Thermoluminescence Dosimetry - Application in Environmental Monitoring

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Abstract: Very soon after the introduction of the application of thermoluminescence (TL) for dosimetric purposes the method was used also for environmental monitoring. Thermoluminescence dosimetry (TLD) systems are permanently developed and improved. The aim of the present paper is to help the reader to introduce TLD for environmental monitoring. Therefore the paper describes the brief theoretical background of thermoluminescence. The elements of TLD systems, such as detectors, readers and measurement cycle including annealing, package and storage, irradiation, readout and mathematical evaluation are summarised. The main characteristics of various TL systems (batch homogeneity, sensitivity, reproducibility, linearity, light sensitivity, fading and energy dependence) are analysed mostly from the viewpoint of use in environmental monitoring. Special attention is given to the performance test requirements of the International Electrotechnical Committee and the importance of intercomparisons. Some environmental dosimetry applications during the last ten years are also reviewed.

Key words: thermoluminescence dosimetry, environmental monitoring, TL materials, thermal treatment, linearity, fading, energy dependence, intercomparison

1. Introduction

Since the suggestion of Daniels et al.¹⁾ to apply thermoluminescence (TL) for radiation dosimetry purposes a lot of effort has been made in the scientific community to explain the mechanism of thermoluminescence, as well as to improve its application for various dosimetry purposes such as personal, environmental and clinical dosimetry. Today most nuclear or radiological facilities apply TL dosimetry.

The dose in the environment depends on many factors. In addition to natural radiation, the contribution from man-made ionising radiation sources should not be neglected. The natural radiation arises from cosmic and terrestrial radiations and the dose rate is in the range of 100-200 nSv/h. The variability of natural radiation can often exceed that from man-made sources. Cosmic radiation consists mostly of high energy ionising radiations and high LET radiation (neutrons).

The advantages of passive detectors for environmental monitoring are that they are small, cheap, do not require in situ electronic power supply and can be used in a large dose

range. They provide measurements of the dose integrated during the time interval (days – months) which means that only an average dose rate value for this period can be estimated.

Readers interested in introducing TL dosimetry can find useful guidance and information in the books by Cameron et al.²⁾, Becker³⁾, McKinlay⁴⁾, Oberhofer and Scharmann⁵⁾, Horowitz⁶⁾, McKeever⁷⁾, Vij⁸⁾, McKeever et al⁹⁾, Chen and McKeever¹⁰⁾ and Furetta and Weng¹¹⁾ as well as in the book chapter by Uchrin and Ranogajec-Komor¹²⁾.

The aim of this paper is to help the reader to introduce the application of TLD for environmental monitoring. For this purpose a short overview of the physical principles is given followed by the description of the most important aspects of practical dosimetry. The last chapter deals with environmental monitoring applications of TLD.

2. Basic physical principles

Thermoluminescence (TL) or thermally stimulated lu-

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Fig. 1. Typical glow curve of $\text{Al}_2\text{O}_3\text{:C}$ detector irradiated with $D = 2\text{mGy}$ (^{137}Cs source). Reader: TOLEDO 634. Heating rate: 10 K/s

minescence is the emission of light during heating of a solid sample (insulator or semiconductor), previously excited by radiation. The TL material absorbs energy during exposure to radiation (ionising, visible light, UV etc.) and stores this energy until heated. The intensity of emitted light as a function of temperature is the thermoluminescence glow curve. Usually the heating rate is constant, and therefore the glow curve is often shown in the literature as the TL intensity as a function of the time of the measurement cycle or the channel number instead of the temperature. In this case it is advisable to show also the time-temperature profile in addition to the glow curve. A typical glow curve of an $\text{Al}_2\text{O}_3\text{:C}$ detector¹³⁾ - which since its development has been of interest for environmental applications - can be seen in Fig. 1. For dosimetry applications the integrated area under the glow curve between two temperature (channel/time) values or the peak height has to be linearly proportional to the radiation dose.

Although the first theoretical elaboration of thermoluminescence was submitted in the middle of the last century¹⁴⁾, there is no general theoretical model up to now to explain the dosimetric characteristics of a certain dosimetry system.

The theoretical explanation of thermoluminescence is based on the electron band theory. A very useful overview of theoretical models can be found in references^{10), 15)} and¹⁶⁾. In this paper only the basic principles of some models will be briefly introduced to indicate the complexity of the TL

Fig. 2. Schematic illustration of processes produced in the crystal by irradiation and a successive heat treatment

phenomena.

As already mentioned, TL materials are insulators or semiconductors with a crystalline structure. According to the simplest model, the one trap-one centre model, in an ideal crystal the electrons occupy the valence bands, which are separated from the conduction band by the so-called forbidden band gap (Fig. 2). In real crystals various defects occur as a consequence of ionising radiation or of impurities in the lattice. In both cases the electrons can possess "forbidden" energies. The excitation of electrons from the valence band to the conduction band may occur as a result of irradiation (step a, Fig. 2), while holes remain in the valence band. Some of the electrons and holes can recombine under emission of light (radioluminescence), while the others will be trapped at the level L_T in the "forbidden" energy gap (step b, Fig. 2). The probability of the release of an electron from the forbidden band into the conduction band (step c, Fig. 2) can be described by the Arrhenius equation:

$$p = se^{-E/kT} \quad (1)$$

where p is the probability per unit time, s is the so called frequency factor (that, according to the simple model, is constant), E is the activation energy, k is the Boltzmann's constant, T is the absolute temperature. With increasing T the probability of recombination increases too. E is related to the energy difference between the trap and the conduction level.

