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Invited paper

TL-efficiency—Overview and experimental results over the years

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Abstract

For several decades, thermoluminescence detectors (TLDs) have been widely used in environmental, personal and medical dosimetry. With the development of heavy-ion accelerators, investigations concentrated on the thermoluminescent (TL) response of different TL phosphors to heavy charged particles (HCPs), motivated by the manifold potential TLD applications in HCP dosimetry. At the beginning, the research interest was focused particularly on the most common TL material, LiF:Mg, Ti. During the last years, considerable effort has been invested to determine precisely TL-efficiency also for other phosphors, either based on theoretical models or on experiments at heavy-ion accelerator facilities. The high amount of TLDs being used in space dosimetry in combination with plastic nuclear track detectors (PNTDs) essentially requires the detailed knowledge of TL-efficiency to permit dose equivalent evaluation. The paper summarizes the theoretical models currently employed to describe TL-efficiency and reviews TL-efficiency measurements from the late 1960s up to now. Difficulties in comparing TL-efficiency data from various laboratories due to different readout systems and experimental protocols will be addressed. Valuable input for a critical discussion is provided by the large pool of recent TL-efficiency data generated in the framework of the ICCHIBAN intercomparison experiment. Particularly the results obtained by the authors for ^{6}LiF:Mg, Ti (TLD-600), ^{7}LiF:Mg, Ti (TLD-700), ^{6}LiF:Mg, Cu, P (TLD-600H), ^{7}LiF:Mg, Cu, P (TLD-700H) and CaF_2:Tm (TLD-300) detectors, purchased from Thermo Fisher Scientific, Inc. (former Harshaw Chemical Co.), will be analyzed since these materials—and specific peaks in their glow curves—show distinctively different TL responses to HCP irradiation.

Keywords: Thermoluminescence detectors; Thermoluminescence efficiency; Heavy ions; TLD-700; TLD-700H; TLD-300

1. Introduction/scope

Thermoluminescence detectors (TLDs) have been widely used in the last decades for dose measurements in environmental, personal, or medical applications. With the construction of the first heavy-ion accelerator facilities, investigations primarily concerned the determination of thermoluminescent (TL) efficiency for various TLD materials to heavy ions, correlated with the question if and how TLDs can be used for heavy-ion dosimetry. At the beginning, research was mostly focused on the commonly used TL phosphor, LiF:Mg, Ti. During the last years, considerable effort has been invested by several research groups all over the world to determine precisely TL-efficiency also for other substances, either relying on theoretical models or based on experiments at heavy charged particle (HCP) accelerator facilities, such as the Heavy Ion Medical Accelerator (HIMAC) of the National Institute of Radiological Sciences (NIRS) in Chiba, Japan. One reason for these investigations is the high amount of TLDs being used in combination with plastic nuclear track detectors (PNTDs) in space dosimetry, requiring a detailed knowledge of TL-efficiency to permit dose equivalent evaluation. This review presents a brief overview of the theoretical models currently in use to describe HCP TL response followed by a historical overview of TL-efficiency measurements after heavy ion irradiations performed from the late 1960s up to now. The major results—focused particularly on ^{7}LiF:Mg, Ti—will be summarized and the problems encountered in the comparison of TL-efficiency values from various research groups due to different readout systems and experimental protocols will be discussed. Valuable input for this discussion is provided by the high amount of recent experimental data generated in the framework of the ICCHIBAN project. A focus will also be laid on results obtained during the last years by the authors themselves for TL-efficiency of TLD-600/700, TLD-600H/700H and TLD-300 detectors since

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these materials—and specific peaks in their glow curves—show distinctively different TL responses to HCP irradiation, e.g., peaks 3 and 5 in TLD-300 or peak 5 and the high-temperature structure (HTS) in TLD-600/700. These features have already been observed in the 1980s and research has been applied to use these characteristics to acquire more information from TL glow curves then "just" the absorbed dose.

2. TL-efficiency

The absolute or intrinsic TL-efficiency, α, of a phosphor is defined as the ratio of the mean energy emitted as TL light, ε_0, to the mean energy imparted to the TL material by the radiation field, ε (Kalef-Ezra and Horowitz, 1982):

$$\alpha = \frac{\varepsilon_0}{\varepsilon}. \tag{1}$$

The determination of this intrinsic efficiency is highly complex since various physical processes are involved (Bos, 2007). Therefore, research has concentrated on the determination of the relative TL-efficiency, i.e., the TL signal produced per unit dose and unit mass by the radiation under study (in our case HCPs) with respect to the TL signal per unit dose and unit mass produced by a reference radiation (mostly ^{60}Co or ^{137}Cs γ-rays). The relative TL efficiency, $\eta_{\mathrm{HCP},\gamma}$ is given by

$$\eta_{\mathrm{HCP},\gamma} = \frac{R_{\mathrm{HCP}}/D_{\mathrm{HCP}}}{R_\gamma/D_\gamma}, \tag{2}$$

where R_{HCP} and R_γ are the TL responses per unit mass for the radiation under study (HCP) and the reference radiation (γ) at dose levels D_{HCP} and D_γ, respectively. The dose levels D_{HCP} and D_γ must be sufficiently low for R_{HCP} and R_γ to fall in the linear region of the TL dose/fluence response (Horowitz, 2006).

As an example, Fig. 1 shows the glow curves of a TLD-700 detector after irradiations with 50 mGy of ^{60}Co γ-rays and 50 mGy of ^{20}Ne ions with an LET of 31.6 keV/μm. The relative TL-efficiency $\eta_{\mathrm{HCP},\gamma}$ for this ion and LET, calculated according to Eq. (2), accounts to 0.648 ± 0.016.

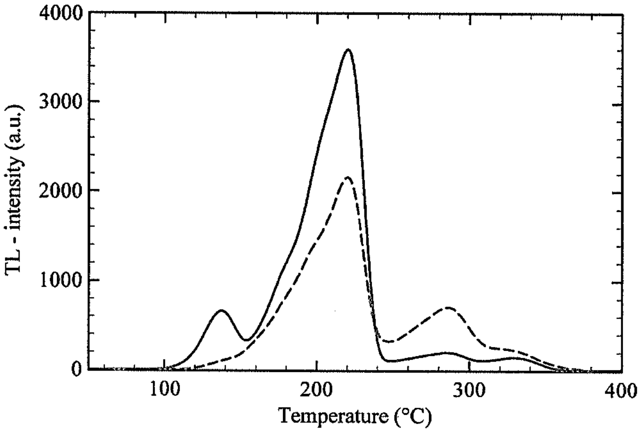

Fig. 1. TLD-700 glowcurves after 50 mGy of ^{60}Co (solid line) and 50 mGy of ^{20}Ne at 31.6 keV/μm DATA:ATI.

