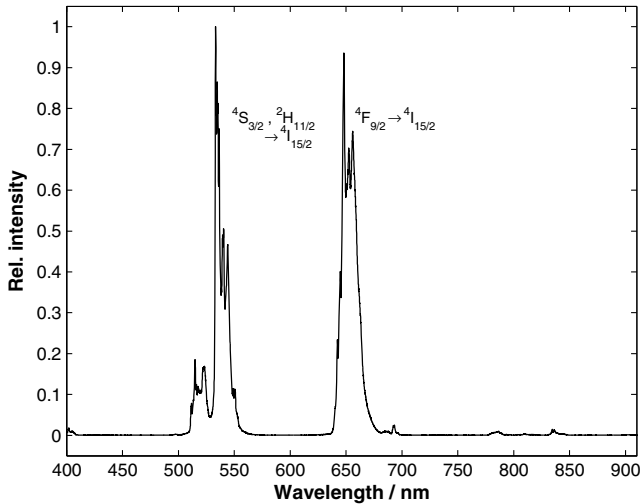


Fig. 1 Upconversion (UC) spectra of $\text{NaYF}_4:\text{Yb}^{3+}, \text{Er}^{3+}$ irradiated at 980 nm.

(A) to reach the excited state [Fig. 2(a)]. However, in the case of ETU, the absorption of the first photon by the activator is followed by an energy transfer from the neighboring sensitizer (S), which results in population of the higher excited state (E^{**}) [Fig. 2(b)]. ETU was first described independently by Auzel,^{7,8} Ovsyankin and Feofilov.⁹ ESA and ETU should be distinguished from the two nonlinear optical processes of two-photon absorption and second harmonic generation, which only occur efficiently by coherent excitation.^{5,6}

3 Experimental Details

3.1 Electron Beam Exposure

Samples of printed badges of the material were prepared on a cover slide. The specimens were processed in the REAMODE, a low energy electron beam equipment at Fraunhofer Institute for Plasma and Electron Beam Technology FEP Dresden, Germany. An electron acceleration voltage of 160 keV and doses in steps of 10 kGy up to 100 kGy and in steps of 50 kGy up to 300 kGy as measured at the sample surface were applied. The number of samples for each dose was five.

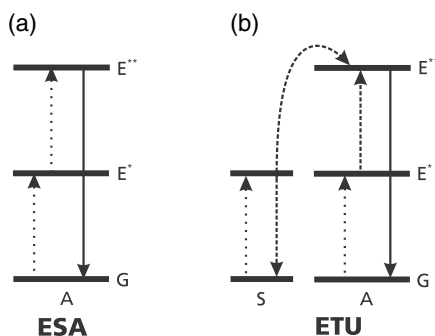


Fig. 2 UC mechanisms for lanthanide-doped crystals: (a) excited state absorption, (b) energy transfer upconversion.: photon excitation, —: energy transfer, —: emission process.⁴

3.2 Interrogation of Sample Material

Samples are placed directly in front of the exciting laser diode and detecting photodiode without any further sample preparation. A silicon photodiode without additional filters is used as detector, hence, acquired data include all emitting states (UC, elastic scattering, down conversion). Emission is integrated in the range from 400 to 1100 nm.

3.3 Data Analysis

The luminescence lifetime τ is obtained by fitting the intensity I over time t plot to the single exponential decay function in Eq. (1), where I_0 is the intensity at $t = 0$. To avoid the influence of the laser on the decay curve, fitting only starts with a precisely defined delay of 80 μs . For each measurement, 10 slopes were collected and averaged.

$$I(t) = I_0 e^{-t/\tau} + C. \quad (1)$$

4 Results and Discussion

4.1 Development of the Portable Optical Device

In contrast to the previous report,² the authors focus on the simple realization of sterilization evaluation with a portable optical device for commercial application. The scheme of our portable device is depicted in Fig. 3 and the actual setup in Fig. 4. The samples as described in Sec. 3.1 are excited with a 50-mW laser diode at 980 nm (No. 2 in Fig. 4) that is controlled by a microcontrol unit (MCU). Light emitted from the sample is collected via a silicon photodiode (No. 1 in Fig. 4) situated next to the laser; afterward the signal is amplified by a transimpedance amplifier. The amplified signal is then digitalized by the analogue digital converter with a sample rate of 500 kSPS. An MCU caches data in a ring buffer and the data are submitted to an external analysis unit via universal series bus (No. 5 in Fig. 4) and finally analyzed with LabVIEW. The power supply is realized with connections No. 3 and 4 in Fig. 4. The obtained data are displayed in the plot of Fig. 4. Luminescence intensity is plotted over samples (sample rate 500 kSPS). Samples are collected at equidistant time periods which allows for a conclusion of the time constant after the fitting.

