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SPECTRAL DEPENDENCE OF OPTICAL BLEACHING OF PTTL IN QUARTZ

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Abstract — The phototransferred thermoluminescence (PTTL) was studied in quartz. The PTTL was excited in X or β irradiated samples by monochromatic light between 250 and 500 nm. Main PTTL peaks appeared at about 110, 160 and 200°C. The temperatures of these peaks as well as their thermal activation energies fit those of X and β induced TL peaks; emission spectra of the PTTL showed the same 370 nm and 450 nm emission bands as the regular TL. Effects of optical bleaching on the PTTL intensities were investigated. The spectral dependence of the bleaching efficiency was measured for the 110°C peak and showed maxima at 310 nm and at 280 nm. For comparison, the excitation spectrum of this PTTL peak was measured and also showed maxima at 310 and 275 nm: these results indicate that the same wavelengths, which are most efficient for the transfer of carriers from deep to shallow traps, are most efficient for optical bleaching as well.

INTRODUCTION

Much interest has been devoted during recent years to phototransferred thermoluminescence (PTTL) studies from both the point of view of basic and applied research. There are many advantages in using this technique for archaeological and geological dating. Various research groups have used this method for the thermoluminescence (TL) dating of quartz[1-4]. For these measurements the samples are normally exposed to ionising radiation at a constant temperature T_1 such as RT, then heated to a given higher temperature T_2, re-cooled to T_1 and illuminated at this temperature with light of wavelengths which do not excite TL in unirradiated crystals. PTTL peaks may appear during repeated heating from T_1 to T_2. The appearance of PTTL peaks during reheating from T_1 to T_2 is attributed to a process in which charge carriers, that had been trapped by the ionising radiation in deep traps and were transferred by light illumination to shallower traps, are thermally released from these traps during reheating and recombine radiatively with carriers of opposite sign.

In a recent study, the PTTL of synthetic quartz was studied in our laboratory[5]; UV excitation spectra of the PTTL as well as emission spectra were measured. In the present work the PTTL was studied in natural BDH quartz crystals and the results were compared with those obtained for the synthetic samples. The dependencies of the PTTL intensities on the β dose as well as on the dose of the UV light were studied. In particular, effects of optical bleaching of PTTL were investigated and the dependence of the bleaching efficiency on the wavelengths was measured.

EXPERIMENTAL TECHNIQUES

The natural Norwegian quartz crystals were obtained from British Drug Houses (BDH) and the synthetic (premium Q grade) crystals from Sawyer Research Products. Single crystals of approximately $0.5 \times 0.2 \times 0.1$ cm^3 were used for the measurements. In order to increase the TL sensitivity of our crystals for most measurements, the samples were annealed to about 900°C. The X irradiation was performed at RT with a W tube (40 kV, 15 mA) and the β irradiation with a ^{90}Sr source. For the PTTL studies the X or β irradiated crystals were heated to about 250°C, then re-cooled to RT and illuminated with monochromatic light in the spectral region of 270 and 500 nm, obtained from a 150 W high pressure Xe lamp and a 0.25 m grating monochromator. The irradiance of the incident light beam was measured with a Molectron pyroelectric radiometer. The PTTL measurements were recorded during heating at a constant rate of 5°C.s^{-1}. Further experimental details have been given elsewhere[5].

EXPERIMENTAL RESULTS AND DISCUSSION

TL peaks of the X or β irradiated samples appeared at about 110, 160, 200 and 330°C. The main PTTL peak appeared at about 110°C and weaker peaks at 160 and 200°C. No PTTL could be recorded in unirradiated crystals and in samples which were heated to above 350°C between the X irradiation and the illumination with monochromatic light in the spectral region between 270–500 nm. The temperatures of the PTTL peaks as well as their thermal activation energies were found to be the same as those of the X or β induced TL peaks (see Table 1). These results indicate that the PTTL and the TL are due to the same traps. Emission spectra of the main TL and PTTL peaks were also measured. The well known emission bands near 370 and 450 nm appeared in the PTTL as in the TL of the quartz crystals, indicating that the emission is due to the same recombination processes. The relative intensities of these emission bands varied, however, with temperature and pretreatment. In the PTTL and TL peaks above room temperature, the emission band near 370 nm was dominant in the thermally pre-treated samples and the broad band near 450 nm was relatively weak. In the untreated crystals, the emission band near 450 nm was relatively strong. The increase of the 450 nm intensity was even more pronounced in the low temperature XL emission.

