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THERMOLUMINESCENCE AND KINETICS OF GAMMA IRRADIATED BHUTANESE STONE

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Abstract. The present paper reports the photoluminescence (PL) and thermoluminescence (TL) studies of natural Bhutanese stone (Biotite) collected from Bhutan. The sample was Bhutanese stone irradiated with Co^{60} gamma source giving a dose from 0.5 kGy to 2 kGy. The heating rate used for TL measurements was 6.7 ºC/s. The heating rate used for TL measurements was 6.7 ºC/s. The samples shows good TL peaks, the corresponding activation energy (E) values are calculated. Structural characteristics were showed by XRD spectrum of the sample. The value of trap depth of Bhutanese stone was evaluated by different methods. Also the annealing quenching effect was studied for gamma exposure of the sample.

1. Introduction

Bhutan is a magic country with magic geology – metamorphic, structural, thermal and rock magnetic history. There have been small scale mining activities in Bhutan for over a thousand years. Slags are found in many parts of country. They point to mining of lead-zinc and iron ores for production of artifacts, weapons, and most notably a series of iron-chain suspension bridges which were constructed in the 14th century by Saint Thangthong Gyalpo (1385-1464).

Mineral exploration began only in the early 1960s. Less than 30 % of total area of the country has been mapped geologically. Geological surveys are carried out jointly by Survey of India and Department of Geology and Mines of the Royal Government. So far surveys have shown that there are deposits of coal, limestone, dolomite, talc, marble, gypsum, slate, lead, zinc, copper, tungsten, graphite, iron, mica, phosphate, pyrite, asbestos, and gold.

It has been variously claimed by different authors that the thermoluminescence of meteorites can be used to provide data on their shock/reheating history, cosmic ray exposure age, orbit, preatmospheric shape, ablation rate, terrestrial age and petrologic type [1-5]. Thermoluminescence dosimetry is a field of recent origin which has proved to be special importance in the estimation of radiation doses. Special attention has been paid to the

development of materials for TL dosimetry. Number of scientists have standardized many materials for TL dosimetry and investigated in detail the fact that if luminescent material satisfies certain minimum requirements (dosimetric properties), they can be used in radiation dose estimation in TLD [6-9].

Thermoluminescence (TL) is the emission of light observed during the heating of insulating or semiconductor materials, provided that they have been previously exposed to ionising radiation (McKeever, 1985 [1]; R. Chen and S.W.S. McKeever, 1997 [9]; M. Martini and F. Meinardi, 1997 [10]). This last feature mentioned, i.e. its dependence on the amount of the energy absorbed during irradiation, called radiation dose, plays a primary role in the dosimetric applications of TL. Several artificial and naturally occurring materials show this favorable property, covering a very wide range of dose (10^{-2} - 10^{8} Gy approximately). They are widely used in radiation protection practices and can be used to measure the doses due to occupational exposure and those accrued as a consequence of nuclear accidents. New materials have been developed to best fit the characteristics required by the main specific applications which are personnel, environmental, medical, retrospective and high–dose dosimetry [11]. Figure 1 shows the geology map of Bhutan area. It represents simplified geological map of Bhutan. Figure 2 shows fine, laminated clayey and sand metasediment with crenulation and oblique schistosity. Figure 3 shows the Biotite schist with well developed crystals.

