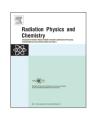
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TL emission spectra measurements using a spectrometer coupled to the Risoe TL/OSL reader

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HIGHLIGHTS

• A spectrometer based on a CCD detector was coupled to a TL/OSL reader via optical fiber.

• TL emission spectra of well-known TL materials were measured.

• The measured spectra showed the expected shapes; thus the system proved a good performance.

• The set-up is very easy to use and it shows good potential to study new materials.

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ABSTRACT

A high sensitivity spectrometer (Ocean Optics QE65 Pro) was coupled to the Risoe TL/OSL reader to measure TL emission spectra of four different dosimeters. This spectrometer is based on a Hamamatsu FFT-CCD detector with a 2-D arrangement of pixels (1044×64), which detects luminescence in a range of 200–950 nm. An optical fiber was used to guide the signal from the sample to the spectrometer. TL spectra from LiF, CaSO₄:Dy, BeO and Al₂O₃:C detectors were obtained and they are presented in this work. The proposed detection system showed good response; spectra shape, in accordance with the literature, were obtained, validating the system.

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1. Introduction

Thermoluminescence is the light emission when certain materials are heated. This phenomenon occurs during the relaxation process in which trapped electrons in crystal defects are released by heating and recombine with holes emitting a photon. Ionizing radiation creates electron-hole pairs that can be quantified through the thermoluminescence signal.

Although the use of thermoluminescence phenomenon for radiation dosimetry ages from the early 1950s (Daniels et al., 1953), the search for new dosimetric materials and for the comprehension of the thermoluminescence mechanism remains nowadays.

Thermoluminescent dosimeters are organic or inorganic crystals with defects or imperfections in their crystalline lattices. According to the energy band theory of solids, the crystal defects or imperfections give rise to energy levels localized at the forbidden gap which may trap electrons or holes created by ionizing radiation

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http://dx.doi.org/10.1016/j.radphyschem.2014.04.014 0969-806X/© 2014 Elsevier Ltd. All rights reserved. incidence. When trapped electrons or holes are stimulated, they may escape to the conduction band and then recombine, emitting luminescence.

The luminescent emission wavelength depends on the recombination centre depth. The deeper the recombination centre, the smaller the luminescence wavelength. The emission spectrum of luminescent materials, along with other studies, helps understanding the defects or imperfections in the crystal lattice and the kinematic phenomenon. This is especially interesting when new materials, natural or synthetic, are being studied.

The emission spectrum can be measured using monochromators or a set of narrow-band filters to discriminate the signal wavelength (Fairchild et al., 1978). These configurations have the advantage of using a photomultiplier tube to collect and amplify the signal, but they have the disadvantage of time-consumption, because only one wavelength is measured at a time. Nowadays, the whole spectrum can be measured at once and with good sensitivity with the new generation of spectrometers (Bakas, 1984). These detectors have a charge-coupled device (CCD) array and can be sensitive from ultraviolet (UV) to near infrared (NIR) region, depending on their configuration.

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In this work, a spectrometer connected to a Risoe TL/OSL reader via an optical fiber was used to perform TL emission spectra measurements. Well-known luminescent materials, like LiF, CaSO₄:Dy, BeO and Al₂O₃:C, were used to test the spectrometric arrangement.

2. Materials and methods

The TL emission spectra were measured using a high-sensitivity spectrometer coupled via optical fiber to a TL/OSL reader.

2.1. Samples

Four different sample pellets were used in this work. All irradiations were carried out using a Gammacell irradiator, Atomic Energy of Canada, with 98.8 TBq activity and 1.18 kGy h^{-1} dose rate from the Center for Radiation Technology of IPEN.

The LiF pellets, with $3.2 \times 3.2 \times 0.9 \text{ mm}^3$ dimensions, were annealed at 400 °C for 1 h before irradiation. 80 °C for 24 h treatment was not undertaken, because it was not intended to reduce the intensity of the low temperature peaks (Fairchild et al., 1978). CaSO₄:Dy sintered pellets were provided by a national company MRA Industria de Equipamentos Eletrônicos Ltda. The pellets were 6 mm in diameter and 0.8 mm thick disks, and they were annealed at 300 °C for 3 h. BeO pellets were obtained from the laboratory stock. They were 4 mm diameter disks with 0.8 mm thickness. These samples were annealed at 750 °C for 15 min. Al₂O₃:C disks were obtained by Rexon Inc. They were 5 mm in diameter and with thickness varying from 0.8 to 1.0 mm. According to the manufacturer, the proper annealing treatment is 800 °C for 15 min.

2.2. Stimulation and detection system

The stimulated luminescence was measured using a fiber coupled spectrometer attached to the Risoe TL/OSL reader, model DA-20 (Fig. 1). A TL/OSL reader, model DA-20, Risoe DTU, Denmark, was used. This reader is equipped with a heating system that uses alternating current controlled by a non-switching continuous full sine wave generator operating at 20 kHz. Samples can be heated up to 700 °C with heating rates from 0.1 to 10 K/s. For the lumine-scence detection a bialkali photomultiplier tube (PMT), model 9235, PRF Technologies was used. The Risoe reader is also equipped with a ⁹⁰Sr/⁹⁰Y beta source.