The electron from the conduction level undergoes recombination with the hole (step d and e in Fig. 2) in the recombination center R which at the same time functions also as a luminescent center (LC) causing a recombination accompanied by a light emission (TL). The intensity of the TL (photons/s) is proportional to the recombination rate, i.e. to the

concentration of free electrons in the conduction band and holes in the valence band. The equation of the recombination rate can be solved analytically only with additional simplifications according to the first order¹⁴⁾, second order and mixed order kinetics^{10,11,15)}.

All these theories are based on the simple one trap-one center model. This model can describe the shape of the TL glow curve i.e. the intensity of the TL as a function of temperature, $I(T)$ as well as the influence of charge concentration, heating rate and trap depth on the glow curve.

The realistic situation is even more complicated and $I(T)$ can be found only by solving a set of simultaneous differential equations without any simplifying assumptions.

3. TLD system

The possible application of the TLD depends on the characteristic of the complete TL system which consists of the detector, the TL reader and the measurement cycle including the mathematical evaluation of the results.

TL detectors

TL detectors are natural or synthetic materials, which emit light the intensity of which is proportional to the dose of irradiation when heated after having been exposed to radiation. Nowadays a lot of TL phosphors are well-known which fulfil the requirements for various practical dosimetry applications.

For many years the most commonly used TLDs were LiF detectors doped with Mg and Ti⁹⁾. Very often the commercial names (codes) of the detectors are cited in the literature. So, TLD-100 stands for the detector LiF:Mg,Ti produced by Harshaw/Bicron (recently Saint Gobain Crystals and Detectors) from natural Li, while TLD-600 and TLD-700 are produced from enriched ⁶Li and ⁷Li isotopes, respectively. Sintered pellets of the same chemical composition produced in Poland from natural LiF are referred under the trade name MTS-N^{17,18,19)}.

The following detectors appear also quite often under their commercial name (the commercial name used by the Harshaw company is given in bracket): LiF:Mg,Cu,P (TLD-100H²⁰⁾), 7LiF:Mg,Cu,P (TLD-700H²⁰⁾), CaF₂:Dy (TLD-200), CaF₂:Tm (TLD-300); CaF₂:Mn (TLD-400), Al₂O₃:C (TLD-500), and CaSO₄:Dy (TLD-900). The highly sensitive LiF:Mg,Cu,P was first proposed by Nakajima, Murayama, Matsuzawa and Koyano²¹⁾. Nowadays this type of detector is produced by different laboratories sometimes under different commercial names. The LiF:Mg,Cu,P detector produced in the Solid Dosimetric Detector and Method Laboratory^{22,23)}, Beijing,

China, appears also as GR-200 or GR-200A in the literature (the difference is in the preparation procedure). LiF:Mg,Cu,P detectors produced in the Institute of Nuclear Physics, Krakow, Poland^{17,18)} are cited also as MCP-N. The code used is not always unified in the literature, therefore the exact description of the chemical composition, the activators and the origin of the detector is highly recommended.

According to the author's opinion, the basic routine dosimetry tasks in personal dosimetry and environmental monitoring can readily be solved with an appropriate selection of commercially available detectors. However, there has been no ideal TL dosimeter for all tasks so far. Therefore, permanent efforts are made in the scientific community to develop a new "ideal" TL detector. The aim of this paper is not to review all kinds of detectors. A look in the Proceedings of the last Solid State Dosimetry Conference²⁴⁾ will give an impression to the reader on the intensive work in this field.

In addition to the detectors mentioned above, the following detectors can also be used for environmental monitoring: CaF₂:Mn^{25,26)} (produced by the Jozef Stefan Institute, Ljubljana, Slovenia), sintered pellets LiB₄O₇ detectors^{27,28)} with different dopants (produced by the "Vinca" Nuclear Research Institute, Belgrade, Serbia and Montenegro) and a new high sensitive LiF:Mg,Cu,Na,Si TL phosphor with a low residual signal, good thermal stability and high sensitivity (produced by the Korea Atomic Energy Research Institute, Taejon, South Korea²⁹⁾). El-Faramawy et al³⁰⁾ also developed a promising copper doped LiB₄O₇ with low fading and a wide linear dose range. Gerome and co-workers reported on the effect of the preparation method of CaSO₄:Dy on its dosimetric properties³¹⁾.

The selected system needs an adequate performance test by the user to estimate the uncertainty even in case of a commercial detector provided manufacturer's performance recommendations.