Special attention was given to the study of optical bleaching of PTTL. Thermal as well as optical bleaching of TL have previously been studied in quartz[2], but no data were given for the wavelength dependence of the efficiency of optical bleaching. In the present work this dependence was measured for the main PTTL peak near 110°C. For these investigations the X or β irradiated crystals were illuminated at RT for relatively long periods of time with intense polychromatic or monochromatic light beams of various wavelengths between 270 and 450 nm and then heated to 250°C. The PTTL was induced in these samples by a relatively small and constant test dose of 310 nm UV light (the irradiance of the test dose was about two orders of magnitude lower than that of the bleaching light). In Figure 1, the intensities of the PTTL peak near 110°C are given as a function of the bleaching time for illumination with various wavelengths. Each point of the curves represents the PTTL intensity induced by the constant test dose of 310 nm light. It can be seen that wavelengths longer than 370 nm did not cause any optical bleaching. In order to investigate the wavelength dependence of the bleaching, the irradiances of the incident light beams at various wavelengths were monitored

Table 1. Temperatures of TL/PTTL peaks, thermal activation energies evaluated by initial rise (E_1) and by symmetry method (E_2), symmetry factor (μ_g) and order of kinetics (l).

Peak temp. (°C)	TL/PTTL	E_1(eV)	E_2(eV)	μ_g	l
110	TL	0.85	0.9	0.42	1
110	PTTL	0.87	0.95	0.43	1.1
160	TL	—	1.1	0.42	1
160	PTTL	—	1.1	0.44	1.2
200	TL	1.15	1.2	0.45	1.2
200	PTTL	1.3	1.25	0.44	1.1

