J. Phys. D: Appl. Phys. 22 (1989) 1758–1762. Printed in the UK

An optical fibre rereadable radiation dosimeter for use at high doses and at elevated temperature

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Received 3 May 1989, in final form 30 June 1989

Abstract. A new type of radiation dosimeter for large radiation doses is described, which is based on silica fibre material. Conventional radioluminescence or thermoluminescence of silica produces emission in the blue region of the spectrum. However, in this new material irradiation, in conjunction with a heat treatment, generates a green emission band. The intensity of the green band can be monitored by either radioluminescence or thermoluminescence using a test dose. The signals are directly related to the total irradiation history of the material. The dosimeter is therefore rereadable. The production mechanism of the green emission centre requires a thermal processing stage, with an activation energy of 0.52 eV. Further, the dosimeter is effective at recording radiation during high-temperature exposure, to at least 400 °C, with the subsequent dosimetry being performed below 200 °C.

1. Introduction

Numerous materials have been considered as potential thermoluminescence (TL) radiation dosimeters because most large-energy-gap insulators have deep trapping levels which can store electrons, or holes, generated by ionising irradiation. Subsequent thermally stimulated release of the metastable charges may then produce luminescence. Key properties sought for TL dosimeters are a reproducible signal–dose response (preferably linear), emission within the spectral range of a low-noise photomultiplier, and more importantly, trap depths which empty in the temperature range from 150 to 250 °C. This ideal depth range ensures that the dosimeter is capable of long term storage at ambient temperature but a measurement heating run is not perturbed by detection of thermal emission.

The metastable character of the process implies that there is no guarantee that the TL information may be read a second time. However, for several of the more common personnel dosimeters photostimulated TL is feasible (e.g. Jain 1984, McKeever 1985) but the sensitivity of the rereading process is greatly reduced, to say 10%, even for favourable dosimeters such as LiF or BeO. At higher dose levels than say 10^3 Gy the materials either saturate or show non-linearities in response to dose. This is unimportant for personnel applications but it has the consequence that for high-dose applications the number of rereadable dosimeters is greatly reduced.

Further, if the glow peaks appear in the range 150 to 250 °C then irradiation at higher temperatures can only be seen from a regenerated signal. In practice high-temperature irradiations frequently lead to annealing of the defect structures which give the conventional TL signals.

The dosimeter material described here is suitable for large irradiation doses and has the advantages of being rereadable, capable of recording doses received at temperatures up to 400 °C and provides an integration of the irradiation history.

2. Experimental details

The material tested was in the form of a step index optical fibre of 125 μm outer diameter doped with a low concentration of rare earth ions and was prepared by a solution doping method (Townsend *et al* 1987, Stone and Burrus 1973).

Irradiations were made with either a $^{90}Sr/^{90}Y$ source or an x-ray set operated at 30 to 35 kVp. The dose rates in the material were estimated to be 18 Gy min^{-1} for the beta source and approximately 3 Gy min^{-1} for

0022-3727/89/111758 + 05 $02.50

the x-ray tube, using either LiF or obsidian TL calibrations against an NPL source. The signal produced during x-ray irradiation, i.e. radioluminescence, was recorded. Thermoluminescence runs were variously made at heating rates of 20 to 100 °C min^{-1}. For those runs in which the spectra of the TL were recorded the lowest heating rate was used. Spectra were recorded via a scanning monochromator, a cooled 9659QA EMI photomultiplier tube and a small computer to record the data. Corrections have been made for the wavelength response of the system. In most cases the spectra were recorded with a wavelength resolution of 10 or 20 nm.

