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Spectral distribution of the thermoluminescent light emitted by gamma irradiated sodium diborate compounds

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Abstract. Samples of amorphous sodium diborate compounds were exposed to gamma radiation from a ^{60}Co source and the thermoluminescent emission spectra recorded. The spectrum is broad and shows two bands centred at around 360 and 500 nm, and a long wavelength tail from 530 nm. From glow curves obtained by using filters the spectral distribution of the light emitted at different temperatures has been determined.

1. Introduction

Sodium diborate compounds become thermoluminescent by exposing them to X or γ radiation [1, 2]. Glow curves recorded at different times after irradiation are shown in figure 1. The glow curves are made up of a low temperature peak, whose intensity decreases as the time elapsed between irradiation and recording of the glow curve increases, and a broad peak at higher temperatures [2]. When the heating rate becomes 250 °C/min the low temperature peak occurs at around 120 °C and the broad peak

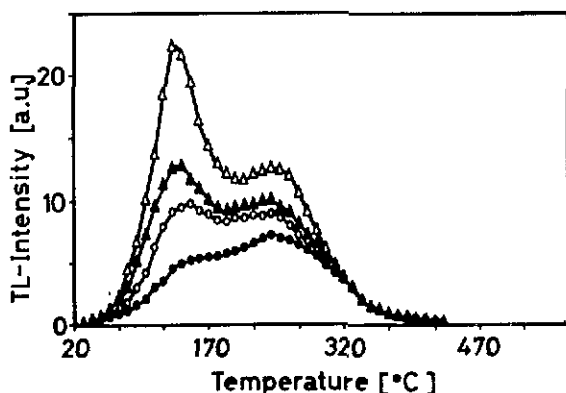


Figure 1. Glow curves taken at different times after irradiation Δ , 20 min; \blacktriangle , 1 day; \circ , 3 days; \bullet , 7 days. Heating rate: 250 °C/min.

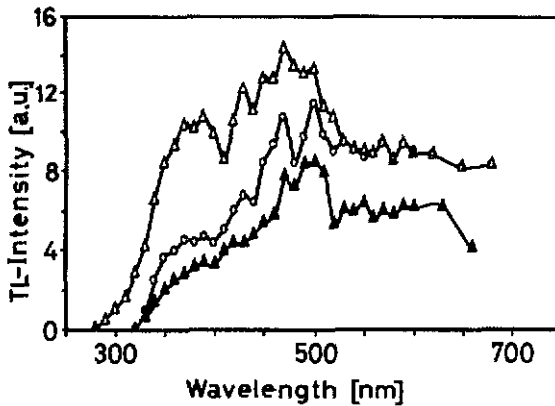


Figure 2. Spectral emission curves taken at different times after irradiation; Δ , 20 min; \circ , 3 days; \blacktriangle , 10 days.

extends from about 170° to 420 °C. In this paper we report on the spectral distribution of the thermoluminescent light. Measurements were performed for the light emitted between room temperature and 420 °C, and at different temperatures corresponding to the low temperature peak, the broad peak, and at a temperature between the peaks.

In the next section the experimental procedure is described. It follows a discussion on the thermoluminescent mechanisms based on the results reported in this and previous work.

2. Experimental procedure

The thermoluminescent emission spectra were measured using a monochromator (Heath EU-700), a photomultiplier (EMI-9558-QA) and an automatic integrating pico-ammeter (Harshaw 2000-B).

After setting the monochromator to a given wavelength the sample was heated, suddenly, up to 430 °C and the intensity of the light emitted during the heating recorded. Taking into account the monochromator specifications and the photomultiplier spectral response the readings were corrected to obtain the light intensities at different wavelengths. The intensity plotted at each wavelength is the average of several measurements. The spectra recorded at different times after irradiation are shown in figure 2.

The spectral distribution of the light emitted at different temperatures could not be found by employing the monochromator because of the weakness of the light intensity. To find out the spectra we resorted to 12 long-wave pass filters, whose successive cut-off wavelengths differ by about 20 nm. Glow curves were recorded employing the filters. Taking into account the transmission curves of the filters and the spectral response of the photomultiplier the spectra for 120, 170 and 250 °C were computed. The wavelengths indicated in figure 3 are the averages between two successive cut-off wavelengths.

