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Optically stimulated luminescence: A brief overview

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ABSTRACT

The use of optically stimulated luminescence (OSL) in radiation dosimetry has been conditioned by the availability of suitable dosimetry (OSLD) materials. The crucial property dictating the suitability of a material as an OSLD is the material's defect structure. This paper reviews some of the recent developments in radiation dosimetry that have been enabled by knowledge of the charge trapping and recombination processes occurring in OSLD materials during irradiation and stimulation. Although many materials have been and are currently being studied, this short review focuses on just two, namely carbon-doped aluminum oxide (Al_2O_3:C) and europium-doped potassium bromide (KBr:Eu). The defect structure and trapping/recombination mechanisms in these materials have led to application in several areas of radiation dosimetry and dose imaging. Two recent areas of development are in space dosimetry (for Al_2O_3:C) and in medical dosimetry (for KBr:Eu). This overview briefly describes these latter modern applications and relates the functionality of the OSLDs to their basic defect properties.

1. Introduction

The genesis of the use of optically stimulated luminescence (OSL) and thermoluminescence (TL) in radiation dosimetry can be found in some of the earliest scientific literature, at a time when consensus about the "scientific method" was just emerging. Sir Robert Boyle's famous treatise on color contains a description of a "glimmering light" from a piece of natural diamond when heated (Boyle, 1664). Today this phenomenon is known throughout our community as thermoluminescence and, although Boyle's experiments predated the discovery of radiation by more than two centuries, the use of TL in radiation dosimetry is now firmly established. What is perhaps less well known is that the origins of the phenomenon that we now label optically stimulated luminescence can be found in the 19th-century French scientific literature, due to the combined work of Edmond and Henri Becquerel (Becquerel, 1843, 1883). The subject of the Becquerels' studies was phosphorescence and on several occasions they described how this form of luminescence can either be increased or decreased (quenched) by illuminating an irradiated sample (infra-red phosphor) with visible light. These latter studies appeared at the beginnings of science's fascination with radiation and its effects but before the birth of quantum mechanics and thus an understanding of electron and hole trapping and recombination was certainly not available. Even so, one has to admire the notions put forward by Weidemann and Schmidt who, as early as 1895, talked about their 'electric dissociation' theory involving the separation of positive and negative charges as being the origin of the phenomenon that they christened "thermoluminescenz" (Wiedemann and Schmidt, 1895). This appears to be the first published use of the modern word. However, we had to wait until the 1960s to see the first use of the phrase "optically stimulated luminescence" when it was used by Fowler in a discussion of luminescence radiation dosimetry methods in medicine (Fowler, 1963).

Developments over the past two or three decades in OSL and TL dosimetry have led to the application of these techniques in many radiation dosimetry fields, including personal, environmental, retrospective, space, neutron and medical dosimetry. Not all of these applications can be discussed sensibly in one short review. Instead the focus of this article is to demonstrate how a fundamental understanding of the charge trapping and recombination processes in Al_2O_3:C and KBr:Eu, at both phenomenological and structural levels, has lead to confidence in the use of these techniques in two particular applications, namely space and medical dosimetry.

2. OSL materials

Carbon-doped aluminum oxide was developed initially as a TL dosimeter in pioneering work at the Urals Polytechnical Institute in Russia (Akselrod et al., 1993, 1990). The extreme sensitivity of the

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material to radiation (i.e. a large TL signal for a small absorbed dose) made this material very attractive for use as a radiation dosimeter. The high sensitivity was due to the creation of a large concentration of oxygen vacancy centers in the material during the crystal growth process. The oxygen vacancy centers (*F*- and F^+-centers) acted as recombination sites yielding a bright emission. The source of the trapping sites was not known. A potential difficulty with the material was its sensitivity to visible light, giving rise to light-induced fading of the TL signal (Moscovitch et al., 1993). By taking advantage of this apparent disadvantage, however, the material could be made into an efficient OSL dosimeter by using light rather than heat to stimulate the radiation-induced luminescence signal (Akselrod et al., 1998; McKeever, 2001; McKeever and Akselrod, 1999). Since then, Al_2O_3:C has gone on to become the foremost OSL dosimetry material, seeing use in a multitude of dosimetry applications.

OSL from europium-doped alkali halides was first used as a technique in medical imaging (Douguchi et al., 1999; Nanto et al., 1993). The sensitivity of the OSL from this material is due to excitation and relaxation of Eu^{2+} ions. More recently, the very rapid decay of the OSL from irradiated KBr:Eu raised the potential of this material being used in real-time dosimetry during medical radiodiagnostic and radiotherapeutic procedures (Gaza and McKeever, 2006; Klein and McKeever, 2008; Klein et al., 2006). Development in these applications continues.

In each of the above materials, the key to successful application lies in an understanding of the defect structure and the electron-hole trapping and recombination processes, even though many details are still lacking. Although a significant amount is known about the emission centers in both materials, the identity of the traps is still unknown. Nevertheless, even a phenomenological understanding of the OSL process can lead to useful insight and confidence in the application.