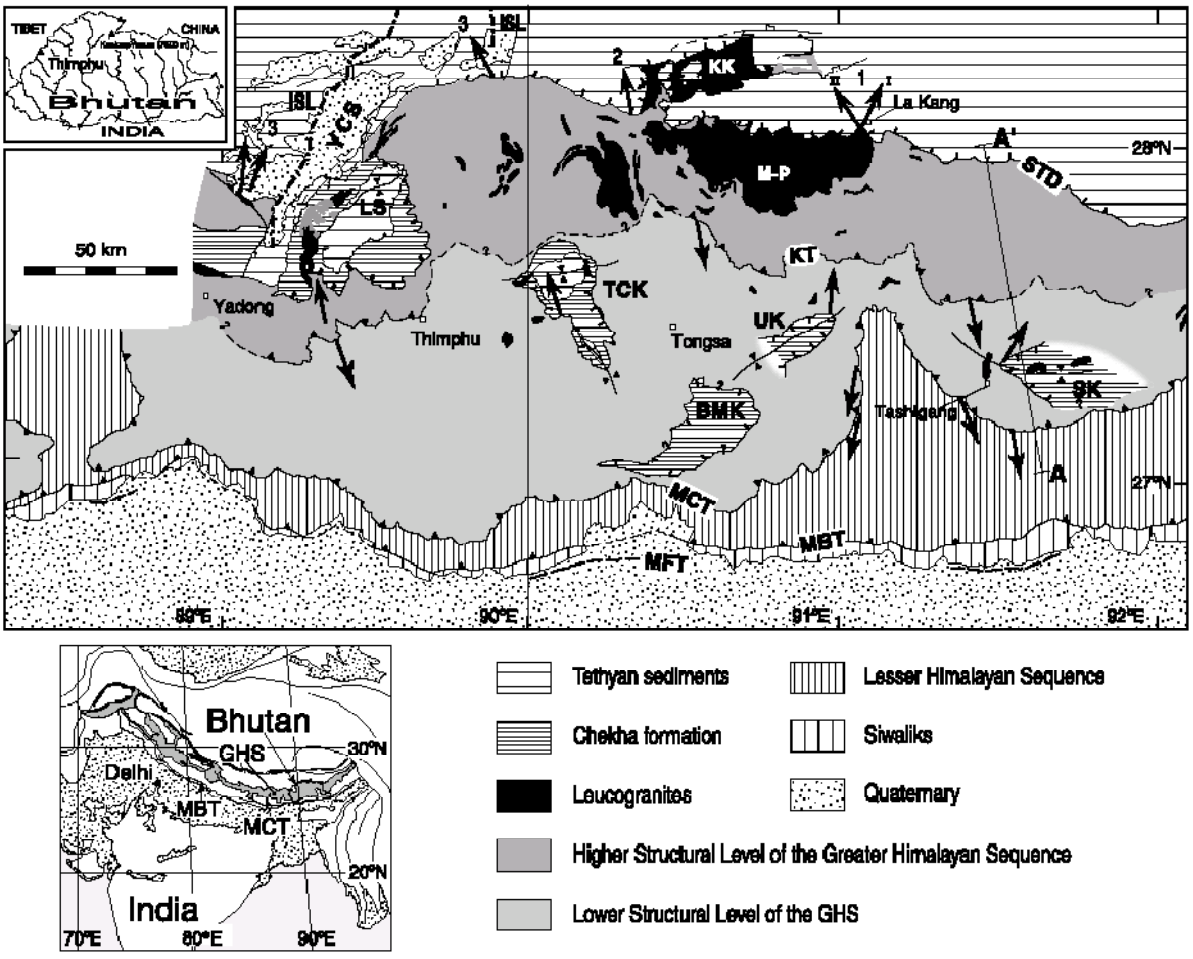

Fig. 1. Simplified geological map of Bhutan.

