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The optically stimulated luminescence (OSL) properties of LiF:Mg,TI, $Li_2B_4O_7$:CU, $CaSO_4$:Tm, and CaF_2:MN thermoluminescent (TL) materials

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HIGHLIGHTS

- Optically stimulated luminescence (OSL) properties of several known thermoluminescent materials were investigated.
- CaF_2:Mn did not show an OSL response with either IR or blue light stimulation.
- $Li_2B_4O_7$:Cu and LiF:Mg,Ti demonstrated very weak OSL signals only under blue light excitation.
- $CaSO_4$:Tm exhibited OSL under both IR and blue light stimulation.
- OSL properties of $CaSO_4$:Tm indicated an optimal OSL readout regime.

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ABSTRACT

This paper reports on an investigation into the optically stimulated luminescence (OSL) properties of several known thermoluminescent materials, namely LiF:Mg,Ti, $Li_2B_4O_7$:Cu, $CaSO_4$:Tm, and CaF_2:Mn. Samples were irradiated to air doses of 15 mGy, 150 mGy and 1.5 Gy and analyzed using a commercially available OSL reader system to determine their luminescence response to continuous blue and infrared light (IR) excitation, centered at 470 nm and 830 nm wavelengths, respectively. CaF_2:Mn did not show an OSL response with either IR or blue light stimulation. $Li_2B_4O_7$:Cu and LiF:Mg,Ti demonstrated relatively weak OSL signals only under blue light excitation. $CaSO_4$:Tm exhibited OSL under both IR and blue light stimulation at sensitivities roughly one order of magnitude less than the OSL response of α-Al_2O_3:C under the same conditions.

1. Introduction

Optically stimulated luminescence (OSL) dosimetry relies upon the illumination of an irradiated sample with light to produce a stimulated emission of light proportional to the radiation dose which previously caused trapping of electrons in the material. OSL has become an accepted personnel dosimetry method during the past two decades, primarily because of the success of a commercially-available dosemeter based upon anion-defective, carbon-doped aluminum oxide, or α-Al_2O_3:C (Luxel®, Landauer, Inc., 2 Science Road, Glenwood, Illinois, 60425-1586, USA, custserv@landauer.com, +1 800 323 8830, www.landauer.com). Thermoluminescent dosemeters (TLDs) long dominated personnel dosimetry, and they are well characterized (Samei et al., 1994; Kearfott et al., 1995, 2000, 2015; Simpkins and Kearfott, 1997; Harvey et al., 2011) and have tissue-equivalent forms essential for meaningful application to medical applications (Kearfott and Grupen-Shemansky, 1990; Kearfott et al., 1990). Unfortunately, TLDs are subject to signal fading post-irradiation (Harvey et al., 2010, 2011), which is particularly significant at high ambient temperatures (Harvey and Kearfott, 2012).

The use of OSL dosemeters in place of TLDs is attractive for a number of technical reasons, including: (1) the current material of choice for this application, α-Al_2O_3:C, is highly sensitive, in that it emits a large amount of stimulated luminescence per unit of the absorbed dose, (2) the optical readout method is fast and relatively simple, and (3) the OSL technique lends itself to repeat readout of samples when an isolated optical simulation does not result in complete clearing of stored signal (Bulur and Göksu, 1997; Akselrod et al., 1998; McKeever and Akselrod 1999; Bøtter-Jensen et al., 1999). Optical stimulation can be activated and deactivated very quickly and thus allowing fast readout of the dose information without a significant depopulation of the dosimetric traps

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(Yoder, 2000). An additional reason that α-Al_2O_3:C was chosen as a dosimetry material for widespread use is that it exhibits very little post-irradiation fading at room temperature over time. Because of this, the accurate estimation of dose is not affected to a large degree by the amount of time elapsed between irradiation of the dosemeter and readout (Akselrod et al., 1998). Although OSL dosemeters are frequently susceptible to environmental light-induced fading (Moscovitch et al., 1993; Walker et al., 1996; West et al., 2006) maintaining the material in a light-tight container during use does not present a significant problem in most applications.

In addition to its application in the area of personnel dosimetry, the OSL phenomenon is widely used in the dating of geologic samples (Stoneham et al., 1996; Banerjee et al., 2000; Bailey et al., 2000; Stokes and Fattahi, 2003). It has been investigated and used for a variety of other applications, including remote dosimetry (Takaki et al., 1997), retrospective dosimetry (Inrig et al., 2008) and medical dosimetry (Akselrod et al., 2006; Gaza and McKeever 2006; Vrigneaud et al., 2013). In most of these applications, the material employed is either α-Al_2O_3:C or a natural material such as quartz, i.e. SiO_2, or feldspar, namely $KAlSi_3O_8$, $NaAlSi_3O_8$, or $CaAl_2Si_2O_8$.

Because of the substantial utility of α-Al_2O_3:C and the prevalence of OSL techniques in geologic dating, a great deal of published research has focused on α-Al_2O_3:C and natural materials, while relatively little published data is available on the OSL properties of other conventional TL materials that may also exhibit OSL. In the case of $Li_2B_4O_7$:Cu and $CaSO_4$:Tm, in spite of their use in commercially-available dosemeters, and the presence of substantial published TL data (Takenaga et al., 1983; Driscoll et al., 1983; Samei et al., 1994; Sunta et al., 1994; Kearfott et al., 1995, 2000; Simpkins and Kearfott, 1997; Lee et al., 2008; Nambi et al., 1974; Lewandowski et al., 1996; Lakshmanan et al., 2005; Yamashita et al., 1971; Harvey et al., 2010, 2011; Harvey and Kearfott, 2011a, 2011b, 2012), little published research exists documenting their OSL properties. However, $CaSO_4$:Tm has been shown to produce a signal using photo- transferred thermoluminescence techniques (PTTL) techniques (Lakshmanan et al., 2005). PTTL is the measurement of a TL signal originating from shallow traps following the optical transfer of charge from deep traps to the shallow traps. Its presence in a TL material indicates that the material has an optically sensitive trap clearance mechanism and therefore may also exhibit OSL under appropriate excitation and detection conditions. $CaSO_4$:Tm is also known to be quite sensitive to environmental light-induced fading (Pradhan, 1993), which suggests that its optical ionization cross section is high and therefore a good candidate for OSL.

