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Sol–gel derived copper-doped silica glass as a sensitive material for X-ray beam dosimetry

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ABSTRACT

The light emission from a sol–gel-derived Cu-doped silica glass was studied under 10 keV X-ray irradiation using a fibered setup. Both radioluminescence (RL) and optically stimulated luminescence (OSL) were analyzed at different high dose rates up to 50 Gy/s and for different exposure times, yielding accumulated doses up to 50 kGy (in SiO_2). Even if a darkening effect appears at this dose level, the material remains X-sensitive after exposure to several kGy. At low dose rate, the scintillation mechanisms are similar to photoluminescence, involving the Cu^+ ions electronic levels, contrary to the nonlinear domain (for dose rates higher than 30 Gy/s). This RL, as well as the OSL, could be exploited in their linear domain to measure doses as high as 3 kGy. A thorough study of the OSL signal has shown that it must be employed with caution in order to take the fading phenomenon and the response dependency on stimulation source intensity into consideration.

1. Introduction

Monovalent copper ions in various matrix systems have attracted considerable attention since the 1990s, due their prominent light emission in the blue–green region, especially when they are excited by ionizing radiations. In effect, when excited by UV light, Cu^+ ions in silicate glass present an absorption cross section two orders of magnitude higher than those of trivalent rare-earth ions [1]. Different mechanisms were demonstrated for light emission of Cu^+ ions, like thermoluminescence (TL) [2,3], lyo- and mechano-luminescence [4], scintillation or optically stimulated luminescence (OSL) [2,5], that could be exploited for X-ray or γ dosimetry. The use of such an efficient emitting ion, when incorporated in optical fiber-based dosimeters, is of great interest for all applications requiring real-time monitoring over a long distance, such as radiotherapy, nuclear waste surveillance or/and when high spatial resolution is needed. In particular, copper-doped quartz glass was used as the scintillating material in a remote optical fiber-coupled dosimeter sensitive to X-rays through both radioluminescence (RL) and OSL processes. Both types of signals were shown to behave linearly with the irradiation dose in a range between 1 cGy and more than 10 Gy [6,7]. A gated fiber–optic-coupled system based on a Cu^+-doped fused quartz fiber was also validated for electron beam dose measurements [8]. However, the short quartz fiber piece used in these studies was doped by thermal diffusion with concentration of only a few ppm copper ions. Moreover, the dose range over 100 Gy has not yet been explored with this kind of material, so that information about both the limit of linearity and radiation hardness is still needed and required.

Recently, we published the study of a sol–gel-derived copper-doped silica glass rod and its corresponding microstructured fiber [9]. This fiber exhibited a green photoluminescence (PL), the intensity of which was linear to the UV laser incident power. In this doped glass, a significant increase in the Cu^+/Cu^{2+} ratio was reported and demonstrated when the densification heat-treatment was carried out at 1200 °C in neutral atmosphere, yielding a Cu^+ concentration of 110 ppm in pure silica and a PL quantum efficiency as high as 40% [10]. In the present paper, we investigated the luminescent properties of this novel sol–gel material under X-ray source delivering doses up to several kGy. Both scintillation and OSL signals were acquired from a Cu^+-doped glassy rod and we showed that, to a certain extent, these reproducible signals are suitable for measuring the dose rate and the total dose delivered by the accelerator.

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2. Experimental

A copper-doped glassy rod was prepared according to a procedure described elsewhere [10]. Briefly, a monolithic porous xerogel cylinder, synthesized through a base-catalyzed sol–gel process and using tetraethylorthosilicate (TEOS) as a precursor, was soaked in a solution of copper (II) hexafluoroacetylacetonate hydrate and dried in air to remove the solvent. The sample was then densified in helium gas atmosphere in order to promote the reduction of Cu^{2+} into Cu^{+} ions. Finally, a thin transparent rod of 0.5 mm diameter was drawn from this bulk sample.

For RL and OSL measurements, a 3 cm-length piece of this thin rod was optically polished at both ends, put in a fitted pure silica sleeve and connected at both ends to 0.5 mm-diameter multimode optical fibers, as shown in Fig. 1. The sample emission signal was guided *via* one of these fibers to an airtight box containing selective filters at 550 nm and a compact photomultiplier tube (PMT, Model H9305-03, Hamamatsu). In OSL experiment, the excitation signal is delivered from a fibered laser diode at 660 nm (LP660-SF60, Thorlabs). This stimulation wavelength was chosen as a compromise between a high stimulation efficiency for copper monovalent ions in silica [7] and the absence of photoluminescence generation. In this case, a spectral filter was designed to select the OSL signal (around 550 nm) while rejecting the laser diode pump at 660 nm.

