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Latest developments in silica-based thermoluminescence spectrometry and dosimetry

D.A. Bradley [a,b,*], S.M. Jafari [a,c], A.S. Siti Shafiqah [d], N. Tamcheck [e], A. Shutt [f], Z. Siti Rozaila [b], S.F. Abdul Sani [b], Siti Norbaini Sabtu [b], Abdulaziz Alanazi [a], G. Amouzad Mahdiraji [g], H.A. Abdul Rashid [h], M.J. Maah [i]

[a] Department of Physics, University of Surrey, Guildford, Surrey GU2 7XH, UK
[b] Department of Physics, University of Malaya, 50603 Kuala Lumpur, Malaysia
[c] Radiotherapy Physics, Level B, Queen Alexandra Hospital, Portsmouth PO6 3LY, UK
[d] Department of Physics, Kuliyah of Science, International Islamic University Malaysia, 25200 Kuantan, Malaysia
[e] Department of Physics, Universiti Putra Malaysia, 43400 Serdang, Selangor, Malaysia
[f] Public Health England, CRCE, Chilton, Didcot, Oxon OX11 0RQ, UK
[g] Integrated Lightwave Research Group, Department of Electrical Engineering, Faculty of Engineering, University of Malaya, 50603 Kuala Lumpur, Malaysia
[h] Faculty of Engineering, Multimedia University, 2010 Cyberjaya, Selangor, Malaysia
[i] Department of Chemistry, University of Malaya, 50603 Kuala Lumpur, Malaysia

HIGHLIGHTS

- We provide a status report for our work on silica-doped media for TL dosimetry.
- The construction and performance of a TL emission spectra facility is described.
- Oxygen-rich/oxygen deficient Ge-doped SiO_2 defects/lattice relaxation phenomena are related to TL spectra.
- Ge-doped SiO_2 are used to obtain high spatial-resolution Am–Be neutron source measurements.

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ABSTRACT

Using irradiated doped-silica preforms from which fibres for thermoluminescence dosimetry applications can be fabricated we have carried out a range of luminescence studies, the TL yield of the fibre systems offering many advantages over conventional passive dosimetry types. In this paper we investigate such media, showing emission spectra for irradiated preforms and the TL response of glass beads following irradiation to an ^{241}Am–Be neutron source located in a tank of water, the glass fibres and beads offering the advantage of being able to be placed directly into liquid. The outcomes from these and other lines of research are intended to inform development of doped silica radiation dosimeters of versatile utility, extending from environmental evaluations through to clinical and industrial applications.

1. Introduction

Over the past three years the consortium effort that this work represents has focused on developing small diameter (0.1 mm through to ~1 mm) GeO_2 doped SiO_2 glass fibres for luminescence dosimetry. For the same reasons, we have also investigated commercial jewellery beads of various diameters (1–3 mm), with the additional benefit of reduced cost and robust utility. Both media are silica based, with compositional studies being carried out to locate and quantify dopants within the silica; see for instance the elemental analysis studies of beads, conducted by Jafari et al. (2014a). The intention has been to produce high spatial resolution thermoluminescence dosimeters (TL) for sensitive radiation detection.

For the preforms from which the fibres are produced, using a custom-built spectrometer we show first results for emission wavelengths, revealing the predominant wavelengths to be towards the blue end of the visible spectrum. In respect of TL yield, we are seeking to define optimal concentrations of GeO_2 in SiO_2 fibres, to-date the dopant concentration being incremented from

* Corresponding author at: Department of Physics, University of Surrey, Guildford, Surrey GU2 7XH, UK.
E-mail address: d.a.bradley@surrey.ac.uk (D.A. Bradley).

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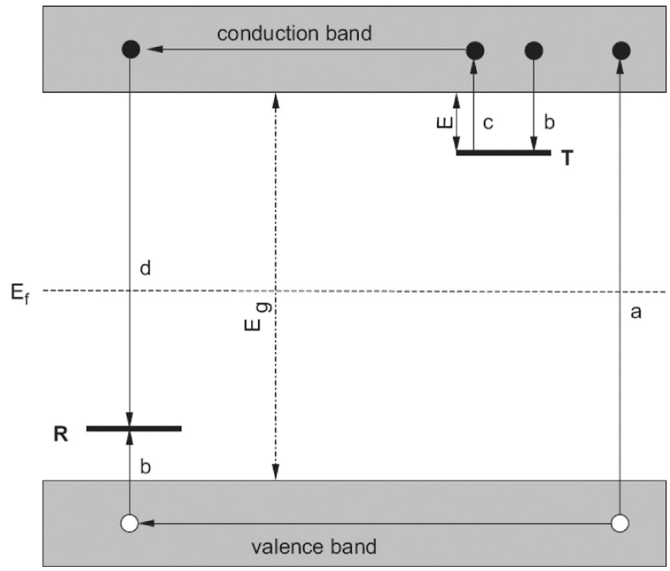

Fig. 1. Energy band model showing the electronic transitions in a TL material according to a simple two-level model. (a) Generation of the electrons and holes; (b) electron-hole trapping; (c) electron release due to thermal stimulation; (d) recombination. The solid circles represent electrons, the open circles holes. Level *T* is an electron trap, level R is a recombination centre, E_f is the Fermi level, E_g is the energy band gap and *E* is the activation energy (referred to in the present text as E_c). Taken from Bos et al. (2007).

that of standard telecommunication fibres (~4 mol%) up to some 12 mol%. At low dopant concentrations and minimal shape deformity the short-range SiO_2 network suffers minimal strain. Conversely, the interface between the pure SiO_2 cladding and the GeO_2 doped silica network becoming increasingly strained with increase in extrinsic doping and shape deformation. As reported elsewhere (Abdul Sani et al., 2015a, 2015b), elevations of Ge dopant beyond 4 mol % tend to reduce TL yield, a result of concentration quenching, sometimes also referred to as self-absorption. The predominant focus of present work is on investigation of the preforms from which the fibres have been formed.

