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Systematic development of new thermoluminescence and optically stimulated luminescence materials

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ABSTRACT

This paper presents an overview of a systematic study to develop new thermoluminescence (TL) and optically stimulated luminescence (OSL) materials using solution combustion synthesis (SCS) for applications such as personal OSL dosimetry, 2D dose mapping, and temperature sensing. A discussion on the material requirements for these applications is included. We present X-ray diffraction (XRD) data on single phase materials obtained with SCS, as well as radioluminescence (RL), TL and OSL data of lanthanide-doped materials. The results demonstrate the possibility of producing TL and OSL materials with sensitivity similar to or approaching those of commercial TL and OSL materials used in dosimetry (e.g., LiF:Mg,Ti and Al_2O_3:C) using SCS. The results also show that the luminescence properties can be improved by Li co-doping and annealing. The presence of an atypical TL background and anomalous fading are discussed and deserve attention in future investigations. We hope that these preliminary results on the use of SCS for production of TL and OSL materials are helpful to guide future efforts towards the development of new luminescence materials for different applications.

1. Introduction

Thermoluminescence (TL) and Optically Stimulated Luminescence (OSL), also called Photo-Stimulated Luminescence (PSL), are techniques widely used in radiation dosimetry [1–4], luminescence dating [1,5,6], and computed radiography [7]. They rely on the stimulated recombination of electrons and holes created by ionizing radiation and trapped at defects in the crystalline lattice of the host material, leading to luminescence whose intensity is related to the energy deposited in the detector by ionizing radiation (i.e., absorbed dose). In TL the stimulation is provided by controlled heating of the detector [3,4]. In OSL, stimulation is provided by controlled illumination [1,2].

In spite of the widespread use of TL and OSL, a demand exists for new materials with tailored properties for specific applications, including OSL neutron dosimetry, 2D dose mapping and temperature sensing, as discussed below.

There are a limited number of OSL materials for personal dosimetry application, particularly for neutron dosimetry. Only two materials are commercially used in OSL dosimetry, Al_2O_3:C and BeO, and this limited availability has been pointed out as a weak point of the OSL technique [2,8]. Moreover, these materials do not have a high cross-section for neutron interaction, which means that they cannot be used as neutron detectors [8]. This problem has been partially solved by preparing detectors made of a mixture of OSL material and neutron converters [9,10] such as ^{6}Li or ^{10}B, which convert neutrons into charged particles [11]. Although this solution is commercially satisfactory [12], higher neutron sensitivities could be achieved using new OSL materials containing ^{6}Li or ^{10}B as part of the crystalline structure, which would require the development of new OSL materials based on compounds such as $Li_2B_4O_7$ or MgB_4O_7.

Two-dimensional dose mapping in medical dosimetry, particularly in quality assurance and dose verification in radiotherapy, is another area of potential application of OSL materials. Although 2D dosimetry has been performed using TL [13–16], an all-optical technique such as OSL would be a better choice for this type of application, as evidenced by the use of the OSL in computed radiography [7]. The main problem with using photostimulable phosphors used in computed radiography, such as BaXBr (X=F, Cl, Br) and CsBr, for dosimetry is their high effective atomic number [17,18] ($Z_{eff}\sim 50$) and signal fading ($>50\%$ in 36 h) [7,19]. One-dimensional dose mapping using Al_2O_3:C OSL detectors has been used in computed tomography [20–22], but the luminescence lifetime of the main luminescence centers in Al_2O_3:C (35 ms) is too long for 2D dosimetry readout by spot-scanning

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laser. OSL systems based on BeO or SrS have been described [23,24], but these systems present problems such as limited spatial resolution or high effective atomic number of the detector material (e.g. $Z_{eff}=34.6$ in the case of SrS) [2].