TL readers

In principle, the reading process is based on the heating of the TL material from ambient temperature up to 300 - 400 °C, while the emitted light is collected and measured quantitatively. The main features common to all TLD readers are the heating unit, the light collection and detection system, the signal-measuring system and the data recording system¹²⁾. Most laboratories use commercially available manual or automatic readers. For routine personal and environmental monitoring, the automatic reader has the advantage because it can evaluate a large number of TL detectors with minimal man-power. On the other hand, for research purposes the

a

b

Fig. 3a. Laboratory irradiation Fig. 3b. Field irradiation of environmental dosimeters during the 12th EML International Intercomparison

manual reader is a better choice, because it offers a variability of experimental parameters (heating rate, variable integration limits of the glow curve etc.).

Several laboratories developed their own proprietary readers^{25,32,33,34,35} to fulfil special tasks or requirements. A special “environmental” dosimetry is the space dosimetry. Apáthy and co-workers³⁵ developed a unique portable reader for this purpose.

TL measurement cycle

All steps of the measurement cycle, such as annealing, package and storage, irradiation, readout and mathematical evaluation, influence the characteristics and uncertainties of the dosimeter system.

Annealing is an essential pre-irradiation heat treatment procedure to achieve reproducible results. One of the advantages of TL detectors is the possibility of their re-use. High temperature annealing is needed in order to maintain the characteristics of a TL detector (shape of the glow curve, TL sensitivity, background signal) after repeated irradiations. The annealing procedure is the following: the sample will be heated to and kept at a given temperature for a certain time, after which it will be cooled with a given cooling rate to ambient temperature. The influences of variations in the heating and cooling rate, as well as the annealing temperature on the glow curve shape are usually significant. This means that for standardisation of the sensitivity i.e. the TL response, all detectors from the batch have to be annealed identically. As a consequence the annealing empties all the traps, i.e. the TL signal will be reset to zero. At the same time the thermody-

namic defect equilibrium is also reestablished¹⁵ during the annealing. Intensive research of the heat treatment effect on the properties of LiF:Mg,Ti dosimeters from the early beginning³⁶ till today³⁷ shows the importance of annealing. It is not possible to describe all the annealing studies, however the attention of the reader should be directed to some papers containing practical and useful informations on annealing of detectors applied in environmental monitoring, such as LiF:Mg,Ti³⁸, CaF₂:Tm³⁹, CaSO₄:Dy⁴⁰, Al₂O₃:C⁴¹, LiF:Mg,Cu,P^{42,43} and LiF:Mg,Cu,Na,Si⁴⁴.

The mode of package and storage influences the results of dosimetric measurements. The TL detector becomes a TL dosimeter after placing it in an adequate holder which enables electronic equilibrium during irradiation. The holder or badge contains various filters usually to absorb the low energy radiation (see below: energy dependence). In environmental monitoring, where long-term field irradiation under sometimes extreme climatic conditions is carried out, the package serves also to protect the dosimeter against UV/sunlight, humidity and mechanical damage. In international intercomparisons the holder ensures also protection against such damages during the long-term international postal transport. Figs. 3a-b show the large variety of dosimeters included in the 12th international intercomparison⁴⁵ organised by US DOE Environmental Measurements Laboratory (EML) with 133 participants from 43 countries.

After having accomplished the procedures described above, the TL dosimeter is irradiated for one of two reasons: calibration (Fig. 3a) and field irradiation (Fig. 3b). As TL dosimetry is not an absolute method, calibration with well-

known and well defined gamma radiation sources is needed. The most commonly used sources are: ^{60}Co , ^{137}Cs , ^{226}Ra . Commercially available irradiators usually contain a $^{90}\text{Sr}/^{90}\text{Y}$ source. Such irradiators can satisfy routine tasks, however, for exact measurements they have to be calibrated by a calibration source.

The field irradiation can be carried out in any environment, i.e. indoor or outdoor measurements are both “environmental” monitoring in a wider sense. In outdoor measurements the dosimeters have to be placed at a distance of 1-2 m from the ground and 5 m from a building to avoid terrestrial radiation and exposure from the building material. Irradiation time varies from several days⁴⁶⁾ to several months^{47,48)}. Budzanowski et al.⁴⁶⁾ proposed, developed and tested a system for rapid assessment of accidental exposures (RACE) using LiF:Mg,Cu,P (MCP-N) detectors. The system was designed for a large number (up to 500) of short-term (1-5 days) exposures or for long-term measurements of kerma rate in the environment. On the other hand, if long-term measurements are necessary (for example for the determination of the natural background kerma rates) it is better to choose a longer monitoring period to keep the ratio of the environmental monitoring and the transport dose high, i.e. to have a low contribution of the transport dose^{47,48)}.