The optical fiber used is a customized UV–vis solarizationresistant fiber, with 1.0 mm core diameter. This fiber presents a relative transmission better than 80% in the range of 250–900 nm

Fig. 1. Risoe TL/OSL reader, spectrometer and optical fiber used to measure the TL spectra of different samples.

and an acceptance angle in air of 25°. Bos et al. (2002) calculated that, for samples of 3 mm diameter, a distance up to 10 mm from the sample to the fiber entrance the light collection keeps constant. For 5 mm diameter samples this distance is 13 mm. In the case of the Risoe TL/OSL reader, model DA-20, the distance is shorter than 5 mm and it is mostly due to a quartz window used to isolate the sample ambience.

Spectra measurements were carried out using a scientific-grade spectrometer Ocean Optics, QE65 Pro, with a Hamamatsu FFT-CCD detector with a 2-D arrangement of pixels (1044×64). This detector is configured with a special diffracting grade HC-1 (groove density of 300 mm⁻¹ and spectral range of 750 nm) and a 200 µm entrance slit, which, according to the manufacturer, provides a pixel resolution of ~8.9 pixels. The sensitive wavelength region is from 200 to 950 nm and the wavelength resolution is 6.5 nm FWHM. The detector is totally plug-and-play type, and the software is provided with it. Data unique to each detector is recorded into a memory chip and can be easily accessed through software. Measurements from 8 ms to 15 min can be performed due to its low dark count, high stability and integrated cooling system.

The manufacturer previously calibrated the spectrometer on wavelength. This calibration may be repeated after a period of time, but as the equipment is brand-new, this calibration was not performed yet. A relative calibration of the system efficiency was obtained using a tungsten-halogen light source. This means that all measurements are relative to that of a blackbody light source with known color temperature.

3. Results and discussion

Using the equipment showed in Fig. 1, the TL spectrum of different luminescent materials was measured. These materials were previously irradiated, and the reading parameters were the same for all samples.

The absorbed dose used was very high, not applicable to personal dosimetry, but it was necessary to obtain a defined signal and to observe luminescence from different recombination centers. Fig. 2 shows the TL spectrum of a LiF pellet irradiated to 100 Gy (60 Co). The luminescence was integrated over 3 s with a heating rate of 2 °C/s.

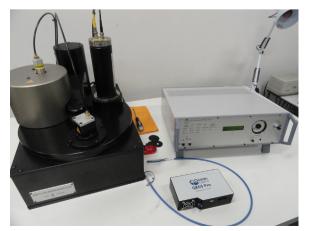
This sample shows a broad dosimetric peak centered around 410 nm. There are many articles about the LiF luminescence response. Although the spectrometric system measured the LiF luminescence showing the dosimetric peak, some details of its emission cannot be noticed. Another measurement with a higher dose was also taken. In Fig. 3, it is possible to see a high definition LiF TL spectrum. For this measurement the sample was irradiated to 1 kGy (60 Co), and it was heated at a rate of 5 °C/s.

From Fig. 3 it is also possible to observe minor emissions at longer wavelengths which cause the appearance of a shoulder, but it is not possible to resolve these peaks. The four glow peaks (peak 2 to peak 5) can easily be observed besides the complex peaks at higher temperatures. Results are consistent with the literature (Piters et al., 1993).

In Fig. 4, a CaSO₄:Dy emission spectrum is shown. The sample was irradiated to a dose of 100 Gy (60 Co), and its luminescence was integrated for 2 s during a heating rate of 2 °C/s.

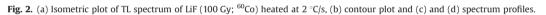
The main dosimetric peak around 270 °C is composed mostly by two dominant emission bands at 480 and 580 nm as expected (Piters et al., 1993). The sharp lines seen in Fig. 4d are characteristic of the rare earth dopant (Dy). The CaSO₄:Dy glow curve and the mechanisms of the defect structure are very complex, but this material showed the best TL sensitivity among the studied materials.

The BeO sample presents only one broad peak; however, this peak apparently consists of an overlapping of other components



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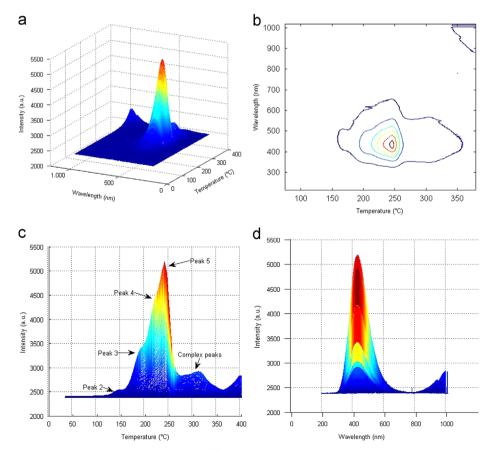


Fig. 3. (a) Isometric plot of TL spectrum of LiF (1 kGy; ⁶⁰Co) heated at 5 °C/s, (b) contour plot and (c) and (d) spectrum profiles.