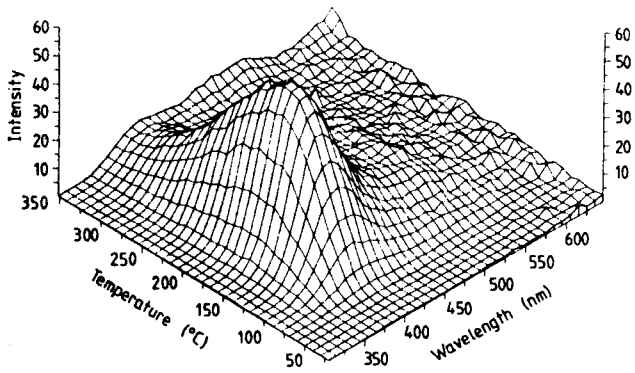

Figure 1. The initial TL spectra from a germano–silica fibre type ND326.

3. Results

The visible spectra obtained during initial radioluminescence (RL) or thermoluminescence (TL) of silica fibres recorded in this work closely resemble those reported elsewhere for bulk silica, silica fibres and indeed for many quartz samples (David *et al* 1977, 1978, McKeever 1984, West and Carter 1980, Yokota 1953). In the fibre work there is a dominant TL peak near 175 °C. Both RL and TL emit strongly in the blue near 400 nm. Although a wide range of doped fibres have been used this blue emission is normally the only major signal in the range from 300 to 750 nm. The intensity of the RL is initially non-zero and increases with exposure to ionising radiation, but with evidence for saturation at high-dose levels. The blue TL signal is nearly linearly related to the room-temperature irradiation dose. Such features are consistent with earlier data for silica.

An advantage of using optical fibre material for a luminescence study is that one has a high-purity glass matrix in which the dopant level can be well controlled. The advantage of this is shown by the fibre labelled type ND326. This particular example is a germano–silicate core fibre containing low concentrations of Nd^{3+}. The dopant level of ND326 is 4 mol% GeO_2, 450 PPM Nd^{3+} and an estimated 2 PPM of OH^- in the fibre core. As shown below this composition leads to quite valuable and novel properties.

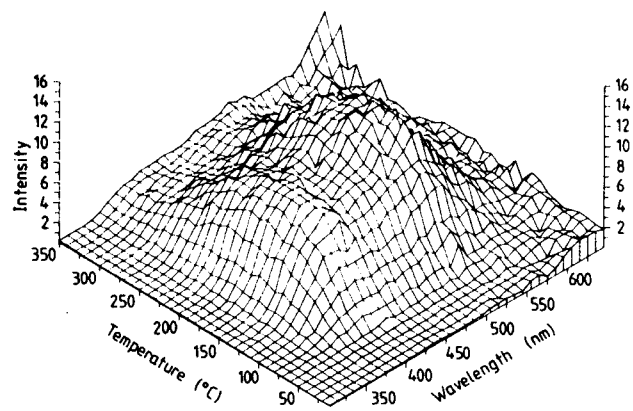

Figure 2. An example of the spectra which occur in subsequent TL remeasurements.

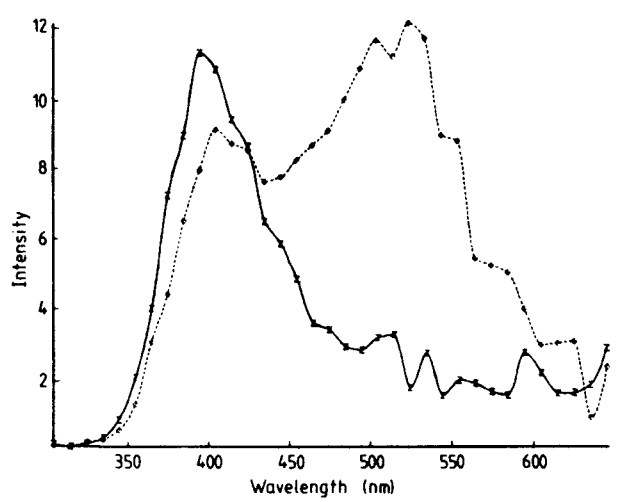

Figure 3. Examples of initial (——) and subsequent (· · · ·) emission spectra taken at 175 °C from figures 1 and 2.