During the *readout* (process of heating the detector to obtain the glow curve) the following parameters have to be optimised in a performance test: temperature and time of preheat, heating rate, temperature and time of readout. Preheat (sometimes called post-irradiation annealing) is used to eliminate the low temperature peaks of the glow curve⁴⁹⁾ i.e. to reduce fading (see part 4.). The choice of an adequate temperature-time profile during pre- and post-irradiation annealing as well as during the readout is important because these parameters influence the glow curve structure and the sensitivity of the TL detector. It was observed that the kinetic parameters E and s (Eq. 1) are strongly dependent on the cooling rate of the pre-irradiation, the high temperature annealing and the heating rate during the readout⁵⁰⁾. Yossian and Horowitz showed theoretically and experimentally that the glow peaks of the TLD-100 undergo an important modification following post-irradiation annealing at 165 °C and that this modification depends on the preheat time⁵¹⁾. Kitis and co-workers⁵²⁾ showed the effects of the heating rate from 0.6 up to 50 °C s⁻¹ on the thermoluminescence of $\text{Al}_2\text{O}_3\text{:C}$. This material rapidly loses its high sensitivity as the heating rate increases. A similar effect was found by Pradhan⁵³⁾ who described also a significant shift in the glow peaks of LiF:Mg,Cu,P sintered pellets with increasing heating rate to

the temperature range where irreparable damage takes place. Due to this effect the use of $\text{Al}_2\text{O}_3\text{:C}$ and LiF:Mg,Cu,P in large scale environmental monitoring systems having fast heating rates is limited. In the practice mostly a heating rate of 10 °C s⁻¹ is adequate while for glow curve structure studies slower heating rates are advisable. At the Rudjer Boskovic Institute, Zagreb preheat at 100 °C for 20 minutes was found to be very useful for fading correction^{26,54,55)} for all kinds of LiF , $\text{CaF}_2\text{:Mn}$ and $\text{Al}_2\text{O}_3\text{:C}$ detectors.

A *mathematical evaluation* enables the determination of the evaluated dose value D_e from the TL readout value (TL response, TL) using the evaluation factor⁵⁶⁾ Fe :

$$D_e = F_e * TL \quad (2)$$

The TL response can be represented either by the maximum intensity (height of the peak) or by the area under the TL curve. The peak area method is more frequently used.

In environmental dosimetry the evaluated dose value is expressed in terms of ambient dose equivalent⁵⁷⁾, $H^*(10)$, for strongly penetrating radiation. $H^*(10)$ is defined as the dose equivalent that would be produced by the corresponding aligned and expanded field in the ICRU sphere at a depth of 10 mm, on the radius opposing the direction in the aligned field. The ICRU sphere is a 30 cm diameter tissue-equivalent sphere. Factor Fe involves the calibration factor¹¹⁾, C_f :

$$C_f = \frac{D_{ref}}{TL_{cal} - TL_{cont}} \quad (3)$$

where D_{ref} is the calibration/reference dose, TL_{cal} is the mean TL response of the calibration dosimeters, TL_{cont} is the mean TL signal of unirradiated control detectors. Fe contains also the algorithm for the calculation in terms of the dose of interest and the combination of results obtained for more than one detector. The algorithm for the calculation in terms of the dose of interest is based on the dose equivalent conversion factors^{56,58)}. Finally, Fe enables corrections to be made that become necessary due to background, individual sensitivity of the detector, fading, non-linearity and energy dependence.

4. The main characteristics of the TLD system

Before application, the characteristics of the complete TLD system have to be tested under standard test conditions. A user friendly and useful standard test programme was proposed by Piesch and Burgkhardt⁵⁹⁾. The International Electrotechnical Committee⁵⁶⁾ is improving the standard “The performance requirements for the TLD system on TL dosimetry systems for personal and environmental monitoring”. The

most important characteristics of the TLD system for environmental monitoring are the following:

Batch homogeneity means the deviation of detector sensitivity in the batch. It can be expressed as the standard deviation of the mean value of all dosimeters in the batch⁵⁹⁾ or as the difference of the TL response of any two detectors in the batch⁵⁷⁾.

Sensitivity is defined as the ratio of absorbed dose and TL response (expressed as the area under the glow curve over a given temperature region or the peak height). Sensitivity depends on many factors, such as the chemical composition of the detector material, the concentration and types of the activators, the physical form of the detector, the type of the reader²⁶⁾ etc.

In environmental monitoring beside the LiF:Mg,Ti detectors very often CaF₂ and CaSO₄ detectors with different activators were used¹²⁾. The system has to be able to measure very low doses, because the man made contribution to the environmental radiation can be only 1/10 of the background radiation. This means that high sensitivity is essential for environmental monitoring. Therefore, new, high sensitive TLD systems based on Al₂O₃:C, LiF:Mg,Cu,P and LiF:Mg,Cu,Na,Si are being developed and characterised permanently for environmental monitoring applications^{13,15,16,22,23,29,43,44,52,53,60)}. The sensitivities relative to TLD-100 expressed as the number of impulses (corresponding to the glow curve area) per unit dose (impulses/Gy) have been found to be 6, 22, 57, 68, 108 for CaF₂:Mn, TLD-700H, Al₂O₃:C⁶¹⁾, LiF:Mg,Cu,Na,Si⁶¹⁾ and GR-200A (LiF:Mg,Cu,P), respectively^{26,55)}.

According to the IEC Standard⁵⁶⁾ the reproducibility expressed as the coefficient of the variation of the evaluated dose value shall not exceed 7.5% for each dosimeter separately and *n* dosimeters collectively, for a given dose. This requirement was highly fulfilled for TLD systems applying TLD-100, LiF:Mg,Cu,P, Al₂O₃:C and CaF₂:Mn detectors⁶²⁾ in a standard test program carried out according to⁵⁹⁾.