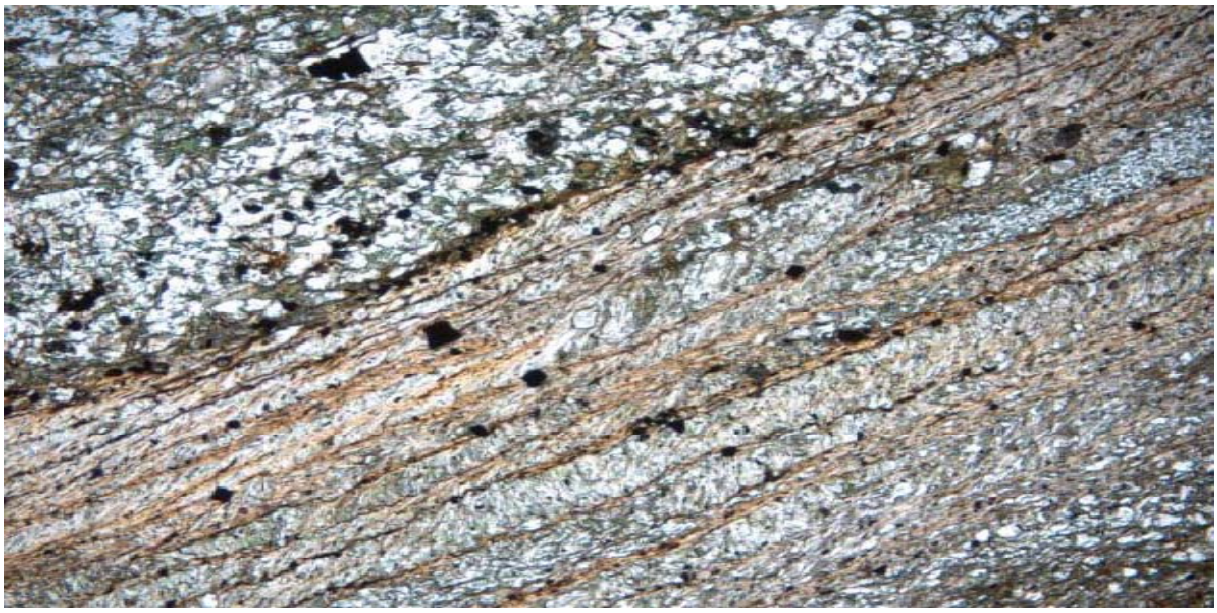

Fig. 2. Fine laminated clayey and sand metasediment with crenulation and oblique schistosity.

Fig. 3. Biotite schist with well developed crystals (B7).

2. Experimental

The natural Bhutanese samples were collected from Bhutan. The TL glow curve was recorded by TLD Reader. The photoluminescence (PL) emission and excitation spectra were recorded at room temperature using Spectrofluorophotometer RF-5301 PC of SHIMADZU make. The excitation source is a xenon lamp. The chemical characterization was done by NGRI Hyderabad with the instrument Perkin Elmer Sciex ELAN DRC II. For gamma irradiation

Co^{60} source was used. For TL measurement every time 2 mg weighed specimen is taken. From the data TL glow curve has been drawn using MS-Excel and the shape factor (μ) has been calculated. The sample was characterized by XRD. The XRD measurements were carried out using Bruker D8 Advance X-ray diffractometer. The x-rays were produced using a sealed tube and the wavelength of x-ray was 0.154 nm (Cu K-alpha). The x-rays were detected using the fast counting detector based on Silicon strip technology (Bruker LynxEye detector). The samples were annealed at different temperatures:

a. samples were pre annealed for 1 h at 400 ºC, and then they were immediately cooled to room temperature;
b. samples were pre annealed for 1 h at 600 ºC, and then they were immediately cooled to room temperature;
c. samples were pre annealed for 1 h at 800 ºC, and then they were immediately cooled to room temperature.

3. Results and discussions

3.1. XRD analysis of Bhutanese stone. The XRD measurements were carried out using Bruker D8 Advance X-ray diffractometer. The x-rays were produced using a sealed tube and the wavelength of x-ray was 0.154 nm (Cu K-alpha). The x-rays were detected using a fast counting detector based on Silicon strip technology (Bruker LynxEye detector). Figure 4 shows the XRD spectra of Bhutanese stone (Biotite). From the Table 1 it shows the cubic structure of the sample and the hkl values are calculated. The Biotite shows the cubic structure and it was verified by XRD spectra. The PL spectra were recorded at room temperature (see Fig. 5). Photoluminescence emission spectrum peak at 390 nm is reported for Bhutanese stone.

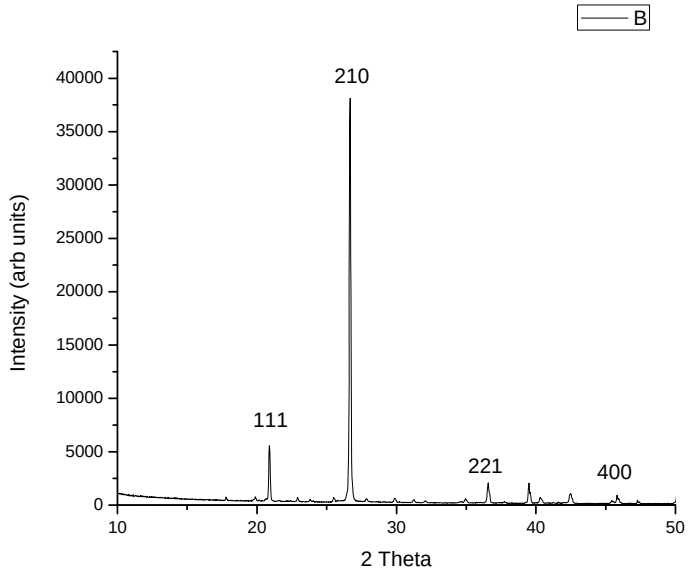

Fig. 4. XRD Pattern of Bhutanese stone.