The scientific reason for the change in the TL-efficiency with increasing ionization density of the particle relates to the dose deposition profiles from HCPs. A slowing down HCP deposits its energy to an overhelming extent by secondary electrons (δ electrons) created in the slowing down process. Depending on the charge and the velocity of the ion, the energies and ranges of these secondary electrons vary greatly. If the γ response of a TL detector system is linear-sublinear, part of the dose around the heavy-ion track is deposited in the sublinear and part in the linear region of the dose response. Therefore, TL-efficiency for heavy ions in TL detectors with a linear-sublinear dose response (as for example LiF:Mg, Cu, P) can never exceed 1. On the contrary, for TL materials with a linear–supralinear–sublinear dose response (as for peak 5 in LiF:Mg, Ti) parts of the doses are deposited in all three regions of the dose response and a relative TL-efficieny > 1 can be observed for these detector materials.

2.1. Intercomparison of relative TL-efficiency

One focal point of this paper is the intercomparison of relative TL-efficiency data obtained from various research groups and published over more than three decades. It was pointed out frequently (Ávila et al., 2002; Horowitz et al., 2006) that an intercomparison of data provided by different laboratories is complicated and could be hampered due to the different experimental protocols applied by the investigators. The following aspects have to be taken into account if someone aims to compare data from the literature and tries to compile it:

- TL material (manufacturer, batch, form);
- reference radiation (^{137}Cs, ^{60}Co, X-rays);
- TLD calibration (single-chip, batch calibration);
- readout system (photocathode spectral response, optical filters, method of heating);
- heating rate;
- annealing cycle (time and temperature profiles);
- method of data evaluation (deconvolution into single peaks, region of interest, height of composite peak).

Nevertheless—as will be shown later—it turns out that intercomparison is possible and even leads to very good agreement between the various groups, if the above mentioned differences in protocols are taken into account carefully. The authors also want to stress the fact, that even if an intercomparsion does not agree perfectly—the "individual" data set from the "individual" research group gives the true values for the "individual" protocol applied by this group, thereby stating, that a certain disagreement between the experimental data from different laboratories does not automatically invalidate particular measurements.

3. Theoretical models

Three major different models/theories have been developed over the years to describe the relative TL response of different materials:

- track structure theory (TST; Waligórski and Katz, 1980a, b);
- modified track structure theory (MTST; Kalef-Ezra and Horowitz, 1982);
- microdosimetric target theory (MTT; Olko et al., 2002).

The value of relative TL-efficiency is strongly related to the structure of the deposited energy of ionizing radiation, in particular to the spatial distribution of ionization and excitations produced by the ionizing particles along their track. The models describing $\eta_{HCP,\gamma}$ in a qualitative and partly also quantitative good way are mostly based on the concept of radial distribution of dose around the ion tracks (Waligórski and Katz, 1980a, b; Kalef-Ezra and Horowitz, 1982). The microdosimetric approach (Olko et al., 2002) relates the distribution of energy deposited by ionizing radiation within a specified volume (representing sensitive targets of nanometer or micrometer dimensions in the detector) to the observable effect (Olko, 2004).

Calculations based on these models have been performed for various TL materials (e.g., LiF:Mg, Ti, LiF:Mg, Cu, P etc.) over the years (Waligórski and Katz, 1980a, b; Ávila et al., 2006; Massillon-JL et al., 2006a, b, Olko, 2004, 2007, Olko et al., 2002, 2004; Geiß et al., 1998a, b). For a detailed review and discussion of the models the reader is referred to Horowitz and Olko (2004), Horowitz et al. (2006) and Olko (2004, 2007). The theories are able to calculate for a variety of particle species the relative TL-efficiency of several TL phosphors in reasonable qualitative and partly quantitative agreement with experimental data. Nevertheless, they have, however, yet to be confronted with sufficiently reliable experimental data to test the extent and range of their validity (Horowitz et al., 2006).

4. Historical overview

The authors want to emphasize that an overview of the history of experimental determination of TL-efficiency can only try to be complete in the sense of showing how our understanding and knowledge of TL-efficiency behaviour after HCP irradiation evolved over the years.

TLDs have been used since the early 1950s (Daniels et al., 1953) for dose determination in medical and scientific applications. The increasing utilization of TLDs in dosimetry demanded research in the properties of the materials. One aim in these studies was the determination of the relative TL-efficiency after proton, alpha particle and heavy-ion irradiation. This search for knowledge—especially for the HCPs—could only be initiated after the construction of the first heavy particle accelerators in the 1960s. Essential contributions were provided by investigations at the Heavy Ion Linear Accelerator (HILAC) facility of the Lawrence Berkeley National Laboratory (LBNL). Tochilin and Goldstein (1968), Patrick et al. (1975, 1976) and Henson and Thomas (1978) used the C, O, Ne and Ar ions at the HILAC and the BEVALAC (a combination of the Bevatron and the SuperHILAC) for their measurements employing LiF:Mg, Ti. Patrick et al. (1976) strongly suggested that $\eta_{HCP,\gamma}$ may not be a function of LET alone but also depend upon the charge of the particle depositing energy in the crystal. Benton et al. (2000) summarized the results from the Berkeley experiments including his own investigations. Jähnert (1972) exposed LiF:Mg, Ti to low-energy protons and observed that the intensity of the high-temperature peaks relative to the whole TL emission increases with increasing LET of the ionizing particle. Investigations for protons of higher energies were conducted by Mukherjee (1980), Hübner et al. (1980) and Schmidt et al. (1988, 1990). Hübner et al. (1980) also used low-energy C and O ions from the cyclotron of the Joint Institute for Nuclear Research (JINR) in Dubna, Russia. Low-energy Ne and Kr ions from the Linear Accelerator (LINAC) in Manchester were applied by Faïn and Montret (1980). From his data for LiF:Mg, Ti it could already be concluded that efficiency is not a unique parameter of LET alone but rather depends on particle species. Hoffmann and Prediger, 1983 and Hoffmann (1996) investigated the response of LiF:Mg, Ti and CaF_2:Tm detectors using the accelerators at LBNL in Berkeley, CA, USA. CaF_2:Tm was investigated as well by Loncol et al. (1996) and Buenfil et al. (1999),motivated by the fact that this material—in a similar way as LiF:Mg, Ti—shows two glow peaks with very different LET dependence so that it could be used as a two-parameter TL detector system. A lot of investigations were dedicated to the measurement of the TL-efficiency to α particle radiation which have been summarized in part and compared by Bartlett and Edwards (1979) and Barber and Ahmed (1986). A comparison of α particle TL-efficiency is complicated due to the very short range of these low energetic particles and demands high accuracy in determining the energy of the impinging α particle.

Already in 1981, Horowitz (1981) summed up the state of the knowledge of TL-efficiency for LiF:Mg, Ti, taking into account the available data from the literature. He further emphasized the non-existence of a unique relationship between TL-efficiency and LET for that particular phosphor. This summary is also contained—with more recent data (Geiß et al., 1998a, b; Ávila et al., 1999, 2006) added—in Horowitz (2006). While Ávila et al. (2006) present a summary of their results for low energetic protons, He, C, N and O ions, Geiß et al. (1998a, b) include data for C ions from 2 to 270 MeV/u. An overview of TL-efficiency data collected till the mid 1990s can also be found in Geiß (1997).