The linearity can be described with the dose response function, *f*(*D*) (also called the supralinearity index)) of a certain TL system usually defined^{9,10,15)} as:

$$f(D) = \frac{S(D)/D}{S(D_1)/D_1} \quad (4)$$

where the value of *S*(*D*) and *S*(*D*₁) can either be the maximum intensity or the area under the TL curve. *S*(*D*) and *S*(*D*₁) are these "TL signals" at the doses *D* and *D*₁. *D*₁ is a dose at which the dose response is linear. In very favourable situations the dose response function *f*(*D*) will be linear in a broad dose range. At high doses, in practically all cases,

Fig. 4. Dose response curves for some TL detectors

supralinearity can start (which, means faster than linear). At very high doses *f*(*D*) may even decrease in some cases.

Intensive research is being carried out on modelling the supralinearity for no final solution has yet been found. Olko at al.⁶³⁾ studied the supralinearity of LiF:Ti,Mg detectors and discussed the data with reference to some models of TL supralinearity. Horowitz⁶⁴⁾ developed a comprehensive theory of the thermoluminescence (TL) dose response, i.e. the unified interaction model (UNIM). The UNIM is based on both radiation absorption stage and recombination stage mechanisms and can describe the dose response for isotropically ionising gamma rays and electrons (in the framework of the trapping centers/luminescent centers geminate recombination model) in a closed mathematical formalism.

The linearity of MTS-N type LiF:Mg,Ti, MgB₄O₇:Dy and CaF₂:Mn detectors was measured in "low"⁶⁵⁾ and "high"²⁶⁾ dose range. In Fig. 4 the dose response curves of these detectors are shown in the dose range 50 mGy-5 Gy. The R² values of linear fitting for all detectors in Fig. 4 were close to 1 (0.99-1). For the same detectors in the dose range of 0.1-5 mGy the linearity expressed as the standard deviation of the calibration factors was found to be 15.3%, 9% and 7.4% for MTS-N type LiF:Mg,Ti, MgB₄O₇:Dy and CaF₂:Mn, respectively⁶⁵⁾.

In the practical TL dosimetry detectors and systems can be used which fulfil the requirements of the IEC Standard⁵⁶⁾, i.e. the response shall not vary from the conventional true value by more than 10% over the range of 30 mSv to 100 mSv if the monitoring time is 30 days.

The stability of dosimeters under various climatic conditions as a function of time i.e. the *fading* is very important in the environmental monitoring since in the environment mostly long-term exposures are the object of the investigations. Fading characteristics of newly developed TL materi-

Table 1. Fading of various TLDs. The TL signal was normalised to the signals of the TLDs stored for 24 hours after irradiation.

Dosimeter Material Type	LiF:Mg,Ti MTS-N	LiF:Mg,Ti TLD-100	⁶ LiF:Mg,Ti TLD-700	LiF:Mg,Cu,P TLD-700H	LiF:Mg,Cu,P GR-200A	MgB ₄ O ₇ :Dy	CaF ₂ :Mn
Fading (%) after							
14 days	Dose 2 mGy ⁵⁵⁾	0*	1*	1*	0*		
21 days	200 mGy ²⁶⁾	5				10*	8
30 days	0.2 mGy ⁶⁵⁾	12				18	5

*Post-irradiation annealing treatment (100°C for 20 min) was applied

als are always investigated under different conditions (temperature, humidity, light effects). The fading depends first of all on the chemical composition of the detector and the dopants, the crystal structure, the thermal treatment during evaluation (pre-irradiation annealing, post-irradiation annealing/preheat, heating rate), the climatic and light conditions during exposure. The fading correction can be carried out in different ways. One possibility is a 24 hours storage before reading of calibration and field detectors, because the shorter is the time between irradiation and readout, the faster the fading becomes. In most cases the fading curves decrease exponentially in the first 24 (sometimes 48) hours and become linear later on^{26,65,66)}. An other method is the thermal cleaning procedure, i.e. preheat to eliminate the low temperature peaks^{26,49,55)}. Table 1 shows the fading of various dosimeters previously stored 24 hours after irradiation^{26,55,65)}. Results signed with * were obtained in combination with an external preheat (100 °C for 20 min).

A simple measurement method for avoiding the need of a fading correction of the absorbed dose from glow curves of TLD-100 was proposed by Weinstein and co-workers⁶⁷⁾. The method is based on locating the channel with the peak maximum and integrating the counts in a defined region around this channel (not all the peak area). A very useful method for fading correction and prediction is the Computerised Glow Curve Deconvolution (CGCD) method^{67,68)}. The analysis of the TL response as a function of the elapsed time of the whole glow curve as well as the individual peaks using the (CGCD) method allows the prediction and correction of the fading without preheat⁶⁹⁾. Sachar and Horowitz⁶⁸⁾ showed an exquisitely complex time-dependent response of the peak 4 and 5 in TLD-100 with CGCD. Peak 4 grows at the beginning of storage and begins to decay only after several months, while peak 5 decays rapidly from the beginning. Peak 4 and 5 have a yearly fading of about 8% when not measured separately (which is mostly the case in practical dosimetry). This indicates the very complex background of the fading requiring

a thorough and prudent analysis previous to the choice of an adequate fading correction. The differences in the stability of the various peaks could be useful in estimating the time between exposure and readout. The method was used by Budzanowski at al. for MCP-N and GR-200A LiF:Cu,Mg,P detectors in the period up to 3 months⁷⁰⁾.