More recently, renewed interest has been expressed in the use of TL for temperature sensing, in particular as passive temperature sensors in biological agent defeat tests [25], but the lack of suitable materials is also one of the main obstacles. The concept is based on the fact that charges trapped at different energy levels within the conduction band are affected differently by the temperature experienced by the particles, and this can be quantified by measuring the TL curves of particles previously irradiated: depending on the time–temperature profile, the TL peaks would be erased differently. For this application, materials with multiple TL peaks that are light-insensitive are required. Unfortunately, a survey of existing TL materials reveals most of them to be light sensitive [3]. LiF:Mg,Ti is an exception, but this material is known for having a complex defect structure, which causes the TL properties to be dependent on the entire temperature history, both before and after irradiation [3].

Because of the complex nature of the TL and OSL processes, which require the presence of both recombination and trapping centers introduced by intrinsic or extrinsic defects in the host material, development of new materials has been serendipitous. Often, the nature of the recombination centers is known because of its characteristic emission spectrum, but that of trapping centers responsible for the TL/OSL signal is not.

Recently, two developments increased the chances of more precisely engineering the TL and OSL properties of materials. The first development is the demonstration that chemical routes such as solution combustion synthesis (SCS) [26–29] may offer a more efficient way to synthesize TL/OSL materials [30–33] and investigate the role of dopants in the TL and OSL process. The second is the understanding that the energy levels introduced by lanthanide (Ln) dopants and their role in the TL (and possibly OSL) process can be predicted based on a few parameters [34–37].

Based on these developments and motivated by the lack of suitable TL and OSL materials for different applications, we initiated a systematic study to develop new TL and OSL materials with properties tailored for the specific applications discussed above. Our approach uses SCS as the main synthesis method, accompanied by characterization of the crystal structure and luminescence properties of the materials produced.

The objective of this work is to present an overview of these efforts by showing the range of materials synthesized by SCS, typical radioluminescence (RL) spectra to show the incorporation of luminescence centers, as well as TL and OSL of some of the samples that exhibited high sensitivity to ionizing radiation. We also discuss the effect of Li co-doping in the RL and TL properties and some unexpected results related to background of the TL measurements and fading. This work does not intend to be an exhaustive study of any single material; see for example references [38,39]. Instead, we focus on general observations that we hope can be useful for other investigators working on the development of new TL and OSL materials.

2. Material requirements

Table 1 summarizes the most important requirements for the specific applications discussed above. In all cases, it is expected that the trapped charge population is stable at room temperature.

In personal OSL dosimetry, additional requirements include a light sensitive trapped charge population, emission in the blue-UV range of the spectrum, tissue equivalency, and predominance of single trapping centers. Emission in the blue-UV range of the spectrum allows for detection of light at shorter wavelengths than stimulation (blue or green), in addition to being a better match for the spectral response of photomultiplier tubes (PMTs). In OSL dosimetry, emission in wavelengths shorter than the stimulation wavelength makes it easier to separate between the stimulation light and the OSL emission using optical filters [2]. Tissue equivalency means that the host material has an effective atomic number similar to water or tissue ($Z_{eff}\sim 7.5$–7.6), so that the detector has a response with dependence on photon energy similar to the material of interest [17]. Predominance of a single trapping center means that that the signal is not associated with overlapping components with different dosimetric properties (e.g., thermal stability). Moreover, luminescence centers characterized by radiation transitions with long lifetime are useful because of the possibility of increasing the signal-to-noise ratio using a time-resolved luminescence technique called pulsed OSL

Table 1
Examples of desirable properties for new TL/OSL materials for different applications.

Application	Desirable properties
All	• Trapped charge population stable at room temperature
Personal OSL dosimetry	• Trapped charge population sensitive to light • Emission in the blue-UV region • Tissue equivalency ($Z_{eff}\sim 7.5$) • Single trapping center associated with the OSL signal • Long luminescence lifetime (>100 μs) in case of POSL applications • Intrinsic neutron sensitivity, i.e. having Li or B in its composition (for neutron dosimetry)
2D OSL dosimetry	• Trapped charge population sensitive to light • Short luminescence lifetime (<100 μs) • Emission in the blue-UV region • Tissue equivalency ($Z_{eff}\sim 7.5$) • Single trapping center associated with OSL signal • Small grain sizes (~μm or less)
Temperature sensing (TL)	• Multiple TL peaks over a wide range of temperatures • Simple TL kinetics (first order) • Trapped charge population insensitive to light