Journal of Sol-Gel Science and Technology 21, 173–176, 2001

A New Manufacturing Process to Obtain Thermoluminescent Dosimeters Using Sol-Gel Method

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Received July 18, 2000; Accepted January 23, 2001

Abstract. A new manufacturing process using sol-gel method to obtain thermoluminescent dosimeters—TLDs is described. A LiF/SiO_2 thermoluminescent dosimeter in the form of reusable solid chip was prepared using an acid-catalyzed sol-gel process, with tetramethoxysilane (TMOS) and LiF powder as precursors. In comparison with the manufacturing process of typical lithium fluoride based dosimeters, the method optimizes the manufacturing process. The new dosimeter has a dose response over a dose range varying from 0.1 Gy to ~160 Gy and can be used to detect higher doses using the typical commercial TLD readers without special adjustments. Furthermore, the process may be used for the preparation of other TLDs using a simple chemical processing.

Keywords: sol-gel, thermoluminescent dosimeter, lithium fluoride dosimeter

1. Introduction

Sol-gel chemistry for the synthesis of new materials is becoming increasingly popular because of its relatively simple components and the variety of applications to which it can be applied [1–4]. The method involves the hydrolysis of a glass precursor—tetramethoxysilane (TMOS) for silica glass—in solution, and, therefore allows for the incorporation of several additives before the glass is formed [1, 5–8]. The ability of the sol-gel process to yield optically transparent materials has been exploited to produce glasses for optical applications [9, 10].

Daniels et al. [11] first suggested the use of thermoluminescence (TL) as a technique in radiation dosimetry. They had appreciated that irradiated materials contains stored energy which could be thermally released. In the case of insulating crystals some of the decay routes for the thermally released charges generate light (thermoluminescence). Since photomultipler tubes can detect very low light levels the overall process of TLD (thermoluminescence dosimetry) offers a sensitive method of detection of radiation history for the crystals [11].

Applications of thermoluminescence dosimetry occurs in medicine, nuclear reactor environments, in spacecraft, and for archaeological and geological dating, mineral prospecting, food irradiation, product sterilization, polymer modification, etc. [11–14]. This wide range of applications covers factors of 10^6 or more in total dose, dose rate and storage time. The use of conventional TLDs in monitoring high dose

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radiation (for example from 10^2 Gy to 10^6 Gy) can be somewhat limited, however, due to the onset of sublinearity (saturation) of the TLD response [12]. New and improved dosimeters are desirable in all these areas [12].

Lithium fluoride based dosimeters have been in use for many years [14]. The material is available in powder, extruded and pressed chip forms [12, 15]. The current method of producing these materials consists of several steps: purifying, growing crystals, grinding, blending, pressing, slicing and dicing [15]. Recently a new manufacturing process have been developed by Bicron (Harshaw) that replaces 4 steps with a pelletising process [15].

We developed a new manufacturing process using sol-gel method to obtain a lithium fluoride based thermoluminescence dosimeter in a chip form, using as precursors TMOS and LiF powder (patent pending). This new TLD obtained has good optical properties and good resistance to annealing and thermal quenching, characteristics desirable for a TLD.

2. Materials and Methods

SGS-LiF Synthesis

The reagents employed in the SGS-LiF thermoluminescent dosimeter (SGS stands for *s*ol-*g*el *s*ilica) synthesis were tetramethoxysilane ($Si(OCH_3)_4$, Aldrich Chemical; grade assay 98%, used without further purification), lithium fluoride (TLD-100 powder, LiF:Mg, Ti, Bicron-NE Harshaw—USA), nitric acid (concentrated, reagent grade, Synth Lab. Prod.—Brazil, used as received) and deionized water (Millipore Q uv/plus, electrical conductivity $\sim 0.05\ \mu S \cdot cm^{-1}$ at 298 K).

Water, TMOS and HNO_3 were combined and the gel was obtained by mixing with a solution containing LiF. The concentration was chosen in order to have the molar ratio TMOS:H_2O equal to 1 : 32 M and the LiF contents varied from 2.3×10^{-2} M up to 2.1×10^{-1} M.