According to the IEC Standard the fading has to be within 5% and 10% for 30 and 90 days, respectively under standard test conditions⁵⁶⁾, while it can be 20% for 30 days storage at 50 °C and 65% relative humidity or at 20 °C and 90% relative humidity.

TLD-700 and LiF:Mg,Cu,P detectors of various origin were prepared and evaluated by different laboratories and tested in an environmental chamber under extreme temperature and humidity conditions^{71,72)} as specified in ANSI Draft N13.29⁷³⁾. The ANSI N13.29 standard requires that the absolute value of the bias and standard deviation of the TL responses must be less than or equal to 0.35, while their sum must be less than or equal to 0.50. The mathematical algorithm is described in ⁷¹⁾ and ⁷²⁾ in detail. All detectors included in this study passed the performance test according to ⁷³⁾. However, all TL systems showed larger fading than expected in spite of fading corrections (annealing and preheat before reading, readout after at least 24 hours, correction fade-factors etc.). This fading phenomena was not observed under normal environmental conditions⁷⁴⁾.

The *light sensitivity* of TL materials is a very complex phenomena. Light sensitivity stands for the photo-induced thermoluminescence (PITL) effect, i.e. the TL directly induced by visible or UV light in a fully annealed material. High PITL sensitivity and linear response allows the use of phosphors as UV dosimeters⁷⁵⁾. It was shown for various aluminium oxide detectors^{76,77)} that the PITL sensitivity depends on the impurity content (concentration and chemical composition of the activators), the preparation conditions and is also influenced by the grain size. The PITL sensitivity of the Al₂O₃:C phosphor was selective to wavelength of UV light, i.e. the

Fig. 5. The PITL, TL and PTTL responses of MTS-N and TLD-100 detectors. The 254 nm UV light exposure rate was $1.6 \text{ mJ cm}^{-2} \text{ s}^{-1}$, the illumination time was 10 min.

254 nm light induced response was more than 20 times higher in comparison to the 366 nm one.

TLD materials exhibit a variation in readout with light exposure (including both visible and UV radiation energy) which may cause problems in environmental dosimetry. In TLD-100 the overall fading of the TL signal (peak 4 and 5) was reduced but the dissipation of the results increased when dosimeters were exposed to UV light following an irradiation with X-rays⁷⁸). The light sensitivity study of LiF:Mg,Ti (GR-100) and LiF:Mg,Cu,P (MCP-N) proved, that MCP-N is approximately five times less light sensitive than GR-100⁷⁹). However, for MCP-N, this sensitivity relates to a loss of approximately half the signal in 14 days of sun exposure. In aluminium oxide TL detectors light exposure increased the fading⁸⁰), while the readout of $\text{Al}_2\text{O}_3\text{:C}$ increases with increasing light exposure⁷⁹). This light sensitivity can be exploited in the optically stimulated luminescence dosimetry (OSLD) when the detector is stimulated with light after exposure to ionising radiation and luminescence is recorded. In the case of $\text{Al}_2\text{O}_3\text{:C}$ detectors a several times higher response using OSLD was achieved than the TL output⁸¹).

Another advantage of light sensitivity of TL materials is the possibility of dose re-estimation by phototransferred thermoluminescence (PTTL)⁷⁶). PTTL is based on the retention of TL information – on exposure to UV light – in TL materials previously exposed to ionising radiation. The procedure for PTTL determination compared to PITL and TL is the following:

PITL: annealing - UV exposure – readout

TL: annealing – irradiation – readout

PTTL: annealing – irradiation – readout - UV exposure – readout

The detection threshold for PTTL re-estimation is mainly defined by the UV sensitivity. Fig. 5 shows the PITL, TL and PTTL responses of two LiF based TL detectors: MTS-N and TLD-100. The PITL and PTTL curves indicate that the UV sensitivity of MTS-N is more than 10 times, while the TL sensitivity is about 1.5 times higher than that of TLD-100. The lowest doses that could be re-estimated were obtained from the PTTL responses as functions of previous gamma irradiation doses (Fig. 6). There were found to be 1.5, 2.0, and 4.0 mGy for $\text{CaF}_2\text{:Mn}$, MTS-N and TLD-100, respectively. Despite of the non-linearity of the PTTL dose response curve in Fig. 6, this method can be used for the re-estimation of dose with these detectors, provided an adequate calibration has been made.

According to the IEC Standard⁵⁶) referring to routine environmental applications, the evaluated value of the TLDs shall not differ by more than 10% after one week exposure to daylight (1000 Wm^{-2}) from the evaluated value of a dosimeter kept in the dark.