3. Results and discussion

Spectral emission curves recorded 20 minutes, 3 and 10 days after irradiation are plotted in figure 2. The emission spectrum obtained after 20 minutes has a peak between 280

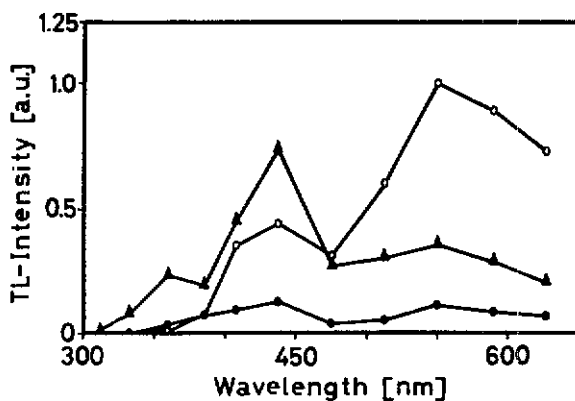


Figure 3. Spectral distribution of the light emitted at ▲, 120; ●, 170; and ○, 220 °C.

and 400 nm, whose intensity in the spectra taken after 3 and 10 days shows a stronger decrease than in the region for wavelengths longer than 400 nm. A similar behaviour is observed in the glow curve for the peaks at 120 °C and the broad peak, as can be seen from figure 1. These results suggest that for wavelengths shorter than 400 nm the light contributes mainly to the low temperature peak of the glow curve. This conjecture is confirmed by the spectral distribution of the light at 120, 170 and 250 °C plotted in figure 3. The spectra at different temperatures higher than 170 °C show no essential differences to the spectrum at 250 °C, except for the relative intensities of the peaks at around 440 and 550 nm. As can be seen from figure 3 the spectral composition of the light at 120 °C has, besides the peak at 360 nm, two peaks centred at approximately the same wavelengths as the peaks appearing at temperatures higher than 170 °C.

Upon irradiation, paramagnetic centres are formed in alkali borates. At room temperature an electron spin resonance signal can be measured corresponding to two kinds of centres [3]. One is a trapped hole in an oxygen atom having a hyperfine interaction with two boron atoms. The other centre is believed to consist of a trapped hole in an oxygen atom having a hyperfine interaction with a boron atom. In what follows we will denote these centres BOHC (boron oxygen hole centre). There is another type of paramagnetic centre consisting of an electron trapped in an alkali metal, which we will denote hereafter AMEC (alkali metal electron centre) [4]. AMEC centres exist at room temperature and their ESR signal can only be observed at lower temperatures. BOHC and AMEC centres give rise to an absorption band in the radiation induced optical spectrum shown in figure 4. Pb^{2+} doping experiments have shown that the band at around 600 nm consists of the contribution of peaks centred at 500–550 and 630 nm. The former is due to BOHC centres, while the latter is related to AMEC centres. The absorption of light by BOHC centres has been investigated further in alkali borates. The results have shown absorption at 540 (2.3 eV) and 340 nm (3.6 eV) [5]. In figures 5 and 6 radiation induced optical spectra and glow curves for different concentrations of soda are shown [1, 6]. As the soda content increases both the absorption of light between 340 and 630 nm (from 2 up to 3.6 eV) and the thermoluminescent intensity increase. These results suggest that paramagnetic centres are involved in the thermoluminescent mechanisms. This connection can also be inferred by comparing the radiation induced optical spectra of both sodium and lithium diborate reported by Beekenkamp [6] (figures 5 and 7), with

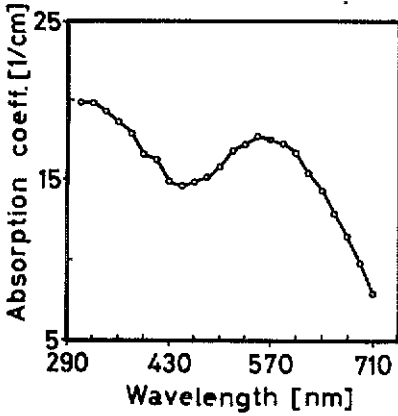


Figure 4. Radiation induced optical spectrum of sodium diborate.