4.2 Evaluation of Dose Dependence

In Fig. 5, luminescence lifetime τ is plotted over the applied dose of electron beam irradiation. A distinct reduction of luminescence lifetime can be observed with increasing dose. The most vigorous reduction of luminescence lifetime τ takes place after the initial exposure to the electron beam and the degree of decrease reduces with a higher dose application. The data show evidence of an exponential decay as

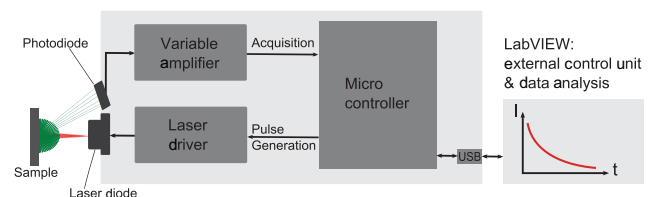


Fig. 3 Schematic view of the optical set up for time-resolved measurements in our portable optical device.

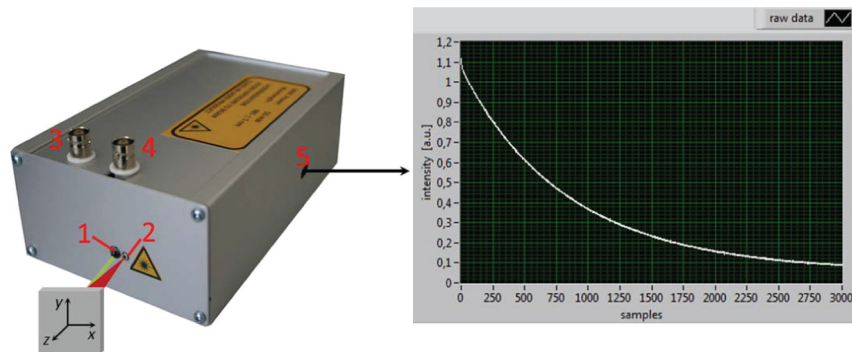


Fig. 4 Actual hand-held device and resulting LabView data.

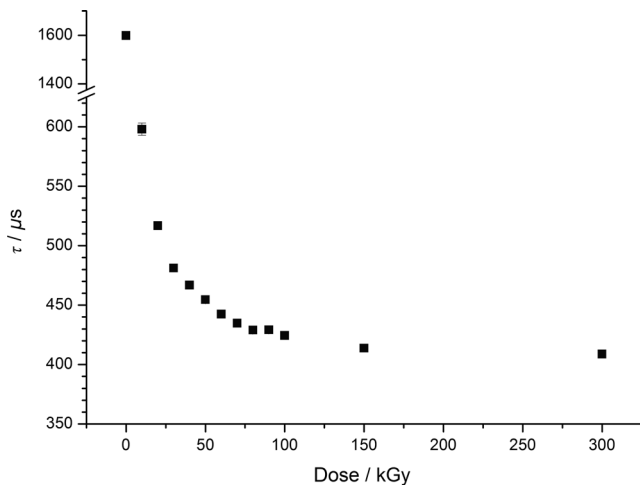


Fig. 5 Dependence of luminescence lifetime τ from applied dose on $\text{NaYF}_4:\text{Yb}^{3+}, \text{Er}^{3+}$ sample.

dose—decay time dependence, but additional evaluation and irradiation experiments are needed for the calculation of a reliable decay constant. However, the obtained data clearly show the capability of our simplified device to evaluate the optical response of the marker material.

5 Conclusion and Perspectives

In this contribution, the authors showed the proof of concept of evaluation of a low energy electron beam dose application with a portable optical device. In spite of the low light collection efficiency and simple amplification electronics, the device turned out to be well suited to measure the change in luminescence decay time of UC phosphors after electron beam exposure. The integrated luminescence in the range between 400 and 1100 nm shows an evident decrease of the luminescence lifetime due to the exposure. Further,

the first hints of an exponential dose dependence were revealed, but additional evaluation and irradiation experiments are necessary.

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