In environmental monitoring, in personal dosimetry and especially in clinical application of TLD the energy range of interest is very wide. Determination of the energy dependence is required by most standard and performance tests. For environmental dosimeter systems, the IEC Standard⁵⁶) requires that in the energy range 30 keV to 80 keV the evaluated value shall not exceed the conventional true value by

Fig. 6. PTTL responses of various TL detectors expressed in equivalent gamma dose

more than a factor of 2 and in the range of 80 keV to 3 MeV the difference cannot be more than 30 %.

The energy response depends first of all on the effective atomic number (Z_{eff}) of the detector. It is known from the theory, and it was also shown experimentally, that dosimeters based on Li have the lowest, those based on Al a higher, and those based on Ca the highest energy dependence^{12,26,55,82,83}. On the other hand, it was shown that the energy absorption characteristics of TLDs depend not only on the effective atomic number of the respective TLD, but are influenced by the added dopant as well^{55,83}. Fairbanks and Dewerd found that LiF:Mg,Ti TLD's obtained from three manufacturers showed similar characteristics in energy response and linearity, the sensitivity, however, varied among the three groups by as much as 40%⁸⁴. For TLD-100 the supralinearity showed an energy dependence, the supralinearity at 30 keV being lower than that at 1250 keV⁸⁵. The photon energy response of various LiF:Mg,Ti (TLD-100, TLD-700, MTS-N) was compared to that of various LiF:Mg,Cu,P (TLD-700H, MCP-N, GR-200) by several authors^{55,82,85,86}. The doses measured by the high sensitivity LiF:Mg,Cu,P were lower than the nominal doses for the energies of 80-160 keV⁸². At 40.5 keV LiF:Mg,Cu,P shows over-response of 24%, while the LiF:Mg,Ti detectors (MTS-N and TLD-700) have an over-response of up to 50% (Fig. 7).

The energy response at about 100 keV of the new LiF:Mg,Cu,Na,Si detectors shows a decrease, similar to that in LiF:Mg,Cu,P. For lower energies the response is higher than that for LiF:Mg,Cu,P, due to the presence of high-Z elements (Na, Cu, and Si)²⁹.

The knowledge of the effective atomic number in rela-

Fig. 7. Measured photon energy dependence of LiF based detectors (dose of irradiation = 2 mGy)

tion to tissue is not sufficient for the prediction of the suitability of a dosimeter to measure the ambient dose equivalent⁵⁶ $H^*(10)$, especially for low energy radiation. This is mainly due to the fact that the dosimeter is placed on the phantom, whereas $H^*(10)$ is defined and calculated for the depth of 10 mm, and for these two cases the spectra of low energy photons are different.

5. Application of TLDs in environmental monitoring

Well characterised and calibrated TLD systems are used worldwide for environmental monitoring. The actuality of this task can be seen, among others, from the Proceedings of a Workshop on Environmental Dosimetry⁸⁷, Proceedings of the last Solid State Dosimetry Conference²⁴, as well as from the Editorial of the Special Issue of Journal of Environmental Radioactivity Journal on Environmental Dosimetry and Radiation Effects in the Environment⁸⁸.

The objective of environmental monitoring is to provide data on natural background radiation and to determine the contribution to the dose to public from man-made sources^{89,90}. For this purpose measurements are carried out starting at the nuclear or radiological facility limits to country-wide locations. A permanent country-wide background monitoring with passive dosimeters serves as a basis for the calculation of the contribution to dose in accidental situations.

The most commonly used TL detector in environmental monitoring is even today the LiF:Mg,Ti. A progress has been made with the introduction of high sensitivity detectors,

LiF:Mg,Cu,P⁹¹⁾, Al₂O₃:C and recently attempts are done in introducing LiF:Mg,Cu,Na,Si²⁹⁾. Their high sensitivity allows for a rapid and short time monitoring^{46,92)}.

An important alternative to thermoluminescence is optically stimulated luminescence with Al₂O₃:C⁸¹⁾ and phosphate glass⁹³⁾ dosimeters. These detectors are widely used in various environmental conditions. Without pretension to entirety an overview of environmental applications of TLDs during the last years will be given.

LiF:Mg,Cu,P was compared to CaSO₄:Dy including laboratory irradiation and environmental exposure before the start-up of a nuclear power plant operation in China⁹⁴⁾. The differences of dose measurements between two thermoluminescent dosimeters were not significant. LiF: Mg,Cu,P was applied for the radiation survey of the Antarctic region⁹⁵⁾. The experimental results showed that the distribution of natural radiation levels in Antarctica is very uneven. Laboratory and a three year field comparison were carried out with LiF:Mg,Ti and LiF:Mg,Cu,P. The expected impact on the results when transferring from one system to the other is about 10%⁸⁹⁾. LiF:Mg,Cu,P based environmental dosimeter card and dose calculation algorithm was constructed⁹⁶⁾ in a way that it can determine directional and ambient dose equivalent⁵⁷⁾ and meet the criteria of ANSI N3.²⁹⁷³⁾

Indoor natural radiation doses existing in dwellings have been estimated using CaSO₄:Dy thermoluminescent dosimeters and the annual effective dose equivalent to the population due to indoor gamma radiation was calculated⁹⁷⁾.