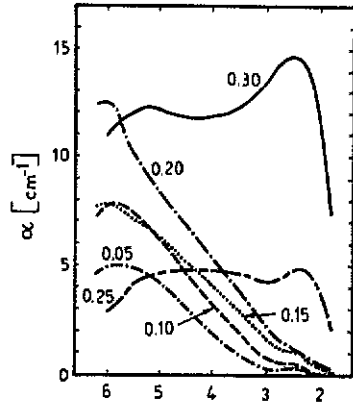


Figure 5. Radiation induced optical spectrum of sodium borates for different molar concentrations of soda (after Beekenkamp [6]).

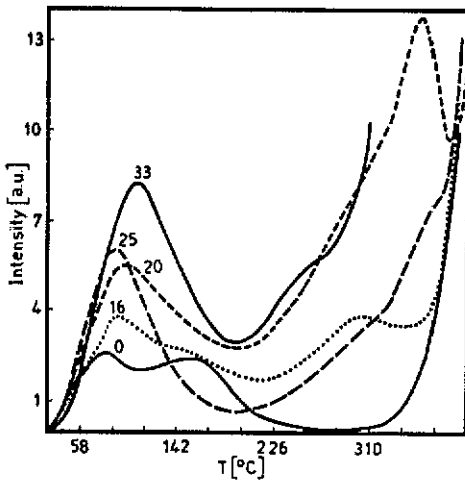


Figure 6. Glow curves of sodium borates for different molar concentrations of soda (after Nasipuri [1]).

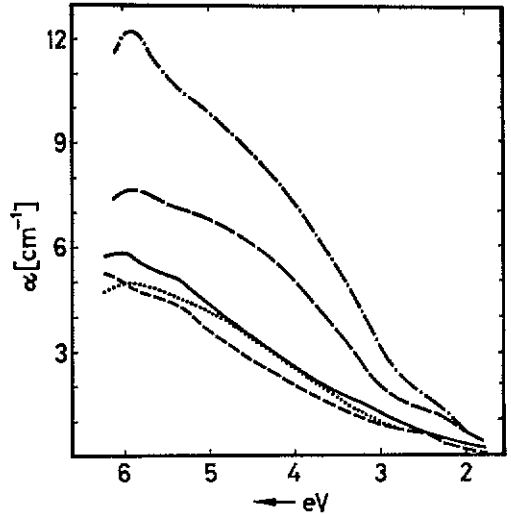


Figure 7. Radiation induced optical spectrum of lithium diborate for different concentrations of lithium oxide (after Beekenkamp [6]). —, 0.05 mol; —, 0.10 mol; —, 0.15 mol; —, 0.20 mol; ···, 0.25 mol.

their glow curves shown in figure 8 (the samples were given the same dose). In lithium diborate both the ESR signal intensity and the band in the radiation induced optical spectrum are smaller. According to ESR thermoluminescence correlation measurements in sodium diborate BOHC centres can only be involved in the emission of light related to the broad peak, i.e. at temperatures higher than 170 °C [2]. Further, AMEC centres are stable up to 150 °C and higher temperatures [5]. From the earlier discussion it follows that both centres can only be related to the thermoluminescent mechanisms giving rise to the broad peak.