Table 1. XRD calculation of Bhutanese stone for cubic structure.

Peak #	2θ	θ	$\sin^2\theta/\sin^2\theta_{min}$	$(\sin^2\theta/\sin^2\theta_{min})*3$	$h^2+k^2+l^2$	hkl
1	20.91	10.455	1	3	3	111
2	26.78	13.39	1.628612647	4.885837942	5	210
3	36.64	18.32	3.000388057	9.001164172	9	221/300
4	50	25	5.423994988	16.27198496	16	400

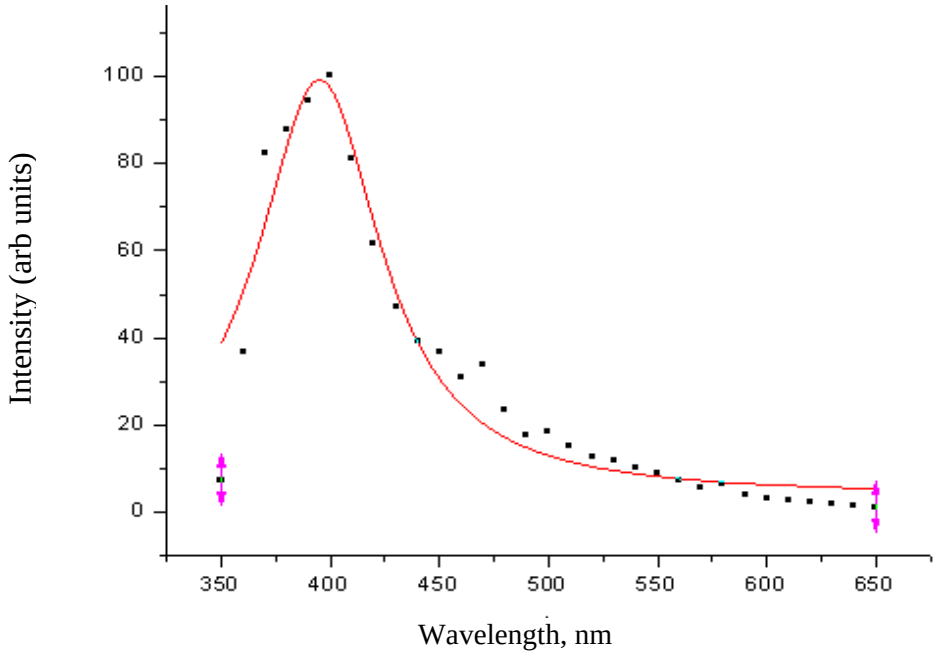

Fig. 5. PL spectra of Bhutanese stone (Biotite).

3.2. Gamma irradiated Bhutanese stone. Figure 6 shows the variation with TL glow curve as a function of gamma dose.

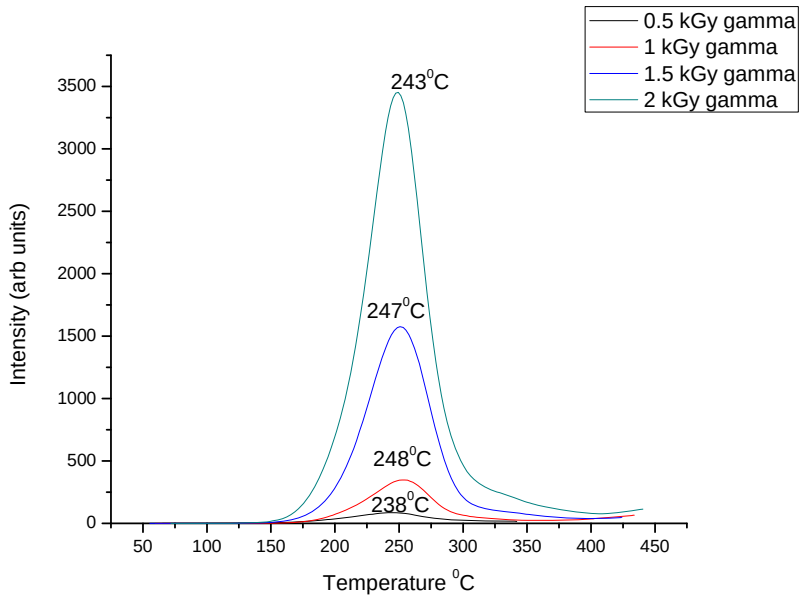

Fig. 6. Gamma irradiated Bhutanese stone as a function of gamma dose.