The homogenized solutions were poured into flasks which were sealed. The wet gels were aged at room temperature for about 4 weeks. After gelation the samples were heated to 100°C at a rate of $6°C \cdot h^{-1}$, where they were allowed to dry for 2 h. The samples were then ramped at the same rate to 450°C, where they were maintained for one hour. Finally they were cooled back to room temperature for a period of 36 h. After the thermal treatment, the SGS-LiF xerogels appeared homogeneous and opaque white, with the

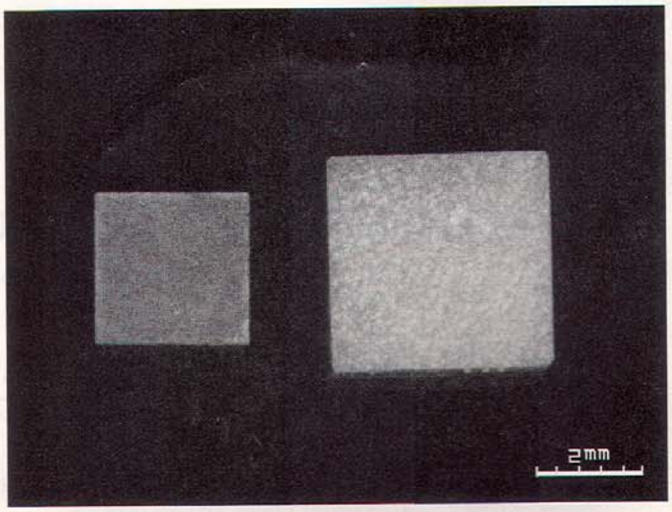

Figure 1. SGS-LiF TLD chip synthesized (right) and TLD-100 chip, Bicron Harshaw.

samples typically in a 4 mm × 4 mm × 2 mm chip form, Fig. 1.

Irradiation Facilities

Gamma irradiation were carried out using ^{137}Cs source and ^{60}Co γ-ray source at dose levels from ~0.1 Gy to approximately 1000 Gy.

The dosimeters were first calibrated in a ^{137}Cs γ-ray source and subsequently irradiated in a ^{60}Co γ-ray source under electronic equilibrium conditions [16]. Fricke dosimetry was used in order to estimate the error induced by the difference in the field homogeneity between the calibration source and the gamma cell unit [17].

3. Results and Discussion

X-ray diffractograms (XRD) of the xerogels were obtained (Rigaku, Geigerflex) at a scanning rate of $0.13° s^{-1}$ in the 2θ range from 5° to 70°, using graphite-monochromated Cu-K_α radiation, Fig. 2.

The X-ray diffraction pattern of the xerogel obtained from the reaction TMOS + H_2O shows a typical amorphous background (Fig. 2), characteristic of amorphous silica with no diffraction pattern indicating the presence of a crystalline. The SGS-LiF XRD pattern (Fig. 2) shows a typical pattern indicating crystalline phases, with peaks at 2θ ~39°, ~45° and ~65° with similar energy of XRD pattern presented by the LiF spectrum, Fig. 2. The low intensity of all peaks observed in SGS-LiF TLD X-ray diffratogram pattern is due to the small amount of LiF presented ~2.3 × 10^{-2} M.

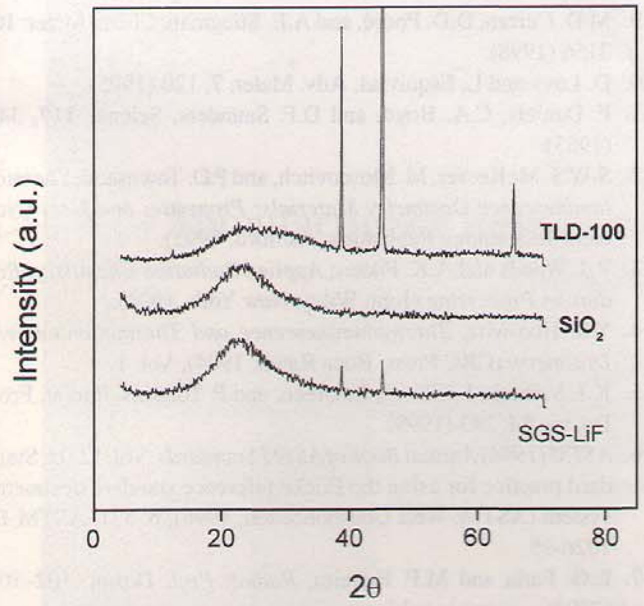

Figure 2. Typical XRD of SGS-LiF TLD chip synthesized, TLD-100 chip Bicron Harshaw chip and SiO_2 chip.