In the 1990s, TL response of the new developed highly sensitive TL material LiF:Mg, Cu, P to α particles (Horowitz and Stern, 1990; Bilski et al., 1994) and protons (Bilski et al., 1997) from the JINR, Dubna, Russia, has been investigated.

A lot of effort has been invested over the last 40 years into the determination of the relative TL-efficiency, especially for the most common used material, LiF:Mg, Ti, but also for other phosphors, such as LiF:Mg, Cu, P and CaF_2:Tm. Due to the non-availability of high-energy particle accelerators most data have been recorded at comparatively low energies—sometimes even for particles which stopped in the detector volume. For two reasons, the demand for data particularly in the high-energy region of a few hundred MeV/u increased over the years: (i) the high number of heavy ion therapy facilities opened the possibility to use TLDs for dose determination or dose control, (ii) the increased presence of humans in space required a low-weight and robust monitoring device capable of assessing dose equivalent which could be provided by combination of TLDs and PNTDs. Since the energy range of space radiation ranges up to a few GeV/u and its spectral composition extends from protons to Fe nuclei of great biological relevance, determination of TL-efficiency is crucial to the performance of the detectors.

5. Recent investigations in TL-efficiency

The intention of this chapter is to summarize and compare TL-efficiency data for several TL materials, in particular LiF:Mg, Ti, LiF:Mg, Cu, P and CaF_2:Tm, gathered by different research groups at heavy ion accelerators over the last 10 years. As long as not stated otherwise, data from the German Aerospace Center (DLR), Cologne, Germany, and the Atomic Institute of the Austrian Universities (ATI), Vienna, Austria, have been obtained in the framework of a joint research project (16P169) at NIRS-HIMAC during the years 2002–2007.

Table 1 summarizes the laboratory procedures, such as reference radiation, heating cycles, measurement systems etc.; applied by DLR and ATI in the framework of this research project. Data evaluation was based on analysis of composite peak height.

Table 2 lists the applied ions, their primary energies, their LET and their range in water. All the irradiations were performed aiming for almost no decrease in ion energy over the thickness of the crystal. Information about the dose control, beam quality and beam uniformity of the ion beams at HIMAC can be found in Uchihori and Benton (2004).

Table 1
Parameters for data read out and data evaluation ATI, DLR

Parameter	ATI	DLR
TL reader	TL-DAT. II (Vana et al., 1988)	Harshaw 5500
PMT	Thorn EMI 9635 QB	Hamamatsu RC095 HA
Nitrogen purity	5.0	5.0
Heating rate	5 °C/s	5 °C/s
Pre-heat		
CaF_2: Tm	No preheat	No preheat
LiF:Mg, Ti	No preheat	No preheat
LiF:Mg, Cu, P	150 °C (10 s)	150 °C (10 s)
Maximum temperature		
CaF_2 : Tm	400 °C	400 °C
LiF:Mg, Ti	480 °C	400 °C
LiF:Mg, Cu, P	240 °C	240 °C
Annealing cycle		
CaF_2: Tm	400 °C (1.5 h)	400 °C (1 h), 100 °C (2 h)
LiF:Mg, Ti	400 °C (1 h)	400 °C (1 h),100 °C (2 h)
LiF:Mg, Cu, P	240 °C (10 min)	240 °C (10 min)
Cooling rate		
CaF_2: Tm	Slow	Slow
LiF:Mg, Ti	Slow	Slow
LiF:Mg, Cu, P	Fast (~ 2 min)	Fast (~ 2 min)
Calibration	Single-chip	Single-chip
Reference radiation	^{60}Co γ-rays	^{137}Cs γ-rays

Table 2
Ion species, energies, LET and range in water applied for the investigations (ion range calculated by means of the SRIM/TRIM 2006 Monte Carlo code, Ziegler et al., 1985, http://www.srim.org)

Ion	Primary energy (MeV/u)	LET$_\infty$ H_2O (keV/μm)	Range in H_2O (mm)
$^{4}He^{2+}$	150	2.2	156.5
$^{12}C^{6+}$	290	12.7	164.5
	400	10.7	278.7
$^{16}O^{8+}$	400	19.4	205.2
$^{20}Ne^{10+}$	400	30.1	165.2
$^{28}Si^{14+}$	490	54.5	160.8
$^{40}Ar^{18+}$	500	92.3	138.8
$^{56}Fe^{26+}$	500	182.6	98.5
$^{84}Kr^{36+}$	400	358.7	75.9
$^{132}Xe^{54+}$	290	1051	22.6

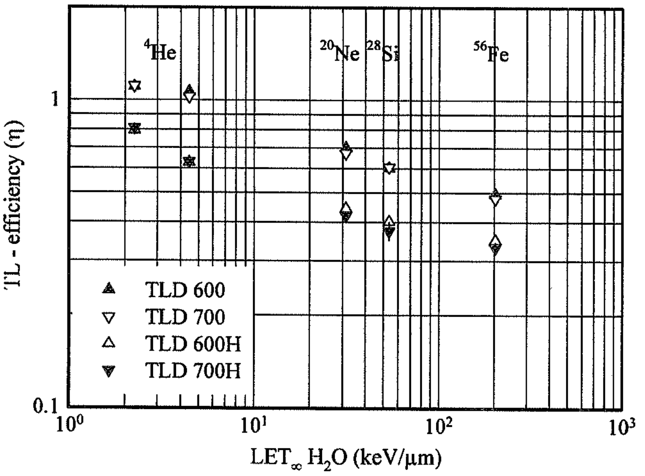

Fig. 2. Peak 5 TL-efficiency of TLD-600 (full triangles up) and TLD-700 (open triangles down) as well as for TLD-600H (open triangles up) and TLD-700H (full triangles down) after irradiation with ^{4}He, ^{20}Ne, ^{28}Si and ^{56}Fe ions. DATA:DLR.

The investigations have mainly been focused on TLD-700, TLD-700H as well as TLD-300. Data have also been obtained for TLD-600 and TLD-600H. As can be seen in Fig. 2, TL-efficiency of LiF:Mg, Ti and LiF:Mg, Cu, P to heavy ions does not depend on the abundance of the different Li isotopes in the phosphor, as long as a possible neutron contribution to the total dose, e.g., during air transport of the detectors, is considered properly.