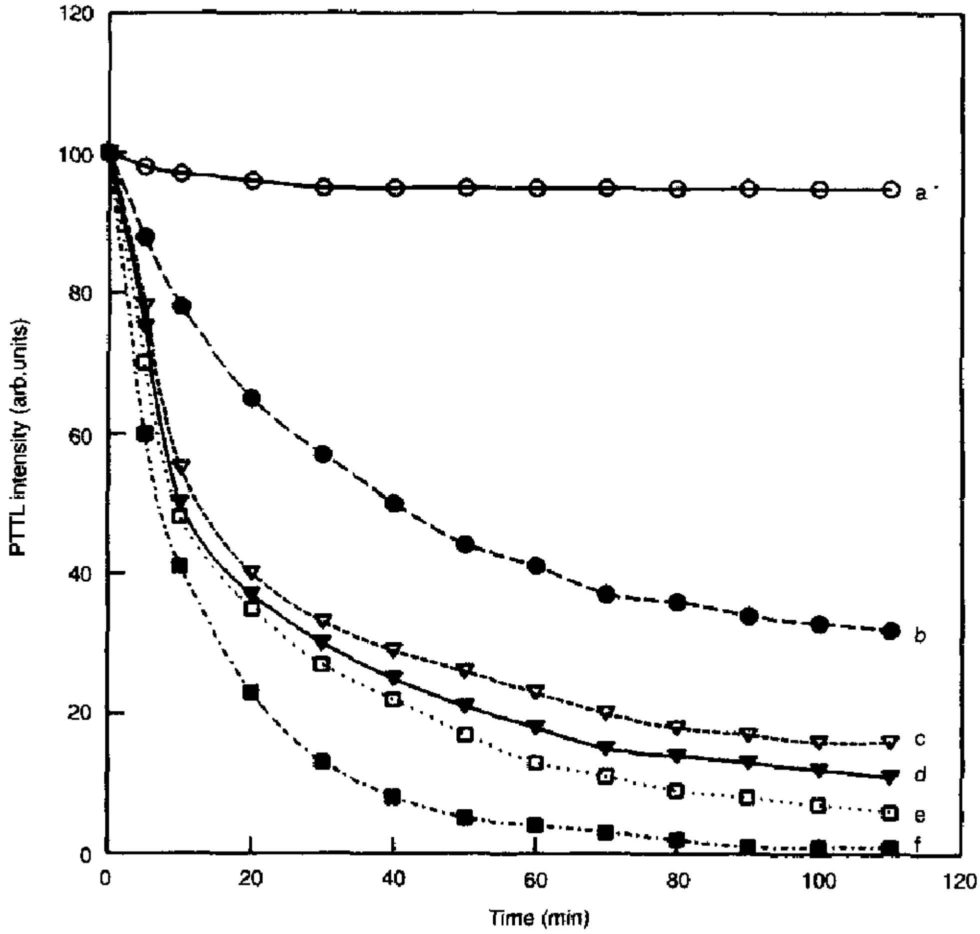

Figure 1. Intensities of the 110°C PTTL peak as a function of time of optical bleaching with various wavelengths: A, 370 nm; B, 270 nm; C, 350 nm; D, 280 nm; E, 330 nm; F, 310 nm (see text).

with a pyroelectric radiometer, and the PTTL intensities before and after the prolonged exposure to the intense bleaching light were measured. The efficiency (η_a) of the optical bleaching was evaluated by the ratio of the PTTL intensities, excited by the test dose of 310 nm light, before (I_1) and after (I_2) the exposure to the strong bleaching light: $\eta_a = (1 - I_2/I_1) \times 100$.

In Figure 2, curve A, the relative efficiency of the optical bleaching is given as a function of the wavelength for constant illumination time and irradiance. It can be seen that the bleaching efficiency reached maxima near 280 and 310 nm. Excitation spectra of the PTTL peak near 110°C were also measured in the same samples and results are given by curve B of Figure 2. The comparison of curves A and B shows that both have maxima at the same wavelengths of 280 and 310 nm, indicating that the wavelengths, which are most efficient for the transfer from deep to shallow traps, are also most efficient for the depletion of the donor levels. Some of our preliminary investigations on the bleaching of optical stimulated luminescence (OSL) in quartz have shown that the same wavelengths are also most efficient for optical bleaching of the OSL in these crystals. The wavelength of 280 nm corresponds to an electron trapping level of about 4.4 eV below the conduction band. A level near this photon energy has previously been attributed to Ge impurities in natural quartz[6]. A weak absorption band at about 320 nm has previously been reported in fused silica and in heavily γ irradiated smoky quartz crystals[7], and has been ascribed to a trapped electron centre; no absorption band could be detected at this wavelength in our X irradiated samples.

In Figure 3, the dependence of the intensity of the 110°C PTTL peak on the β dose is given for a natural crystal. The PTTL was excited by a constant test dose of 310 nm light. For small β doses up to about 50 Gy the dose dependence of our samples was nearly linear, a tendency to saturation can be seen for higher β doses. Essentially the same behaviour was observed for the synthetic samples.

The dependence of the PTTL intensity on the dose of 310 nm UV light was also investigated. In Figure 4, the dependence of the 110°C PTTL peak on the illumination time is given for a constant irradiance of an intense 310 nm light beam and for a constant β dose. A maximal level was reached after approximately