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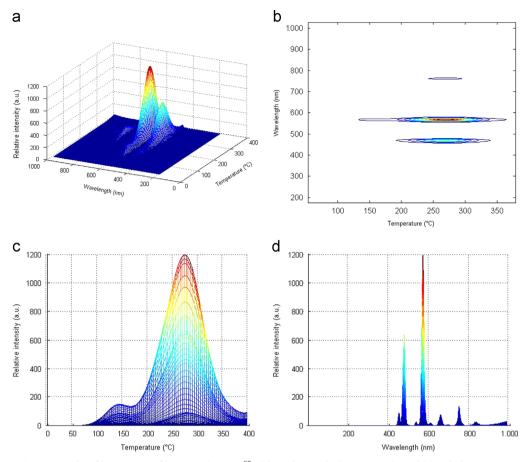


Fig. 4. (a) Isometric plot of TL spectrum of CaSO₄:Dy (100 Gy; ⁶⁰Co) heated at 2 °C/s, (b) contour plot and (c) and (d) spectrum profiles.

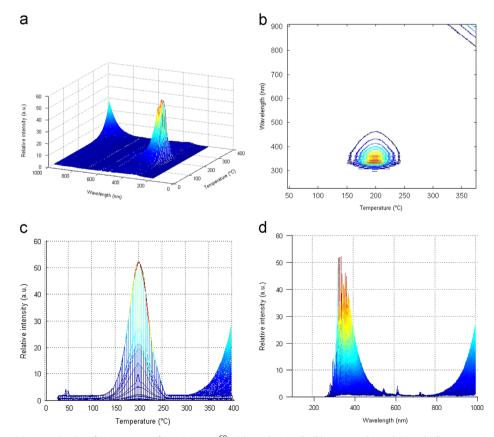


Fig. 5. (a) Isometric plot of TL spectrum of BeO (100 Gy; ⁶⁰Co) heated at 2 °C/s, (b) contour plot and (c) and (d) spectrum profiles.

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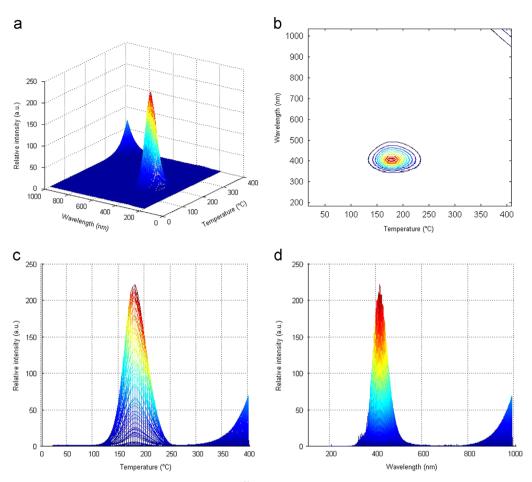


Fig. 6. (a) Isometric plot of TL spectrum of Al₂O₃:C (100 Gy; ⁶⁰Co) heated at 2 °C/s, (b) contour plot and (c) and (d) spectrum profiles.

(Fig. 5). It is centered at 330 nm, and it shows a shoulder to longer wavelengths. The glow curve of BeO depends upon the origin of the material, and the peak shifts with the absorbed dose (McKeever et al., 1995).

The TL emission spectrum of Al_2O_3 :C is shown in Fig. 6. The sample was irradiated to a dose of 100 Gy (⁶⁰Co), and its luminescence was integrated for 2 s during a heating rate of 2 °C/s.

The expected glow curve for Al_2O_3 :C is a single peak centered at approximately 210 °C (at a heating rate of 10 °C/s), but this material presents strong thermal quenching, and the peak shifts to higher temperatures due to increasing heating rates. The maximum emission peak, at 410 nm, is caused by electrons trapped on F⁺ centers (Summers, 1984).

4. Conclusions

Well-known TL materials were used to test a spectrometric system, which is based on a CCD detector coupled directly to a TL reader via optical fiber. The TL spectrum of these materials was obtained, and it was possible to observe the expected characteristics of each spectrum. LiF pellets present four glow peaks, so a higher dose (1 kGy) was necessary to see these peaks. CaSO₄:Dy, BeO and Al₂O₃:C present a single glow peak, which consist of an overlapping of other components, and a dose of 100 Gy was enough to obtain a good response. The heating rate was kept very low compared to that of routine, 2 °C/s, because at low rates the luminescence process is optimized. This system showed good

potential to study new materials, once it is very easy to use, and it showed a good performance.

Acknowledgments

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