5.1. *LiF:Mg, Ti—the main peak 5*

As shown in Fig. 1, the glow curve of LiF:Mg, Ti comprises several peaks (e.g., the main peak 5 and the high temperature peaks). During the last four decades, research was focused primarily on the main-dosimetry peak 5. As forecasted by theoretical models (Waligórski and Katz, 1980a, b; Horowitz, 1981) and confirmed experimentally for example by Berger (2003) and Geiß (1997), peak 5 TL-efficiency in LiF:Mg, Ti does not

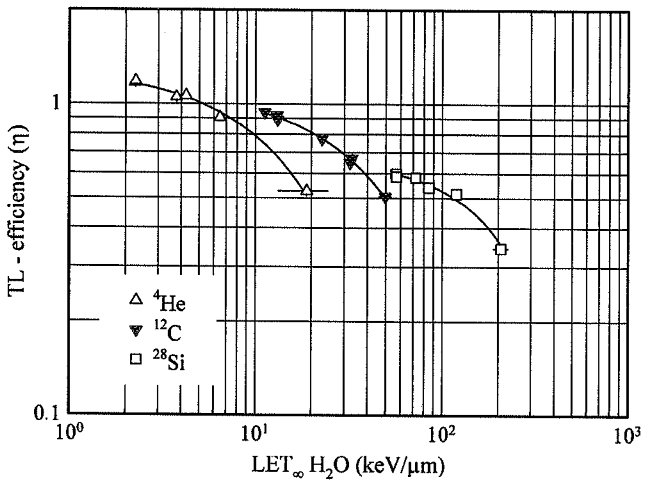

Fig. 3. Peak 5 TL-efficiency of TLD-700 for three different ion species (^{4}He, ^{12}C, ^{28}Si), adapted from: Berger et al. (2006a).

depend on the LET alone, but also on the particle species. This implies that for the same LET of different particles different TL-efficiencies will be observed. This fact is further illustrated in Fig. 3, showing TL-efficiency of TLD-700 detectors after irradiations with $^4He^{2+}$ (2–20 keV/μm), $^{12}C^{6+}$ (11–50 keV/μm) and $^{28}Si^{14+}$ (50–200 keV/μm) ions.

The physical explanation for the LET and ion dependence of TL-efficiency is given by the fact that, for the same LET, the velocity of a particle with greater charge must be higher which leads to lower local ionization density and, consequently, to a higher value of $\eta_{HCP,\gamma}$ (Olko, 2007).

Table 3 shows comprehensive data for the peak 5 TL-efficiency of TLD-700 detectors collected by ATI and DLR (Berger, 2003; Berger et al., 2006a, b; Hajek et al., 2006). Though both groups used detectors from different batches, applied different readout systems and annealing cycles, the agreement between the two data sets is very good.

The main purpose of this research was to acquire a comprehensive data set of TLD-700 TL-efficiency to apply this knowledge to the determination of dose equivalent from space radiation, using a combination of TLDs and PNTDs. The TLDs act as low-LET detectors, while the CR-39 PNTDs assess the high-LET (> 10 keV/μm) part of the space radiation spectrum (as example see Reitz, 1994; Benton and Benton, 2001). To subtract the high-LET contribution from the dose measured by TLDs, knowledge of the TL-efficiency is essential.

The critical reader will certainly stress the question if the TL-efficiency function determined for a certain batch can readily be used for a different batch from the same manufacturer. Batch dependence of TL-efficiency was seen as one of the major problems for data intercomparison. DLR has exposed two different TL batches (S-4678, S-4762 I and II; the latter purchased over a 6 months time period) to HCPs from the HIMAC. Table 4 compares TL-efficiency data for these batches to prove that the values do not vary from batch to batch (S-4678, S-4762), nor within detectors purchased from the same batch over a certain period of time (S-4762 I and II).

Table 3
Relative TL-efficiency for the main peak 5 of TLD-700 (data: ATI, DLR)

Ion	LET_∞ H_2O (keV/μm)	$\eta_{HCP,\gamma}$ (ATI)	$\eta_{HCP,\gamma}$ (DLR)
$^4He^{2+}$	2.3 ± 0.0	1.174 ± 0.020	1.111 ± 0.018
	3.6 ± 0.0	1.050 ± 0.040	1.029 ± 0.019
	4.3 ± 0.1	1.061 ± 0.024	1.031 ± 0.019
	6.2 ± 0.2	0.905 ± 0.023	0.941 ± 0.029
	8.0 ± 0.9		0.855 ± 0.021
	11.02 ± 1.3		0.704 ± 0.036
	18.9 ± 5.6	0.526 ± 0.019	
$^{12}C^{6+}$	11.2 ± 0.0	0.940 ± 0.020	
	12.5 ± 0.1	0.890 ± 0.005	0.820 ± 0.040
	23.5 ± 0.3	0.773 ± 0.022	0.698 ± 0.050
	32.5 ± 0.9	0.660 ± 0.030	0.622 ± 0.030
	49.6 ± 3.6	0.505 ± 0.007	0.515 ± 0.020
$^{16}O^{8+}$	19.8 ± 0.0		0.760 ± 0.002
	22.4 ± 0.1		0.681 ± 0.041
	39.4 ± 0.6		0.612 ± 0.018
	47.4 ± 1.1		0.550 ± 0.015
	66.0 ± 3.6		0.467 ± 0.011
$^{20}Ne^{10+}$	31.6 ± 0.0	0.648 ± 0.016	0.649 ± 0.010
	44.8 ± 0.3	0.637 ± 0.008	0.572 ± 0.005
	55.4 ± 0.7	0.559 ± 0.012	0.540 ± 0.007
	76.3 ± 2.5		0.496 ± 0.009
	91.8 ± 4.7		0.464 ± 0.007
	121.2 ± 11.7		0.398 ± 0.010
$^{28}Si^{14+}$	56.3 ± 0.0	0.559 ± 0.020	0.560 ± 0.040
	72.2 ± 0.4	0.581 ± 0.013	
	85.0 ± 0.8	0.540 ± 0.005	
	119.5 ± 2.5	0.516 ± 0.012	
	207.0 ± 17.9	0.344 ± 0.008	
$^{40}Ar^{18+}$	96.2 ± 0.2	0.497 ± 0.018	0.495 ± 0.030
	109.5 ± 0.5	0.504 ± 0.004	
	120.7 ± 0.7	0.501 ± 0.022	
	140.5 ± 1.3	0.493 ± 0.002	
	158.1 ± 2.1	0.491 ± 0.023	0.481 ± 0.030
$^{56}Fe^{26+}$	198.4 ± 0.4	0.491 ± 0.007	
	201.6 ± 0.8	0.473 ± 0.016	0.462 ± 0.005
	266.8 ± 3.1	0.438 ± 0.007	
	309.6 ± 6.8	0.430 ± 0.013	
	333.9 ± 7.6	0.425 ± 0.012	0.410 ± 0.040
	400.7 ± 14.5	0.444 ± 0.018	
	413.8 ± 16.3	0.398 ± 0.006	0.380 ± 0.030
$^{84}Kr^{36+}$	448.6 ± 0.5	0.460 ± 0.006	0.420 ± 0.040
$^{132}Xe^{54+}$	1365.0 ± 0.0		0.373 ± 0.031
	1913.0 ± 175		0.340 ± 0.014
	2651.0 ± 508		0.301 ± 0.013

Based on these findings, we now compare TL-efficiencies obtained for TLD-100 (Massillon-JL et al., 2007—data for ^{3}He), TLD-700 (DLR; ATI; Patrick et al., 1975; Geiß et al., 1998a) and MTS-7 (Bilski, 2006) exposed to helium and carbon ions of energies ranging from a few to a few hundred MeV/u. This LET interval covers practically the entire range of ionization density which is available for He and C ions in ground-based experiments. The combined data are shown in Fig. 4(a)for He and in Fig. 4(b) for C ions. It has to be noted, that while most