The luminescent rod in its sleeve, as shown in Fig. 1, was fixed 1 mm above a plastic plate. The Cu-doped rod was placed at room temperature in an irradiation machine (Probix) including an X-ray tube with a tungsten anode that provides photons of 8.4 and 10 keV energies. This energy range is far from the one usually involved in radiotherapy systems, but is rather common in radiographic imaging ones or in investigations targeting the potential technologies to operate in a space environment. Furthermore, for energies lower than 195 keV in silica, no Cerenkov effect interferes with the RL signal. A circular 2 cm-diameter mask was used in order to delimit the irradiation zone regarding the sensitive rod, thus reducing stem effects. The Probix machine continuously delivers X-ray dose rates from 1 to 50 Gy/s, enabling us to test the hardening properties of the material. All the experiments were performed at room temperature.

The RL emission spectroscopy was carried out by using a fibered spectrometer (QE65 pro, Ocean Optics) connected to the microscope objective. As the collected signal was very weak and noisy, statistical treatment involving a high number of measurements had to be performed to extract a smoothed spectrum.

3. Results and discussion

Fig. 2 shows the RL signal from the Cu-doped rod under irradiation for about 100 s with various dose rates, reproducibly collected through the optical fiber and without any filter before the PMT. The inset exhibits the average PMT response in permanent regime, after a background subtraction, as a function of the dose rate. The behavior is clearly linear up to 30 Gy/s with a slope of about 14.7 mV s/Gy, but the sensitivity of the system significantly increases at the higher dose rates. Such a result is counter-intuitive since it is expected that, like in the case of photoluminescence, a high irradiation intensity normally fills the electron levels of the Cu^{+} ions, leading to a saturation of the light emission. A possible explanation could be found in the permanent modification of the material under irradiation at the highest dose rate, creating new shallow defect centers, which allow new emission bands. However, this hypothesis must be ruled out since in this case, after exposure

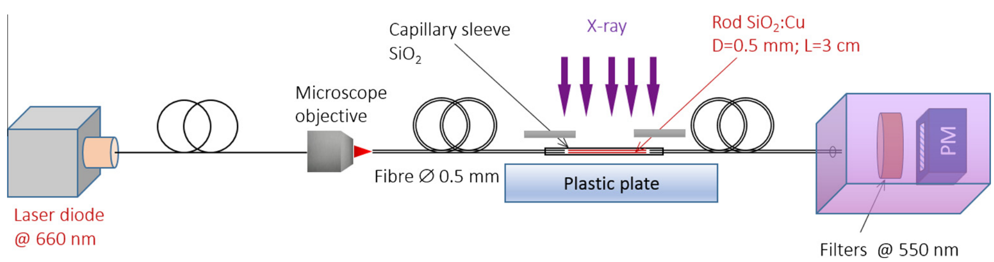

Fig. 1. Schematic view of the RL and OSL measurement setup and a photograph of the luminescent rod excited by a UV lamp.

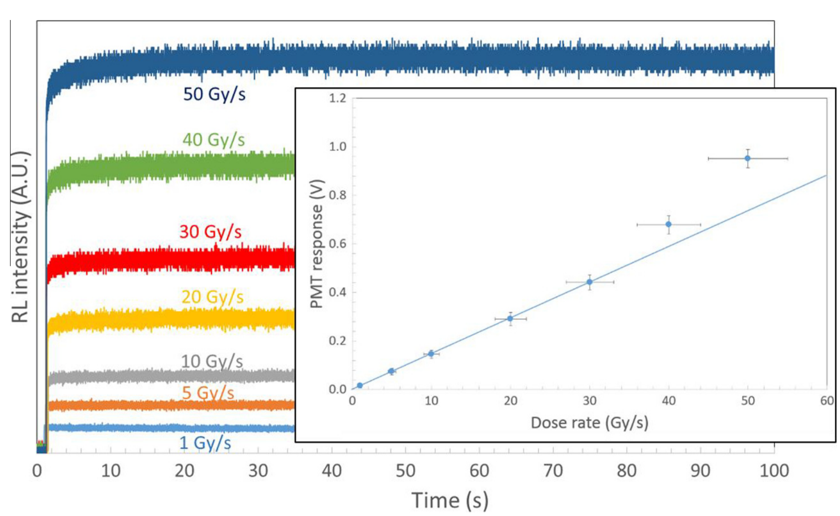

Fig. 2. Rough RL signal as a function of time, recorded after opening the X-ray shutter at different dose rates. Inset: mean RL signal as a function of the dose rate.