The survey for one site in Bulgaria has been performed with passive detectors assembled from TLD (CaSO₄:Dy) for gamma radiation measurements and solid state nuclear track detectors (SSNTDs) for alpha measurements (radon)⁹⁸⁾.

A special application of thermoluminescence and optically stimulated luminescence is the dose reconstruction method after radiation accidents or when the source of radioactive contamination was not under proper control. This latter situation occurred in the Southern Ural region as a result of radionuclide releases from a plutonium facility in the region. An intercomparison was carried out to measure doses accumulated in bricks due to man-made sources of radiation⁹⁹⁾. The bricks, collected from 60-100 year old buildings in a contaminated village, had accumulated a relatively high dose due to natural sources of radiation incorporated inside the bricks themselves and from the surrounding environment. After subtraction of the natural background dose, the absorbed dose in bricks due to contaminated river sediments and banks was calculated.

A simple method to eliminate background radiation for

estimating elevated dose in the vicinity of a nuclear facility (due to downward radiation arising from the plume discharge, as well as to direct and scattered radiations) was presented in the literature¹⁰⁰⁾. Two different types of passive dosimeter, a photoluminescence dosimeter (PLD) and a CaSO₄:Tm TLD were applied. The PLD results were in agreement with the reference value, while with TLD the agreement was achieved only after energy dependence correction.

Four different techniques for the assessment of environmental gamma dose in northwestern Italy were compared: high pressure ionisation chamber, TLD dosimeters (GR-200), in situ gamma ray spectrometry, and dose evaluation from the activity concentration of radionuclides in the soil. The results obtained with these four techniques agree well within 20%¹⁰¹⁾.

Prokic reported on a multi-element dosimeter system for environmental monitoring¹⁰²⁾. It consists of three solid sintered and properly filtered CaSO₄ activated with Dy or Tm TL detectors and of one low Z, MgB₄O₇:Dy,Na detector. This TL dosimetry system met all specified criteria and provided accurate dosimetry required for environmental monitoring.

As mentioned earlier, for an accurate environmental dosimetry, well prepared performance tests are necessary. Even a well established routine or scientific laboratory needs a control from time to time. Intercomparisons provide unique opportunities to participants to check their calibration, the use of different corrections and the methods of measurements. Intercomparisons of TLD systems can help the participants also to achieve international standards. Intercomparison in environmental monitoring is strongly advised also by the Recommendations of the European Radiation Dosimetry (EURADOS) Working Group 12¹⁰³⁾. Several large-scale international intercomparisons were organised for passive environmental dosimetry systems. The US Department of Energy Environmental Measurements Laboratory organised 12 intercomparisons for passive environmental dosimeters^{45,104)}. Typically over 100 participants from 25-30 countries submitted various types of dosimeters (see Fig 3a-b). The last detailed report of the US intercomparisons was submitted after the 11th intercomparison¹⁰⁴⁾. This large-scale intercomparison of environmental dosimeters was organised in 1996 and involved 121 participants from 31 countries. Significantly, an increased application of cer-doped LiF was evident in comparison to the earlier intercomparisons. The intercomparison included field and different energy laboratory irradiations. More than 80% of the field dosimeter results were in agreement within 30% with the reference values, but only 42-59% of participants were within 10% of the

reference values in laboratory irradiations.

Ranogajec-Komor et al. organised two different intercomparisons^{54,62,73,74)} which were based on the joint-cross calibration in the laboratories involved and an exchange of the dosimeters within the respective national environmental monitoring programs. Both intercomparisons were focused to the fading in the field. In the first one LiF:Mg,Ti, CaF₂:Mn and phosphate-glass dosimeters were involved⁵⁴⁾. It was found that the fading corrections did not improve the agreement between the dosimeter systems, the differences in the fading factors having been due to the properties of the dosimetric systems. In the second intercomparison⁷⁴⁾ the results indicated that the new high sensitivity (LiF:Mg,Cu,P and Al₂O₃:C) detectors were very suitable for environmental monitoring. Even in a shorter monitoring period (less than 100 days) the agreement among dosimetric systems with the new high sensitivity detectors was good (less than 10% in natural radiation environments). The large fading obtained earlier⁷²⁾ with the same systems (excluding Al₂O₃) in extreme temperature and humidity conditions was not observed under "normal" environmental conditions at various sites.

Summary

Environmental radiation monitoring with thermoluminescence dosimeters is applied nowadays on a wide scale. The use of TLDs is attractive because of their small size, low price and the absence of any need for servicing in the field. With appropriate choice of the dosimetry system, especially the detector and the heat treatment, very wide dose range can be measured, from several μ Gy to 10⁵ Gy. In environmental radiation dosimetry low doses are more interesting, because the contribution of man-made dose under normal conditions is small compared to the natural background level. Therefore the permanent development of new high sensitivity TL detectors improves dose measurement. A dosimetry system has to be tested according to the international or national standard performance test programs in order to achieve good accuracy. To achieve international standards, intercomparisons of TLD systems are highly recommended.

Permanent efforts have to be made in theoretical modelling and problems to improve our understanding of the influence of various parameters on the characteristics of thermoluminescence dosimetry system.

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