3.1. Fibre type ND326

In initial measurements of RL and TL this fibre type produced the characteristic blue emission and glow peak associated with other silica type fibres. The more exciting and novel features only emerged on subsequent re-irradiation of the material after it had received both an irradiation (e.g. greater than 1 kGy) and a heat treatment (e.g. to 400 °C). Figure 1 presents an isometric plot $I(\lambda, T)$ of intensity as a function of wavelength taken during TL. The standard blue emission is clearly visible near 400 nm and the broad TL peak is a maximum near 175 °C. This is not a single glow peak but it can be resolved into several overlapping component features. By contrast with this behaviour subsequent RL or TL spectra contain a second major feature in the green (530 nm). This is demonstrated by figure 2. For comparison purposes figure 3 presents examples of spectra obtained from the isometric data for an initial and a later rerun of the TL.

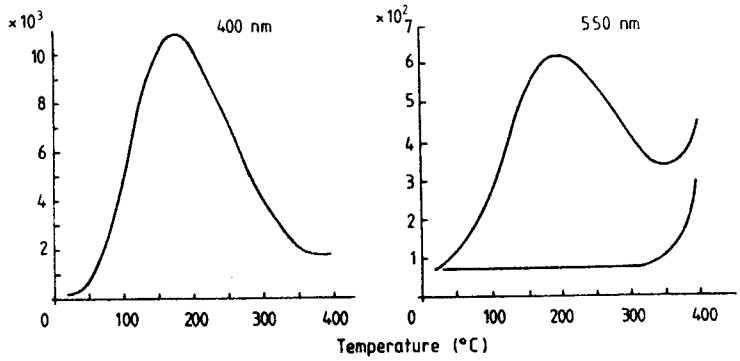

Figure 4. Examples of conventional glow curves taken at wavelengths of 400 and 550 nm with a band width of 20 nm and a heating rate of 20 °C min^{-1}. The blackbody signal at 550 nm is also shown.

Conventional TL glow curves can similarly be extracted from the $I(\lambda, T)$ data, or by operation of the monochromator at fixed wavelength. Examples of these classical-style glow curves are given in figure 4 at two wavelengths. Because there is overlap of the blue and green emission bands the green signal was monitored at 550 nm to minimise the overlap from the blue feature. It is apparent that the glow curves are not identical for the blue and green emission. There is a small shift to higher temperature and more high-temperature components in the case of the green signal compared with that in the blue. The differences are revealed in more detail in the kinetic analysis study of this material by Kirsh *et al* (1989). In broad terms both TL curves comprise a set of glow peaks originating from traps whose depths range from 0.86 to 1.72 eV.

3.2. Intensity of the green emission

During radioluminescence at room temperature the blue signal increases with radiation dose and only a weak green signal is recorded. If the material is subjected to the heating cycle of the TL measurement, that is from 20 to 400 °C, the subsequent radioluminescence signals in the blue region are regenerated as for the original measurement. In other words, from the viewpoint of the blue emission, the 400 °C heating apparently anneals the fibres. By contrast, the behaviour of the green band is markedly different. After an irradiation and heating the subsequent green emission is increased in intensity in both the RL and TL. This green signal enhancement is related to the total radiation dose received by the fibre prior to the last heating cycle. Figure 5 shows the green TL response of the fibre to a test dose as a function of radiation history at room temperature prior to the last heating cycle. The data of figure 5 were obtained using the $^{90}Sr/^{90}Y$ source with a test dose of 18 Gy. Similar results were found with the x-ray source. Larger test doses gave proportionally larger green signals. In the present trials the linearity was tested for test doses ranging from 10 to 180 Gy for a prior dose of 360 Gy and for samples in which the intervening heating cycle extended to 450 °C.