Table 4
Batch dependence of relative TL-efficiency for peak 5 of TLD-700 (data:DLR)

Ion	LET_∞ H_2O (keV/μm)	$\eta_{HCP,\gamma}$ (S-4678)	$\eta_{HCP,\gamma}$ (S-4762 I)	$\eta_{HCP,\gamma}$ (S-4762 II)
$^4He^{2+}$	2.3 ± 0.0	1.111 ± 0.018	1.126 ± 0.020	1.116 ± 0.002
	4.3 ± 0.1	1.031 ± 0.019		1.026 ± 0.021
$^{20}Ne^{10+}$	31.6 ± 0.0	0.670 ± 0.009	0.655 ± 0.015	0.672 ± 0.09
	55.4 ± 0.7	0.549 ± 0.005		0.580 ± 0.10
$^{56}Fe^{26+}$	201.6 ± 0.8	0.450 ± 0.008	0.455 ± 0.007	0.472 ± 0.011

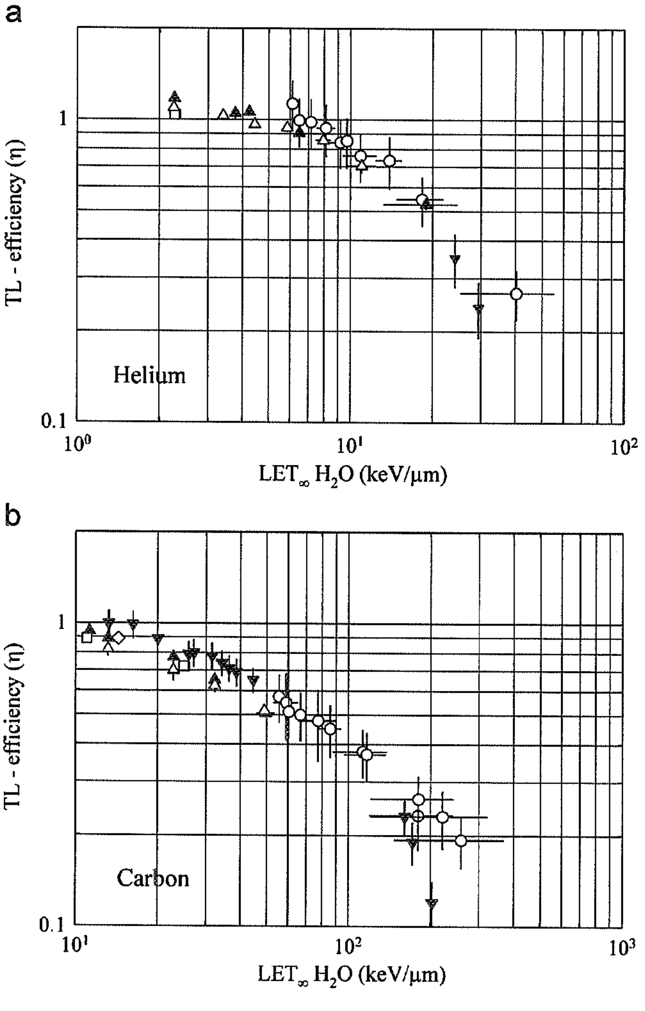

Fig. 4. (a) and (b) TL-efficiency intercomparison for helium (2–40 keV/μm) and carbon (10–250 keV/μm) ions. Full triangles up: ATI; open triangles up: DLR; full triangles down: Geiß et al. (1998a); squares: Bilski (2006); circles: Massillon-JL et al. (2006a, 2007); hex:Patrick et al. (1975).

of the groups applied ^{60}Co or ^{137}Cs γ as reference radiation data from Geiß et al. (1998a) is based on 250 keV X-rays. Despite the fact that detectors from different batches and manufacturers are compared and the methods used by the individual laboratories for TL readout and analysis (peak height, deconvolution) are not identical, the data agree very well—especially for the high energy parts—within thegiven uncertainties.

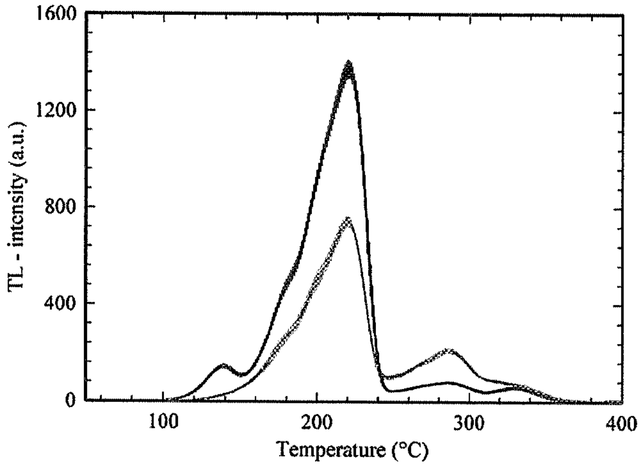

Fig. 5. TLD-700 glow curves after 1 mGy of ^{60}Co γ-rays (black) and 1 mGy of ^{56}Fe at 202 keV/μm (grey). The presented data are averaged over 6 chips and include error bars. DATA:ATI.

5.2. *LiF:Mg, Ti—the high temperature peaks*

It was known from the early 1970s (e.g., Jähnert, 1972) that the high-temperature peaks in LiF:Mg, Ti show a very different LET dependence than the main peak 5. This behaviour of the high-temperature peaks can be attributed to the earlier onset of supralinearity compared to peak 5. Hoffmann and Prediger (1983) stated that LiF:Mg, Ti therefore gives a two-parameter signal from which dose and LET can be derived. The relative TL-efficiency of the high-temperature peaks with respect to ^{60}Co was already calculated by Waligórski and Katz (1980a, b), showing that levels far exceeding 1 can be reached. They concluded that the high-temperature peaks in LiF:Mg, Ti would be good candidates for mimicking the response of biological systems to heavy-ion irradiations.

For experimental purposes, the following aspects need to be considered in analyzing the high-temperature peaks in LiF:Mg, Ti.

(i) It is essential to perform a reproducible background subtraction. The background, hereby, refers to the signal from an unirradiated detector, e.g., the increasing black-body radiation and the signal induced by the readout system (electronic noise etc). Fig. 5 shows average glow curves after irradiations with 1 mGy of ^{60}Co γ-rays and 1 mGy of $^{56}Fe^{26+}$ ions of 202 keV/μm. Background subtraction has been performed both by measuring each chip a second time and by applying a mathematical fit to the black-body radiation. As can be seen from Fig. 5, the high-temperature peaks are clearly resolved even at very low doses of 1 mGy for both low- and high-LET radiation.