Using a standard TL reader, measurements of TL from an irradiated sample typically concern the emitted total light intensity, the emission resulting from insulating/semiconductor material containing defects, intrinsic and extrinsic. With present interest being on Ge-doped silica preforms, attempts are made to measure changes in emission resulting from changes in defect concentrations. Our starting point in studying the TL glow curve is use of the standard energy level description for defects in such media (Fig.1). Although described for crystalline media, the model is also applicable to the short-range order of amorphous media, including the silica studied herein (Bradley et al., 2014). The band model of Fig. 1 is typically represented by E_c, the activation/conduction energy, also referred to as the conduction energy, pointing to the least energy needed to drive de-trapping, E_v the valence band energy and E_f the Fermi energy.

2. Energy band diagram

Optical activity concerns the absorption and luminescence process. In the glass-matrix lattice, the extrinsically introduced Ge atoms occupy interstitial or substitutional positions, the modification in bond energy allowing the donor electrons to move freely from ion to ion. At ~9 eV, the energy difference between the valence and conduction band of silica is wide, as shown in Fig. 2. When light energy is absorbed and if the absorbed energy is sufficiently great, then electrons may be raised from ground

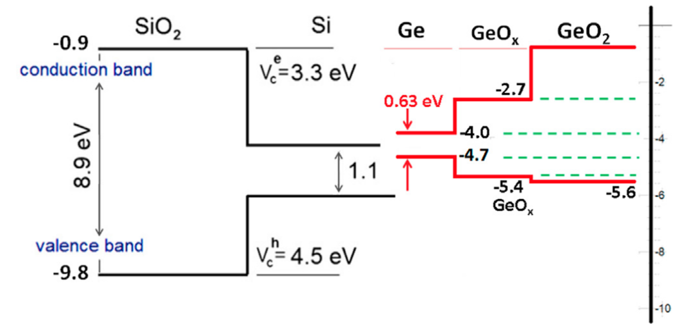

Fig. 2. Modification of the energy band structure of silica by impurity atoms (Ge) or lattice irregularities in the silica glass that create localized electron states (energy values all in eV). This figure was constructed with the help of available literature (Randall, 1945; Skuja, 1992, 1998; Imai and Hirashim, 1994; Amossov and Rybaltovsky, 1994), showing the various possible defect energy levels in Ge doped silica.

energy to the conduction band, giving rise to a current. At intermediate energies, the electrons can be trapped by impurity atoms (Ge in the present case) or lattice irregularities in the silica glass that create localized electron states. These electron states normally have very narrow energy level and are sometimes capable of trapping electrons for long durations (from seconds to years). The release of this energy in the form of photons are categorized in terms of typical time scales, as in fluorescence ($\sim 10^{-7}$ s) and phosphorescence (10^2 s). Our main interest is in the photon emitted by the phosphorescence process, utilized in many applications. The localized energy states created by impurity atoms have been the focus of many studies, detailed by among others, Randall (1945), Skuja (1992, 1998), Imai and Hirashim (1994) and Amossov and Rybaltovsky (1994), the outcome of these providing for a broad summary of the energy levels for Ge-doped silica glass (Fig. 2) and the signature absorption and luminescence bands or colour centres (Table 1).

3. Energy level of GeO_2–SiO_2 glass

In the extrinsic doping of an insulator or semiconductor the point defects are produced as a mixture of vacancies (between neighbouring atoms) and interstitials (with other atoms placed between neighbouring lattice atoms). These will distort the structure plane, altering compactness and local charge and strain. The point defects produce colour centres as a result of the modification of electronic neutrality, line defects are from dislocations

Table 1
Optical absorption and luminescence band defect centres in silica (Skuja, 1998)*

Defect acronym(s)	Suggested-structural model(s)	Position-of optical absorption band peaks (eV)	Peaks of PL bands (eV)
Si ODC(II), Ge ODC (II)	Oxygen vacancy	6.8–7.0	4.2–4.3
B_2(Si),	Oxygen divacancy	4.95–5.05,	4.3–4.4
B_2(Ge)	Oxygen divacancy	5.1–5.4	4.3–4.4
Si_2^0	dicoordinated Si/Ge	3.15	2.7–2.8
Ge_2^0, Ge^{2+}	dicoordinated Ge	3.7	3.0–3.2
Non-bridging-oxygen hole centre (NBOHC)	≡Si–O●	4.8	1.85–1.95

* Skuja has commented on the continuing controversy over the basic structure of ODCs (oxygen deficient centres), with different naming conventions for ODCs as a result of this, including 'non-relaxed oxygen vacancy', 'defect of type oxygen vacancy', 'divacancy', 'B2-centre' and 'silicon lone-pair center' (SLPC) for ODC(II). PL indicates photoluminescence.

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