The sample was irradiated with Co^{60} gamma source giving a dose from 0.5 kGy to 2 kGy and the heating rate used for TL measurements was 6.7 ºC/s. The samples displayed good TL peaks at 238 ºC, 243 ºC, 244 ºC, and 246 ºC respectively. The corresponding

activation energy (E) values are calculated in Tables 2 and 3. All glow curves show the second order glow curve and the intensity increases as a function of gamma dose. It shows very good TL glow curves and the activation energy is found from 0.9 eV to 1.5 eV. The calculation was done by peak shape method. The frequency factor is found from 3×10^{10} to 1×10^{16} s^{-1} (see Table 2). Corresponding energy values (trap depth) were calculated by different methods proposed by different authors. The energy values are found between 0.1 eV and 0.8 eV (see Table 3).

Table 2. Shape factors (μ), activation energy (E) and frequency factor (S) of gamma irradiated natural Bhutanese stone collected from Bhutan.

Gamma Dose	T1	Tm	T2	τ	δ	ω	μ = δ / ω	Activation energy E, eV	Frequency factor S, s^{-1}
B7 0.5 kGy	203	238.3	273	35.32	34.68	70	0.495	0.95	3×10^{10}
B7 1 kGy	221	248	277	27	29	56	0.518	1.30	7×10^{13}
B7 1.5 kGy	219	246.4	278	27.4	31.6	59	0.536	1.28	5×10^{13}
B7 2 kGy	220	243	271	23	28	51	0.549	1.51	1×10^{16}
400 AQ 0.5 kGy	205	253	288	48	35	83	0.422	0.727	8×10^{7}
600 AQ 0.5 kGy	215	248	278	33	30	63	0.476	1.06	2×10^{11}
800 AQ 0.5 kGy	218	257	291	39	34	73	0.466	0.924	7×10^{9}

Table 3. The trap depth for the prominent glow peaks of the studied Bhutanese stone, evaluated from second order kinetics as a function of gamma dose.

Methods	0.5 kGy	1kGy	1.5kGy	2kGy
$E\ (eV)\ =\ T_m(K)/500$	0.47	0.49	0.49	0.48
$E(eV)=23KT_m$	0.47	0.49	0.48	0.48
$E(eV)=38KT_m$	0.77	0.81	0.80	0.79
$E(eV)=\frac{2KT_m^2}{\delta}$	0.27	0.36	0.32	0.36
$E_\omega=C_\omega\frac{KT_m^2}{\omega}-b_\omega(2KT_m)$	0.15	0.24	0.24	0.29
$E_\tau=C_\tau\frac{KT_m^2}{\tau}-b_\tau(2KT_m)$	0.12	0.22	0.23	0.28
$E_\delta=C_\delta\frac{KT_m^2}{\delta}-b_\delta(2KT_m)$	0.17	0.26	0.26	0.30

3.3. Chemical analysis of Bhutanese stone. Chemical analysis of our sample was done by NGRI (National Geophysics Research Institute), Hyderabad using Perkin Elmer Sciex ELAN DRC II. The method determines only tracing element of our sample through ICP-MS. The concentrations of tracing elements are shown in Table 4. The tracing elements Rb, Cu, Nd, and Nb have the major concentrations (Table 4).

3.4. Annealing quenching effect of gamma irradiated Bhutanese stone. Figure 7 shows the TL glow curve of gamma irradiated Bhutanese stone given a dose 0.5 kGy from

Co^{60} source. Heating rate used for TL measurement was 6.7 ºC/s. The powdered Bhutanese stone was annealed in air atmosphere in the temperature range 400 ºC, 600 ºC, and 800 ºC at atmospheric pressure. Samples show TL glow peaks at 254 ºC, 248 ºC, and 257 ºC respectively as a function of annealing quenching temperature. Corresponding activation energy and frequency factor are shown in Table 2. TL glow curve shows first order kinetics for AQ effect. The activation energy lies between 0.7 and 1.0 eV. The frequency factor is between 8×10^{7} and 7×10^{11} s^{-1}.

Table 4. Chemical composition of Bhutanese stone (Biotite) tracing elements.