(ii) If TL-efficiency is determined either from the deconvoluted high-temperature peaks or the composite HTS, exposures have to be performed in the linear part of the γ-ray dose response of the high-temperature peaks. The discussion whether a linear dose response can be found is still ongoing. A linear dose response in the applied dose region for the TL-efficiency studies has been measured by Pradhan et al. (1985) (Fig. 3) and by Massillon-JL et al. (2006a) (Fig. 3). Horowitz et al. (2007) measured a pronounced supralinearity even at very low doses (see Fig. 6 in Horowitz et al., 2007), concluding from his experiments that the HTS from LiF:Mg, Ti is barely observable at low dose levels of around 10 mGy (Fig. 7 in Horowitz et al., 2007). However, he stated that "there may be measurement protocols and batches for which a linear region exists and other protocols and batches for which no linear region exists". In the framework of the ICCHIBAN project, the relative HTS to peak 5 ratio with respect to ^{60}Co was evaluated for HCP doses between 1 and 100 mGy. The results indicate an entirely dose-independent behaviour in this dose region (Uchihori and Benton, 2004; Hajek et al., 2006). The same dose-independence was observed for the low-dose region by Bilski (2006, see Fig. 4), and by Yasuda and Fujitaka (2000), Yasuda (2001). Further studies by Berger and Hajek (2007) confirmed the linear HTS dose response after ^{60}Co γ-rays in the dose region between 2 and 95 mGy for their applied readout and annealing procedures. The main differences in the HTS are related to the different annealing procedures of slow and fast cooling employed by the various groups—showing a decrease of the high temperature emission by increasing cooling rate. Therefore the subtraction of the background signal becomes more crucial. It shall be emphasized that our knowledge of the high-temperature emission in LiF:Mg, Ti is far from being complete and further studies need to be performed using both the HTS and the deconvoluted glow peaks in this region.

Figs. 6(a)–(c) show a compilation of the TL-efficiency for peak 5 and the high-temperature peaks in LiF:Mg, Ti. For this compilation, numerical values have been used from Yasuda and Fujitaka (2000), Yasuda (2001), Berger (2003), Berger et al. (2006a), Hajek et al. (2006) and Massillon-JL et al. (2006b, 2007). While the data in Figs. 6(a) and (b) were assessed from the HTS around peak 7, data from Massillon-JL et al. (2006b, 2007) are based on the deconvoluted peak 7. Nevertheless, the overall tendency is the same for all three figures: the TL-efficiency for the high-temperature peaks shows a strong correlation with LET and particle species. LET is not the only parameter governing TL-efficiency for the HTS as well as peak 5.

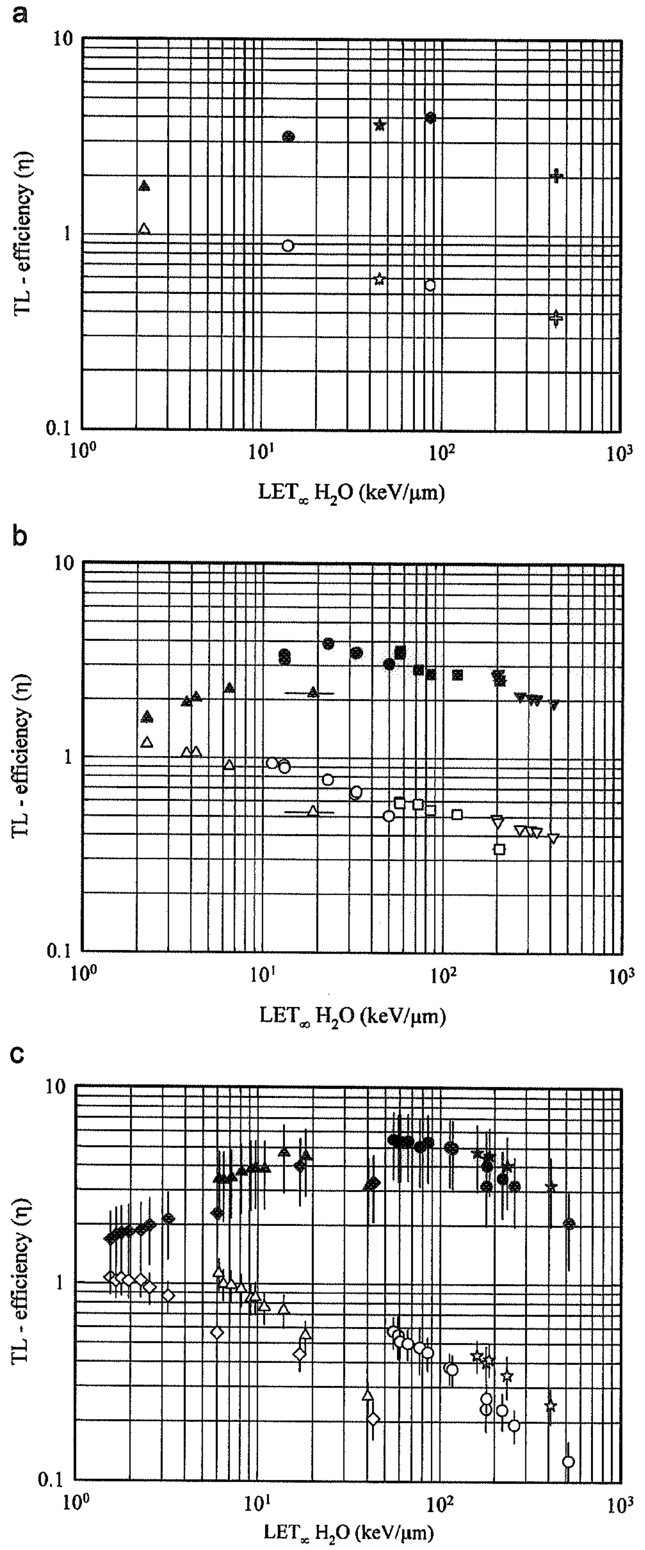

Fig. 6. (a)–(c) TL-efficiency for peak 5 (open symbols) and the high-temperature peaks (full symbols) in LiF:Mg, Ti. Adapted from: (a) Yasuda and Fujitaka (2000); (b) Berger et al. (2006a), Hajek et al. (2006) and (c) Massillon-JL et al. (2007). Diamond: ^1H; triangle up: ^4He /(^3He for Massillon-JL et al., 2007); circle: ^{12}C; star: ^{20}Ne; square: ^{28}Si; hex: ^{40}Ar; triangle down: ^{58}Fe; cross: ^{96}Kr.