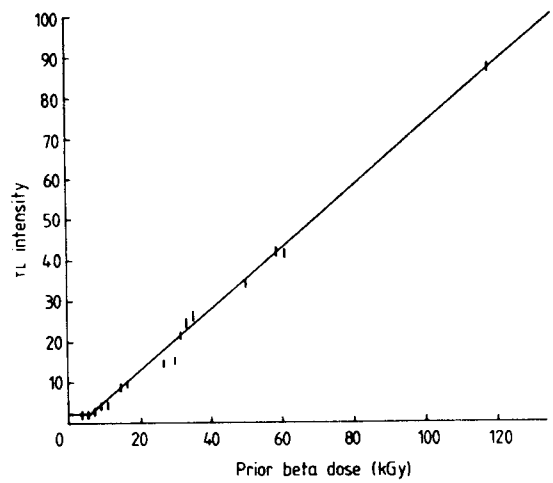

Figure 5. The signal intensity monitored in the green for an 18 Gy test dose as a function of prior irradiation dose.

3.3. Effects of heat treatment

As emphasised above, the production of the green emission required that after room temperature irradiation the sample should be heated. There appears to be a threshold temperature of 320 °C for production of the green signal. The process is thermally activated as in a sequence of irradiations and heatings to successively higher temperatures the enhancement of the green band is described by an Arrhenius plot over the range 320 to 450 °C with an activation energy of 0.52 ± 0.05 eV, as shown in figure 6.

3.4. High-temperature irradiation

Since both an irradiation and a thermal treatment are required to generate the green emission centre the two steps may be combined by irradiation at elevated temperature. This effect was tested up to 400 °C. After the higher-temperature irradiations the subsequent glow curves showed minor changes in form, when detecting in the green region.

4. Discussion

The type of Nd^{3+}-doped silica fibre reported here has two major advantages as a high-dose radiation dosimeter. These are that it is rereadable by either RL or TL, and it is an effective dosimeter for ionising radiation even if the radiation is made at high temperature. Both properties are technologically interesting. At the radiation doses used to emphasise the green emission the fibre is unsuitable for personnel dosimetry but it is of potential value for use with food processing or in fusion and nuclear reactor environments where high radiation levels and high temperatures may exist.

Discussion of the precise nature of the defect sites is difficult and, although there are many examples of thermal, or prior irradiation, sensitisation of TL dosimeters no universal mechanism has been established. Similarly, changes in spectra have been noted and, perhaps fortuitously, may correspond to the onset of supralinearity or saturation of the TL response. Arguments advanced include close association of traps and luminescence recombination centres and/or aggregation of point defects. Numerous examples of all these features are presented in the books or reviews by Aitken (1985), Jain (1984) and McKeever (1985). Articles which have specific references to quartz and silica include those by Aitken (1985), Chen *et al* (1988), David (1977, 1978), Griscom (1984), Halliburton (1985), McKeever (1985) and Townsend and Kirsh (1989). For the present work, defect aggregation may be important for the growth of the green emission centre as a thermally activated process is involved, as shown by figure 6. The small induction stage noted at lower doses and typified by the initial points of figure 5 is consistent with an aggregation model. Thus aggregation to give a green emission centre requires thermally activated motion of point defects generated by the radiation. However, once aggregation has occurred the new defects are thermally stable. This offers the possibility of green emission during subsequent RL and TL which is proportional in intensity to the size of the test dose. An aggregation model is also compatible with the fact that irradiation at high temperature can lead to the green emission from the fibres.

In silica and quartz there are numerous examples of electron paramagnetic resonance active defect sites which increase as a result of annealing but none which match the present fibre example in terms of activation energy, except that H_2O diffuses in the relevant temperature range of 300 to 400 °C (Griscom 1985). Similarly there is an extensive literature of measurements of the emission spectra taken during various types of excitation. In particular the blue emission region has been postulated as being the result of the decay of a relaxed exciton, or electron–hole pair. Specific reference to green luminescence is more limited for 'pure' material although there are examples with natural quartz. One example which does provide both a green and a blue TL signal is reported after ion beam damage of quartz. However, in this example the original 'pure' quartz gave the green signal and the amorphised (i.e. silica) region produced the blue light (Townsend 1987).