ANALYTE	MASS	CONV. MEAN	
Sc	45	293.315	0.0293315
V	51	2588.618	0.2588618
Cr	52	550.763	0.0550763
Co	59	423.278	0.0423278
Ni	60	142.317	0.0142317
Cu	63	1892.395	0.1892395
Zn	66	850.855	0.0850855
Ga	71	445.775	0.0445775
Rb	85	24739.466	2.4739466
Sr	88	119.136	0.0119136
Y	89	1051.556	0.1051556
Zr	90	915.092	0.0915092
Nb	93	4951.254	0.4951254
Cs	133	1521.964	0.1521964
Ba	137	444.534	0.0444534
La	139	1886.374	0.1886374
Ce	140	4305.023	0.4305023
Pr	141	555.407	0.0555407
Nd	146	1730.689	0.1730689
Sm	147	350.874	0.0350874
Eu	151	13.297	0.0013297
Gd	157	308.414	0.0308414
Tb	159	40.758	0.0040758
Dy	163	332.292	0.0332292
Ho	165	49.491	0.0049491
Er	166	102.417	0.0102417
Tm	169	29.466	0.0029466
Yb	172	162.494	0.0162494
Lu	175	36.527	0.0036527
Hf	178	40.65	0.004065
Ta	181	790.906	0.0790906
Pb	208	164.515	0.0164515
Th	232	509.497	0.0509497
U	238	2.274	0.0002274
Rh	103		
Total			5.2341683

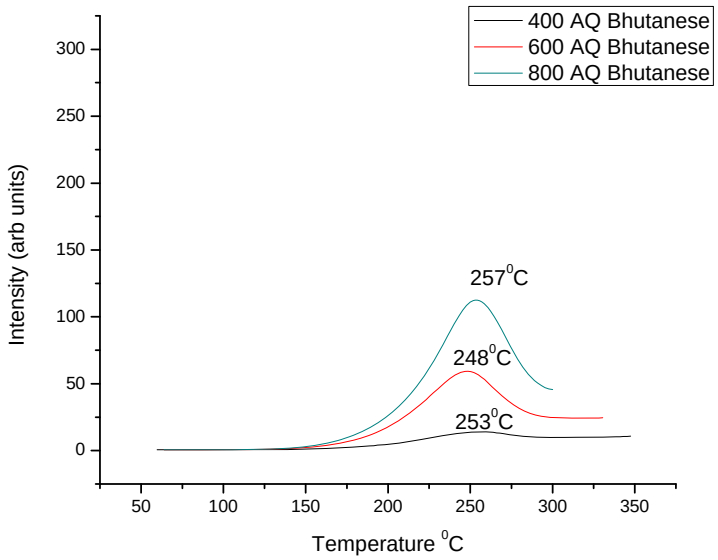

Fig. 7. Annealing effect of gamma irradiated Bhutanese stone (at dose 0.5 kGy).

Acknowledgements

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References

[1] S.W.S. McKeever, *Thermoluminescence of solids* (Cambridge University Press, Cambridge, 1985).
[2] Vikas Dubey, Jagjeet Kaur, N.S. Suryanarayana // *International Journal of Industrial Engineering and Technology* **2** (2010) 383.
[3] Vikas Dubey, N.S. Suryanarayan, Jagjeet Kaur // *Journal of Minerals & Materials Characterization & Engineering* **9** (2010) 1101.
[4] P.C. Choubey, Jagjeet Kaur, N.S. Suryanarayana, V. Dubey, G. Das // *Mater. Phys. Mech.* **10** (2010) 47.
[5] J.T. Randall, M.H.F. Wilkins // *Proc. Roy. Soc. A* **184** (1945) 366.
[6] F. Urbach // *Wiener Ber. IIa* **139** (1930) 363.
[7] L.I. Lushihik // *Sov. Phys. - JETP* **3** (1956) 390
[8] R. Chen // *J. Electrochem. Soc.* **116** (1969) 1254.
[9] R. Chen, S.W.S. McKeever, *Theory of Thermoluminescence and Related Phenomenon* (World Scientific, Singapore, New Jersey, London, Hong Kong, 1997).
[10] M. Martini, F. Meinardi // *La Rivista del Nuovo Cimento* **20 (8)** (1997) 1.
[11] M. Martini, S. Paravisi, C. Liguori // *Radiation Protection Dosimetry* **66 (4)** (1996) 447.