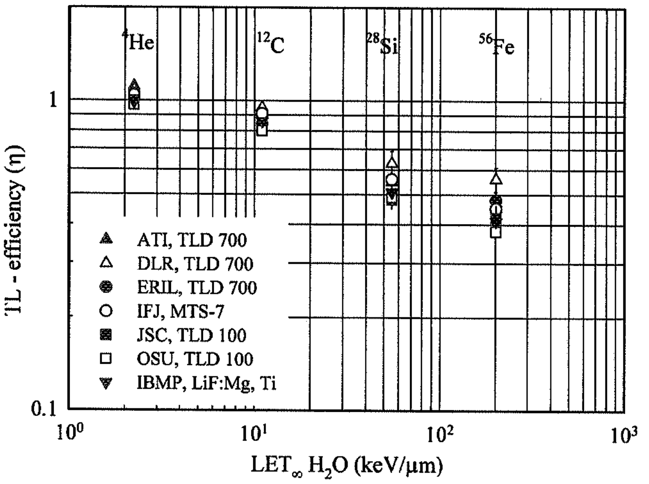

Fig. 7. Peak 5 TL-efficiency for various LiF:Mg, Ti materials irradiated under the same conditions during the ICCHIBAN-2 project (adapted from Uchihori and Benton, 2004). ATI—Atomic Institute of the Austrian Universities, Vienna, Austria; DLR—German Aerospace Center, Cologne, Germany, ERIL—Eril Research, San Francisco, USA, IFJ—Institute of Nuclear Physics, Krakow, Poland (MTS-7 ^{7}LiF:Mg, Ti), JSC—Johnson Space Center, Houston, USA; OSU—Oklahoma State University, Stillwater, USA, IBMP—Institute for Biomedical Problems, Moscow, Russia (Natural LiF:Mg, Ti - equivalent to TLD-100).

5.3. *LiF:Mg, Ti—results from the 2nd ICCHIBAN experiment*

The ICCHIBAN project (InterComparison of Dosimetric Instruments for Cosmic Radiation with Heavy Ion Beams at the National Institute for Radiological Sciences in Chiba, Japan) was initiated at the 4th Workshop on Radiation Monitoring for the International Space Station (WRMISS) in Farnborough, United Kingdom (Yasuda et al., 2006). The space dosimetry community deemed it necessary to perform an intercalibration of space radiation monitoring devices in terms of particle detection efficiency and other dosimetric properties. The first ICCHIBAN run for passive detectors (ICCHIBAN-2) was held at HIMAC in May 2002. Passive detectors from all participating groups were irradiated under the same irradiations conditions, enabling the detailed intercomparison of TL-efficiency data obtained by the different groups.

The detailed readout and data evaluation procedures applied by the individual laboratories have been reported in Uchihori and Benton (2004). Further results of the ICCHIBAN irradiations can be found in Berger et al. (2006c), Bilski (2006), Gaza et al. (2004), Spurný (2004) and Yukihara et al. (2004, 2006). Fig. 7 shows the compilation of TL-efficiency data for LiF:Mg, Ti detectors (TLD-100, TLD-700, MTS-7 and natural LiF:Mg, Ti) from seven different laboratories, each applying its own experimental protocol for readout and analysis. A closer look on the combined TL-efficiency values reveals an average TL-efficiency of 1.034 ± 0.047 for He and 0.451 ± 0.054 for Fe, respectively. The statistical uncertainties are ±4.5% for He and ±11.8% for Fe.

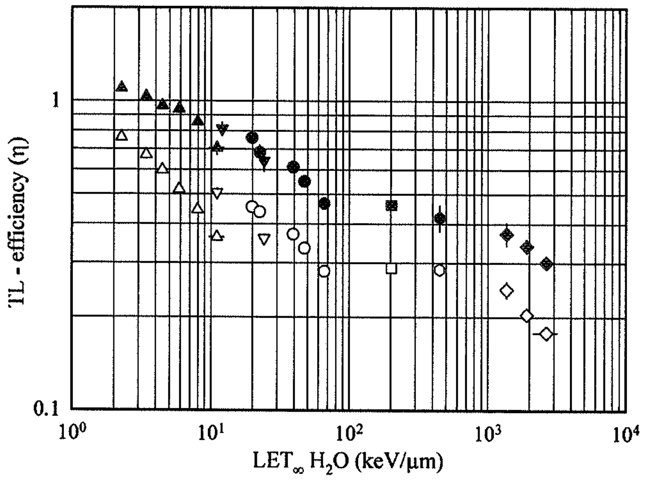

Fig. 8. TL-efficiency for peak 5 in TLD-700 (solid symbols) and TLD-700H (open symbols). Triangle up: ^{4}He; triangle down: ^{12}C; circle: ^{16}O; square: ^{56}Fe, hex: ^{96}Kr; diamond: ^{136}Xe DATA: DLR.

5.4. *LiF:Mg, Cu, P*

Application of LiF:Mg, Cu, P is mostly concentrated on environmental dosimetry due to its very high sensitivity compared to LiF:Mg, Ti. LiF:Mg, Cu, P is an interesting TL material as its main peak shows a linear-sublinear γ-ray response, which can be expected to result in very low TL-efficiency after heavy-ion bombardment. Only a limited amount of investigations have been performed up to now regarding the experimental determination of TL-efficiency for this material. Also, assessment has been restricted to protons and α particles (Horowitz and Stern, 1990; Bilski et al., 1994, 1997). TL-efficiency calculations have been performed for MCP-N (LiF:Mg, Cu,P) detectors by Olko et al. (2002, 2004) and Olko (2004, 2007).

In the framework of the authors' HIMAC research project the different behaviour of TL-efficiency for TLD-700 and TLD-700H was investigated by DLR for a wide range of ions and energies. Results are shown in Fig. 8 for TLD-700 (solid symbols) and TLD-700H (open symbols), respectively. The previous argument that TL-efficiency is not governed by LET alone can also be applied to TLD-700H detectors. By combination of TLD-700 and TLD-700H, a two-detector system is available which gives very different dose readings for the same LET and can therefore be used to obtain more information about the radiation field under study (Berger et al., 2006c; Olko et al., 2002).

Table 5 gives an intercomparison of TL-efficiency data for TLD-700H and MCP-7 detectors as determined by ATI (Berger et al., 2006a), DLR and IFJ (Bilski, 2006).

5.5. *CaF$_2$:Tm*

CaF$_2$:Tm phosphors have been investigated in the last 30 years by various research groups due to the very distinct TL-efficiency behaviour of their main glow peaks 3 and 5 (Hoffmann and Möller, 1980; Hoffmann and Prediger, 1983; Hoffmann, 1996; Loncol et al., 1996; Buenfil et al., 1999).

Table 5
Relative TL-efficiency for LiF:Mg, Cu, P (TLD-700H and MCP-7) as determined by IFJ (Bilski, 2006), ATI (Berger et al., 2006a) and DLR

Ion	LET$_\infty$ H$_2$O (keV/μm)	$\eta_{\mathrm{HCP},\gamma}$ (Bilski) (MCP-7)	$\eta_{\mathrm{HCP},\gamma}$ (DLR) (TLD-700H)	$\eta_{\mathrm{HCP},\gamma}$ (ATI) (TLD-700H)
^{4}He^{2+}	2.3 ± 0.0	0.771 ± 0.026	0.762 ± 0.008	0.760 ± 0.050
^{12}C^{6+}	11.2 ± 0.0	0.507 ± 0.017	0.489 ± 0.026	0.410 ± 0.020
^{16}O^{8+}	19.8 ± 0.0		0.454 ± 0.026	
^{20}Ne^{10+}	31.6 ± 0.0	0.369 ± 0.013	0.347 ± 0.025	
^{28}Si^{14+}	56.3 ± 0.0	0.313 ± 0.011	0.295 ± 0.015	0.280 ± 0.030
^{40}Ar^{18+}	96.2 ± 0.2	0.311 ± 0.009	0.321 ± 0.030	
^{56}Fe^{26+}	201.6 ± 0.8	0.288 ± 0.010	0.256 ± 0.015	0.270 ± 0.010