Some Characteristics of the SGS-LiF Thermoluminescent Dosimeter

The TL detectors were read out in a Harshaw 4500 TLD Reader utilizing the same time temperature profile used for typical TLD-100 elsewhere. The signal/noise ratio measured with an internal light source was 35100 under 890 V, 30 s reading cicle and $10^{\circ}\mathrm{C}\cdot\mathrm{s}^{-1}$ heating rate.

The glow curve (intensity of the TL signal vs. time or temperature) [14] comparing the detection levels of the SGS-LiF and the TLD-100 is shown at Fig. 3, indicating the highest detection level of each TLD. We observe that the SGS-LiF can detect, with the same current level at the photomultiplier tube, a dose ~50 times higher than TLD-100. It should be noted that the most intense peak in the SGS-LiF is the (so-called [18]) *peak 3*, while in the TLD-100 it is the (so-called [18]) *peak 5*. This is due to the saturation of *peak 5* for higher doses.

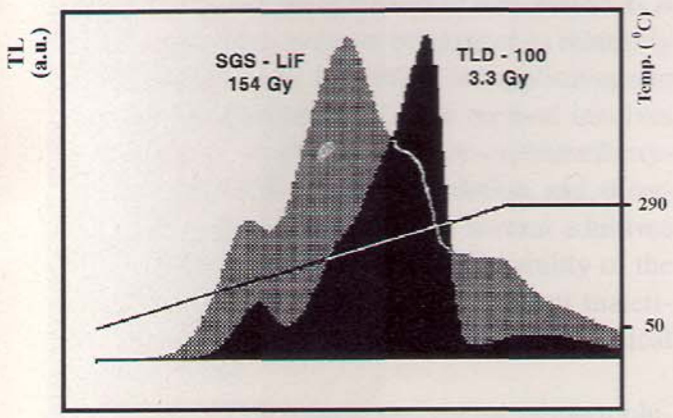

Figure 3. Typical glow curve absorption spectra from SGS-LiF TLD chip synthesized (gray) and LiF-100 Bicron Harshaw chip following gamma radiation.

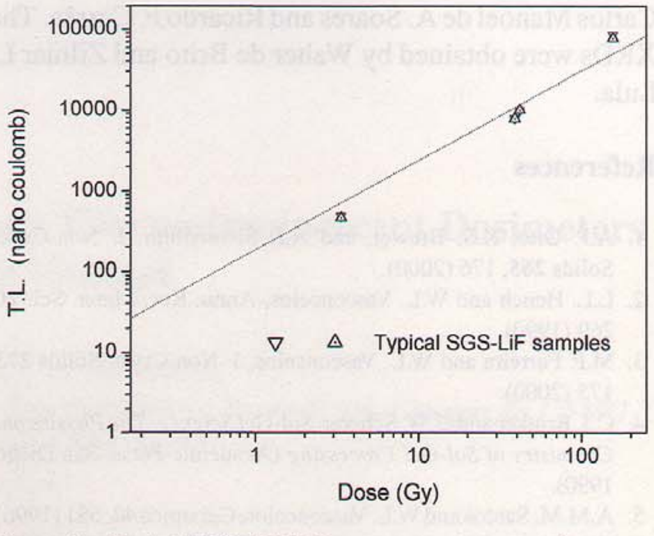

Figure 4. Typical SGS-LiF TLDs response on gamma doses up to $\sim 1.5 \times 10^2$ Gy. The nano coulomb stands for the electric charge at the photomultiplier tube (The dashed line is a guide to the eyes).

The minimum measurable dose ~0.1 Gy was estimated irradiating a series of samples with monoenergetic fields of ^{137}Cs around this range at our Secondary Dosimetric Laboratory.

The typical SGS-LiF thermoluminescent dosimeter response on the gamma dose ranging from ~1 Gy to $\sim 1.5 \times 10^2$ Gy is shown at Fig. 4, showing a linear dose response.

4. Conclusion

A new manufacturing process using an acid-catalysed sol-gel method to obtain thermoluminescent dosimeters-TLDs is described. The new dosimeter in the form of reusable solid chip has a dose response over a dose range varying from 0.1 Gy to ~160 Gy and can be used to detect higher doses using the typical commercial TLD readers, without special adjustments. The method optimizes the usual TLDs manufacturing process and may be used for the preparation of others TLDs using a simple chemical processing.

Acknowledgments

All the irradiation procedures with ^{137}Cs γ-ray source and ^{60}Co γ-ray source were performed respectively by

Carlos Manoel de A. Soares and Ricardo F. Corrêa. The XRDs were obtained by Walter de Brito and Zilmar L. Lula.

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