In the case of LiF:Mg,Ti, there have been published confirmations of an optically stimulated afterglow (OSA), along with a characterization of the excitation and emission spectra relating to this afterglow (Jaek et al., 2002; Matsushima et al., 2013). However, few data exist related to its OSL dose sensitivity or behavior in a conventional continuous-wave OSL (CW-OSL) measurement regime. In the case of CaF_2:Mn, there is a limited amount of published OSL research. Additionally, Henniger et al. (1982) published the results of an experiment using broadband optical illumination and a delayed OSL (DOSL) technique. Allen and McKeever (1990) have performed some PTTL and OSL experiments, reporting OSL excitation spectra for a single emission wavelength of 495 nm but with no sensitivity information. Investigations of the material's sensitivity, annealing and long-term OSL fading behaviors using a highly specialized 'cooled' OSL technique have also been published (Miller and Eschbach, 1991).

This experiment seeks to expand upon this research. Specifically, it was designed to determine whether LiF:Mg,Ti, $Li_2B_4O_7$:Cu, $CaSO_4$:Tm, and CaF_2:Mn exhibit OSL when using narrow-band IR and blue light excitation in a room-temperature CW-OSL regime, and to what relative degree. Because the defined experimental setup, which stimulated with two wavelengths of light and detected in the ultraviolet (UV) region, does not examine the entire spectrum of possible excitation and emission wavelengths. This experiment is intended as an initial investigation into the promise of several of these materials as OSL dosemeters.

2. Materials

The four commercially available TLD crystal types evaluated as part of this experiment were LiF:Mg,Ti (TLD-100, Harshaw Chemical Company, 29001 Solon Road, Cleveland, OH, 44139, USA), $Li_2B_4O_7$:Cu (Panasonic Industrial Company, 3 Panasonic Way, 7E-6, Secaucus, NJ 07094, USA, radiationmeasurement@us.panasonic.com, +1 201 348 2517, www.panasonic.com), $CaSO_4$:Tm (Panasonic Industrial Company, 3 Panasonic Way, 7E-6, Secaucus, NJ 07094, USA, radiationmeasurement@us.panasonic.com, +1 201 348 2517, www.panasonic.com), and CaF_2:Mn (TLD-400, Harshaw Chemical Company, 29001 Solon Road, Cleveland, OH 44139, USA).

The LiF:Mg,Ti and CaF_2:Mn were in the form of 3.2 mmx3.2 mmx0.6 mm chips. The $Li_2B_4O_7$:Cu and $CaSO_4$:Tm material was originally in the form of small grains attached with an epoxy to dosemeter badge backings (UD-802AQ TLD badges, Panasonic Industrial Company, 3 Panasonic Way, 7E-6, Secaucus, NJ 07094, USA, radiationmeasurement@us.panasonic.com, +1 201 348 2517, www.panasonic.com). The materials were carefully scraped from their respective backings and placed on 9.7 mm diameter stainless steel planchets with a non-fluorescing adhesive. This adhesive was verified as non-fluorescing by confirming that blank samples, containing only adhesive on the planchet, produced no detectable OSL output during the course of the experiment. The grains covered a circular surface area roughly the size of the chips. Samples of α-Al_2O_3:C (Luxel®, Landauer, Inc., 2 Science Road, Glenwood, Illinois, 60425-1586, USA, custserv@landauer.com, +1 800 323 8830, www.landauer.com) were used as reference standards. Individual samples were formed using a hole punch to produce 6.35 mm diameter round samples from the clear polystyrene-laminated α-Al_2O_3:C ribbon. All samples were washed with ethyl alcohol, C_2H_5OH, to remove any potentially fluorescent contaminants before use.

3. Methods

A flexible, commercially available automated OSL/TLD reader system was employed for the experiment (Model DA-15, Risø National Laboratory, Frederiksborgvej 399, P.O. Box. 49, 4000 Roskilde, Denmark, risoe@risoe.dtu.dk, +45 4677 4677, www.risoe.dtu.dk). This reader allows continuous blue light stimulation centered at 470 nm using clusters of 42 light emitting diodes (LEDs) (NSPB 500S, Nichia Corporation, 491 Oka, Kaminaka-Cho, Anan-Shi, Tokushima 774-8601, Japan, +81 3 3456 3746, www.nichia.co.jp) collectively delivering 50 mW cm^{-2}. In addition, this unit features infrared (IR) stimulation capability, centered at 830 nm, using a 1 W IR laser diode delivering 300 mW cm^{-2}. The reader also had an integral beta irradiator employing a pneumatically activated 1.48 GBq $^{90}Sr/^{90}Y$ source, capable of delivering approximately 15 mGy s^{-1} to a sample. The detection subsystem of this reader consists of a low-background bi-alkali photomultiplier tube with a quartz window (Model 9235QB, ADIT Electron Tubes, 300 Crane Street, Sweetwater, Texas 79556, USA, sales@electrontubes.com, +1 325 235 1418, www.electrontubes.com) operated in photon counting mode. High-pass filters (U340, Hoya Filters, 2-7-5, Naka-Ochiai, Shinjuku-ku, Tokyo 161-8525,