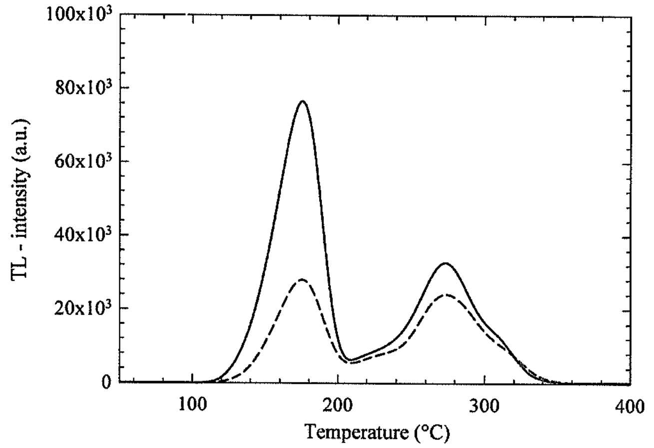

Fig. 9. Glowcurve of TLD-300 after 100 mGy of ^{137}Cs γ-rays (solid line) and 100 mGy of ^{56}Fe (202 keV/μm) (dashed line).

Table 6
Relative TL-efficiency for peak 3 in CaF$_2$: Tm; data are obtained from analysis of the composite peak height

Ion	LET$_\infty$ H$_2$O (keV/μm)	$\eta_{\mathrm{HCP},\gamma}$ peak 3 (ATI)	$\eta_{\mathrm{HCP},\gamma}$ peak 3 (DLR)
^{4}He^{2+}	2.3 ± 0.0	0.724 ± 0.112	0.699 ± 0.012
	4.5 ± 0.1	0.548 ± 0.056	0.626 ± 0.011
^{16}O^{8+}	19.9 ± 0.0		0.527 ± 0.017
^{20}Ne^{10+}	31.5 ± 0.1	0.468 ± 0.044	0.444 ± 0.015
	54.2 ± 1.1	0.419 ± 0.041	0.402 ± 0.014
^{40}Ar^{18+}	89.5 ± 0.2		0.349 ± 0.008
^{56}Fe^{26+}	202.0 ± 0.9	0.351 ± 0.038	0.350 ± 0.010
	312.1 ± 7.2		0.287 ± 0.009

As an example, Fig. 9 shows glow curves from CaF$_2$: Tm after irradiation with 100 mGy of ^{137}Cs γ-rays (solid line) and 100 mGy of ^{56}Fe^{26+} (202 keV/μm) (dashed line), respectively. The intensity of peak 3 decreases very fast with increasing LET, while the intensity of the composite high-temperature peak shows only a very slow decrease at this high LET values.

A data comparison for the peak 3 and the peak 5 TL-efficiency of TLD-300 is given in Tables 6 and 7. It is clearly seen that, while the peak 3 TL-efficiency decreases rapidly with increasing LET (similar to LiF:Mg, Cu, P), peak 5 TL-efficiency is > 1 up to LET values of $\sim$ 50 keV/μm. Recent data from Hajek et al. (2008) and Massillon-JL et al. (2008) further emphasize that LET is not the only parameter governing TL-efficiency—as was observed previously for LiF:Mg, Ti and LiF:Mg, Cu, P.

Table 7
Relative TL-efficiency for peak 5 in CaF$_2$: Tm; data are obtained from analysis of the composite peak height

Ion	LET$_\infty$ H$_2$O (keV/μm)	$\eta_{\mathrm{HCP},\gamma}$ peak 5 (ATI)	$\eta_{\mathrm{HCP},\gamma}$ peak 5 (DLR)
^{4}He^{2+}	2.3 ± 0.0	1.055 ± 0.133	1.021 ± 0.018
	4.5 ± 0.1	1.114 ± 0.121	1.099 ± 0.019
^{16}O^{8+}	19.9 ± 0.0		1.199 ± 0.031
^{20}Ne^{10+}	31.5 ± 0.1	1.341 ± 0.117	1.153 ± 0.060
	54.2 ± 1.1	1.136 ± 0.070	1.014 ± 0.038
^{40}Ar^{18+}	89.5 ± 0.2		0.784 ± 0.024
^{56}Fe^{26+}	202.0 ± 0.9	0.836 ± 0.109	0.752 ± 0.048
	312.1 ± 7.2		0.597 ± 0.016

Contrary to LiF:Mg, Ti which is commercially available in the form of sintered chips, for the TLD-300 single crystals dopant variations may lead to an observable batch dependence. Similarly to LiF:Mg, Ti (Berger et al., 2006b), also CaF$_2$: Tm gives a two-parameter signal which can be used to gather more information about the radiation field under study than just absorbed dose. The same accounts for a combination of CaF$_2$: Tm, LiF:Mg, Ti and/or LiF:Mg, Cu, P detectors (Berger et al., 2006c; Olko et al., 2002).

6. Conclusion

A comprehensive review of TL-efficiency was presented which relied on the manifold experiments that had been conducted by several laboratories during the last decades. The paper concentrated on some of the most frequently used TL phosphors, i.e. LiF:Mg, Ti (Harshaw TLD-100/600/700, TLD Poland MTS-7), LiF:Mg, Cu, P (Harshaw TLD-600H/700H, TLD Poland MCP-7) and CaF$_2$: Tm (Harshaw TLD-300). It was demonstrated that TL response to different heavy charged particles with the same LET is not a unique function of ionization density, but rather depends on the microscopic pattern of energy deposition. For the same value of LET, the dose from a particle with greater charge is locally deposited in larger volumes around the particle track, corresponding to lower local ionization density in the core of the particle track and therefore enhanced TL-efficiency. For glow peaks with a linear-supralinear-sublinear γ-ray dose response, relative

TL-efficiency with respect to γ-rays may exceed unity if the locally absorbed dose falls in the supralinear region.

When relative TL-efficiency with respect to γ-rays is concerned, numerical values evaluated by different researchers proved to be in good agreement within the statistical uncertainties—independent of the individual experimental protocols that had been used. Theoretical models applied today include track structure theory (TST), modified track structure theory (MTST) and microdosimetric target theory (MTT). These calculational approaches are able to predict the qualitative behaviour of relative TL-efficiency as a function of LET and particle type. Quantitative agreement with experiments is currently limited to certain particle energies and necessitates improvements in the developed models.

In view of potential TLD applications in different areas of heavy charged particle dosimetry, the picture of TL response characteristics can be regarded as very complete for the most frequently applied TL phosphors. For radiation fields dominated by low-LET particles e.g., the space radiation environment, legitimate demand for further experimental efforts regarding the LET interval between ~ 0.3 and $2\,\mathrm{keV}/\mu\mathrm{m}$ is anticipated.

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