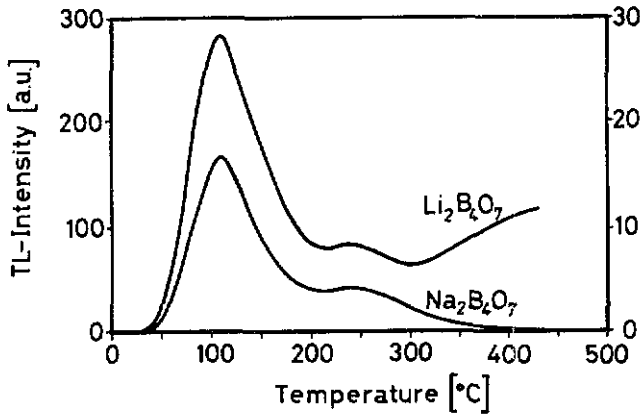


Figure 8. Glow curves of sodium and lithium diborates. The samples were given the same dose.

From figure 3 it follows that both the thermoluminescent mechanism related to the peak at 120 °C, in which paramagnetic centres are not involved, and the thermoluminescent mechanisms related to the broad peak, in which paramagnetic centres are likely to be involved, give rise to similar spectra for wavelengths higher than 350 nm. This result can be accounted for by resonant transfer of energy between recombination and luminescent centres (activators) [7]. A non-radiative transition releases energy which is taken up by a centre becoming excited; then the centre returns to its ground state by emitting a photon. If transitions occurring at different temperatures excite the same activator, the wavelength(s) of the light emitted by the activator will be observed in the spectra recorded at different temperatures.

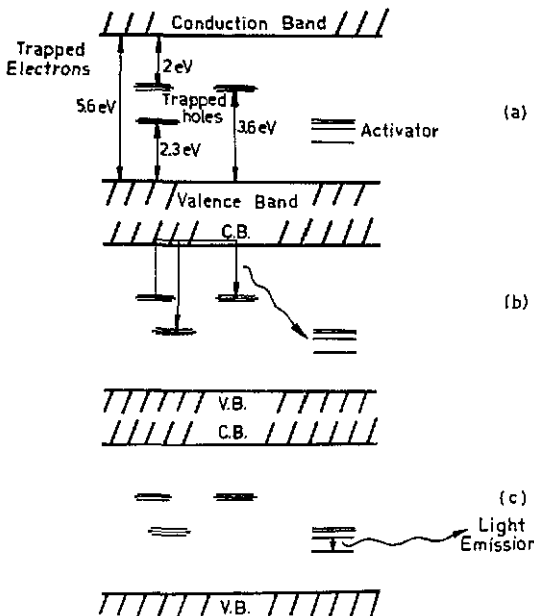


Figure 9. Model for thermoluminescent mechanisms related to the broad peak. (a) Energy levels of the electron trap due to AMEC centres and of hole traps due to BOHC centres; (b) electron transitions from AMEC to BOHC centres and resonant energy transfer from both transitions to an activator; (c) light emission by an activator.

Resonant energy transfer has been proposed by D Shearer [8] to account for the experimental findings in lithium diborate and later by Y Kutomi *et al* [9]. This result is to be expected when it is realized that AMEC and BOHC centres occur in both lithium and sodium diborate and that both compounds have similar thermoluminescent properties [2].

The earlier discussion shows that resonant transfer of energy satisfactorily explains the thermoluminescent mechanisms in sodium diborate compounds. Possible mechanisms giving rise to the broad peak are shown schematically in figure 9. The band gap was taken from reference [10], and the energy levels of AMEC and BOHC centres have been estimated from their contributions to the radiation induced optical spectrum. According to the non-radiative transitions shown in figure 9(b), the transitions to the low-lying hole trap can transfer enough energy to the activators for them to emit light whose wavelength is 350 nm or longer, while the transitions to the other hole trap can only be involved in the emission of light whose wavelength is approximately 630 nm or longer. In figure 9 only an activator is shown. It is possible that an activator gives rise to two or more peaks, either because it can be excited to different energy levels or because it can occupy sites having surroundings with different symmetries. The nature of the activator has not been investigated. According to the spectral distribution of the light for wavelengths longer than 400 nm, Mn^{2+} could be the activator; it emits light at approximately 450 nm, and 530 or 620 nm, the wavelength depending on the site symmetry [11]. Doping sodium diborate with Mn should provide information on the possibility of traces of Mn^{2+} being the activator. This work is in progress at our laboratory.

Acknowledgment

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