2.1. Al_2O_3:C

When grown in a reducing atmosphere Al_2O_3 single crystals can be produced with high concentrations of *F*, F^+, F_2 and F_2^+ oxygen vacancy centers giving the material a high OSL sensitivity (Akselrod et al., 1993; Itou et al., 2009; Yang et al., 2009). The main emission from such material is centered near 420 nm due to relaxation of the 3P state to the 1S state of the *F*-centers, with a luminescence lifetime of ~35 ms. An additional emission band is observed near 330 nm due to the de-excitation of the 1B state to the 1A state of F^+-centers, with a much shorter lifetime <7 ns (Yukihara and McKeever, 2006). Weaker emission can be observed at 500 nm due to F_2-centers (Itou et al., 2009). After irradiation a broad OSL excitation band is generated between approximately 600 nm to less than 260 nm, with no apparent peak (Whitley and McKeever, 2000). The broad excitation band is suggestive of multiple traps being emptied by the stimulation light in this wavelength range and this suggestion is supported by OSL (Whitley and McKeever, 2000), thermally stimulated conductivity (TSC) (Whitley et al., 2002) and TL analyses (Akselrod and Akselrod, 2002), all of which demonstrate the existence of many trapping centers for both electrons and holes.

In addition to the pre-existing concentrations of *F*- and F^+-centers, further such centers are induced by irradiation. The radiation-induced populations undergo interconversions as *F*-centers are converted to F^+-centers and vice-versa during heating (and presumably during optical stimulation). These population changes are coincident with the emission of TL (or OSL; Fig. 1), which is caused by either electron recombination with holes at F^+-centers yielding 420 nm emission, or hole recombination at *F*-centers yielding 330 nm emission. The multiplicity of trapping and emission centers leads to complex and sample-dependent TL and OSL characteristics. It is this very complexity that can be exploited to advantage in certain

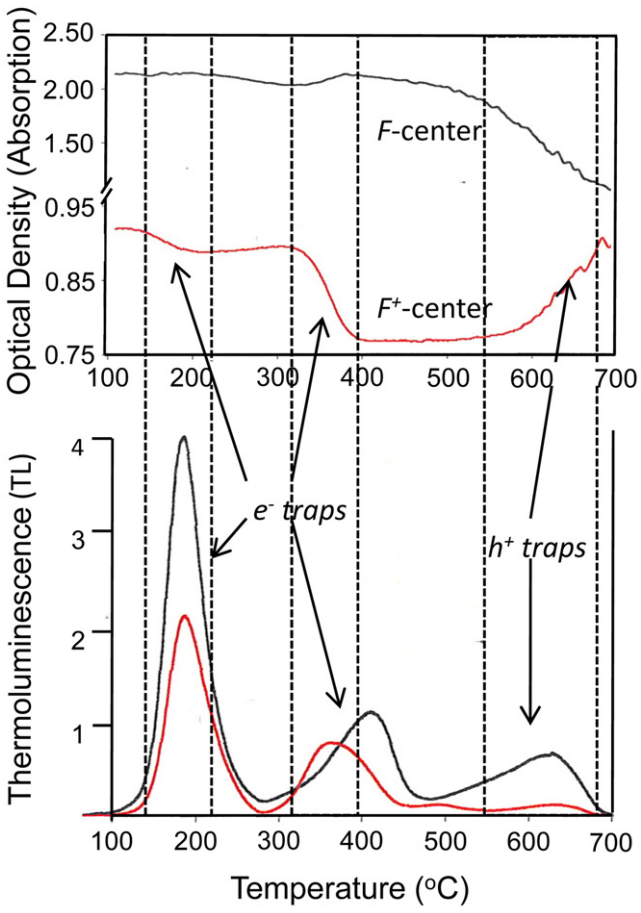

Fig. 1. Changes in the *F*- and F^+-center concentrations (top) in Al_2O_3:C as a function of heating after irradiation (1 kGy ^{60}Co gamma at room temperature) followed by heating at a rate of 1 °C/s. The optical densities were monitored at 205 nm (*F*) and 255 nm (F^+), with a spectral band width of 2 nm. Changes in the background were subtracted from each curve. Thermoluminescence (bottom) recorded at 0.33 °C/s for two separate samples of Al_2O_3:C following irradiation. The steps in the color center data are approximately correlated with the TL peaks and the *F*- and F^+-data are anti-correlated. The downward steps in the F^+-center data indicate the release of electrons from traps and their subsequent recombination with F^+-centers to form *F*-centers. Likewise, the upward step in the F^+-center data and corresponding downward step in the *F*-center data, suggest the release of trapped holes. Unpublished data, courtesy of Dr. J.C. Polf; samples provided by Dr. M. S. Akselrod.

dosimetry applications, as discussed in Section 3. Nevertheless, for the moment we highlight the fact that the presence of shallow electron traps can lead to a thermal instability in the OSL sensitivity that needs to be eliminated. Thus, great effort is expended in growing OSL crystals without shallow traps (Akselrod et al., 1998). Deep traps are of both polarities, electrons and holes. Changes in the populations of these centers can result in sensitivity increases or decreases, and supralinearity or linearity in the dose response curves. Solutions to rate equations for complex kinetic models, which include a plethora of traps (electron and hole, deep and shallow) with pre-existing recombination sites, lead to an acceptable prediction of behaviors of the different Al_2O_3 samples that have been described in the literature (Pagonis et al., 2007; Yukihara et al., 2003). Thus, the broad features of the OSL from Al_2O_3:C are documented and understood.

2.2. KBr:Eu

Potassium bromide doped with divalent europium exhibits OSL emission at ~420 nm due to the $4f^65d$-$4f^7$ transition in Eu^{2+} ions. The OSL stimulation maximum is at 620 nm corresponding to