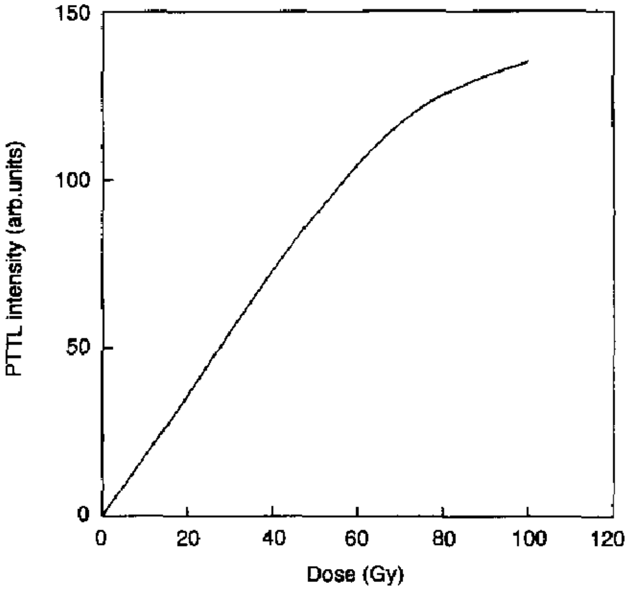

Figure 3. Dependence of 110°C PTTL peak on β dose. The PTTL was induced in all cases by a constant small test dose of 310 nm UV light (1 min; 5 μW.cm^{-2}).

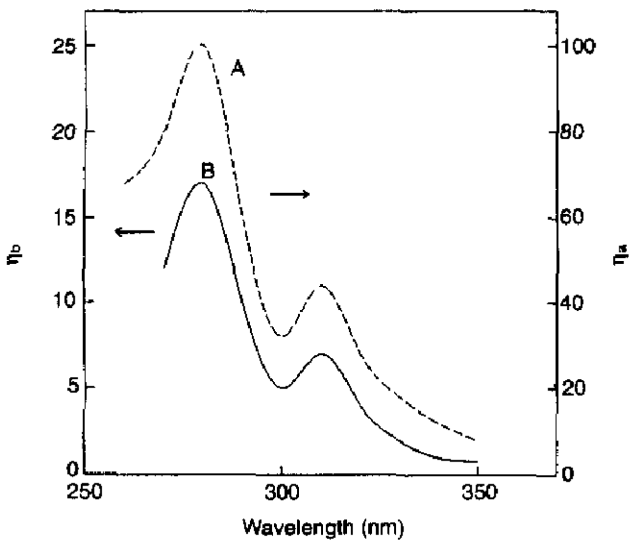

Figure 2. Relative efficiency of (A) the optical bleaching (η_a) of 110°C PTTL peak and (B) of the PTTL excitation (η_b) as functions of illumination wavelengths.

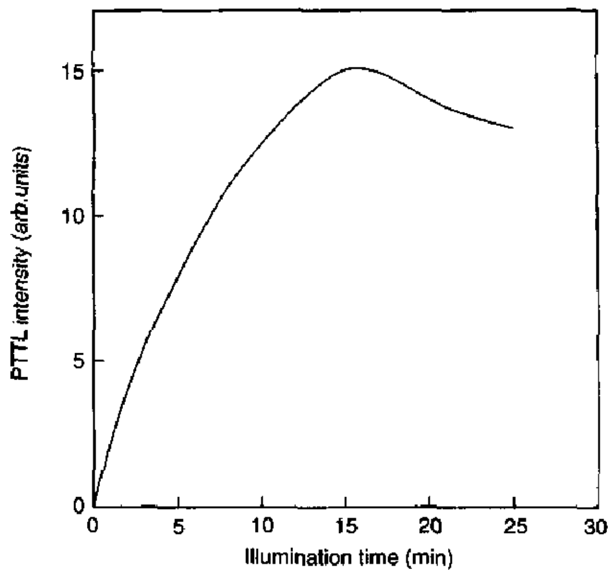

Figure 4. Dependence of the 110°C PTTL intensity on the time of UV illumination. The samples were irradiated with a constant β dose and then illuminated with 310 nm light of constant irradiance of 0.5 mW.cm^{-2}.

15 min, and after longer illumination, a decrease in PTTL intensity was observed. This behaviour is attributed to a contrary effect of optical bleaching of the PTTL, due to the depletion of the donor levels, which becomes dominant after prolonged exposure to the intense 310 nm light. These results are in accordance with the finding that wavelengths of 310 and 280 nm light, which were found to be highly effective for the transfer of carriers and PTTL excitation are also most effective for the optical bleaching.

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