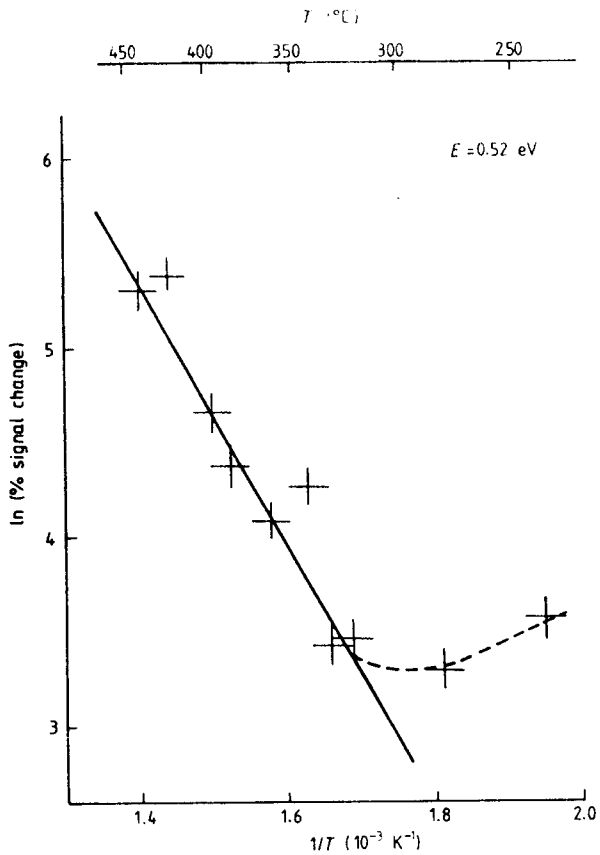

Figure 6. The efficiency for the enhancement of the green emission band, for a constant prior irradiation and test doses, as a function of the maximum temperature used in the heating cycle between irradiation and TL testing.

The TL literature of SiO_2 has been well reviewed by McKeever (1984), Halliburton (1985) and Griscom (1984, 1985) but, despite the number of papers, there are no unequivocal models for the defects. It is generally agreed that above room temperature the TL results from electron release. Many features, such as the ubiquitous '100 °C' peak (or 85 °C at the present 20° min^{-1} heating rate) are now linked to an intrinsic defect complex with Al and OH impurities. Indeed in 'superpure' quartz from GEC the 100 °C peak is reduced by 10^4 compared with standard pure quartz although individually the Al and OH levels are only reduced by 10^2 (Townsend 1986). In this fibre study there is no obvious peak at 85 °C, which is consistent with efficient removal from the fibre preform. The higher-temperature peak near 150 °C (125 °C in this case) may represent a defect complex of simple point defects, as it can be enhanced by the high-defect density found in the region of collision damage at the end of an ion beam track (Arnold 1982).

Sensitivity changes resulting from prior irradiation history are well documented for quartz and at low dose levels below 2 Gy have been used in archaeological dating (David *et al* 1977, 1978, Aitken 1985). A similar

enhancement of sensitivity is produced by heat treatment above 450 °C (McKeever *et al* 1983, Chen *et al* 1988). However in none of these previous examples has a new recombination centre appeared, nor has such a definitive need for both irradiation and subsequent heat treatment been apparent. Thus in summary it is not yet appropriate to enter into more detailed speculation on the structure of the stable new 'green' recombination centres which are formed during heating of irradiated fibre.

Acknowledgments

We wish to thank SERC for a research studentship (JET) and gratefully acknowledge helpful discussion with Dr S W S McKeever, Dr Y Kirsh and Dr H M Rendell.

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