REVIEW OF THE THERMOLUMINESCENT DOSIMETRY METHOD FOR THE ENVIRONMENTAL DOSE MONITORING

by

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Review paper
https://doi.org/10.2298/NTRP2102150S

Passive solid state dosimeters, such as thermoluminescence dosimeters, provide integrated measurement of the total dose and are widely used in environmental monitoring programs. The objective of this paper is to provide a comprehensive review on the use of thermoluminescent dosimetry methods for monitoring radiation dose in the environment. The article presents the part of the research results of the project PREPAREDNESS (EMPIR 2016 call for *Metrology for Environmen*t joint research project) with a particular objective to harmonize procedures used by dosimetry services, relevant authorities and Institutes across the Europe. To achieve this, different monitoring routines that are based on passive environmental dosimetry methods are investigated. Differences in performing specific steps such as preheating, reading, annealing, minimizing fading, and others, are analyzed. The investigation was performed by means of qualitative literature review that showed the lack of information about specific steps. The conclusion of this work is that thermoluminescent dosimetry measurement system has to be type-tested even though the testing procedure is complicated. In addition to this, control dosimeters should be introduced, International Organization for Standardization protocols should be followed during calibration, and finally, parameters influencing the measurement uncertainty have to be identified and well understood in order to produce accurate dose measurement results.

Key words: thermoluminescent dosimetry, environmental dose monitoring, annealing, fading, readout, calibration, transit dose

INTRODUCTION

The *Preparedness* project started in August 2017. This is a project within the European Metrology Programme for Innovation and Research (EMPIR) 2016 call for *Metrology for Environment* joint research [1]. The results of the project are planned to address the protection of the public and the environment in the case of a nuclear or radiological event by means of reliable measurement instruments and methods. The role and the example of radiation measurements that need to be undertaken in case of radiological emergency event are summarized in [2]. Environment monitoring is one of those measurements and can be done by different methods [3]. The *Preparedness* project has four work packages (WP) and the main aim of WP4 (Passive Dosimetry) is to harmonize passive dosimetry for environmental radiation monitoring across Europe. In order to achieve this aim, WP4 has been divided into four tasks, 4.1-4.4. (Investigation of the current status of passive environmental dosimetry, Technical and methodological investigations on passive area dosimetry, Measurement of the ambient dose equivalent using electret ion chambers, and Harmonization of European dose rate measurement procedures using passive dosimeters, respectively). The task 4.4 analyzes the published data, and the newly gathered data from tasks 4.1-4.3 that are subsequently used to make recommendations about specific monitoring steps that will lead to harmonization of environmental dosimetry. This paper will provide insights from that task.

Passive solid state dosimeters provide an integrated measurement of the total dose. The total dose includes the dose from monitored man-made radiation sources, natural background radiation and undesired doses received at different steps (stages) of passive dosimetry method, such as preparation, transport or readout. Even though this implies that measured dose has to be evaluated and interpreted, the benefits of using inexpensive, re-usable and rugged dosimeters in remote locations permits one to establish adequate procedures to per-

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form environmental monitoring over wide areas. Nowadays, a large number of worldwide stations are being monitored with passive dosimeters. These are mainly thermoluminescence dosimeters (TLD). Other dosimeters based on different detector types, such as optically stimulated dosimeters (OSLD), photographic film, radiophotoluminescent dosimeters (RPLD) or electret ion chambers are also used [4-8].

The TLD are used in many scientific and applied fields such as radiation protection, medicine or environmental and space research. The use of TLD in the environmental monitoring programmes started in 1960 when highly sensitive TLD had become available. Initially, the objective of the environmental monitoring was to measure long-term accumulated dose over the period of 3-6 months at large number of places distributed across the environment. However, with the development of nuclear programmes, the need to measure a short-term dose from man-made sources of radiation has emerged. Besides outdoor measurements, passive dosimeters are also used for measurements in indoor environments [9-11]. Many developments in TLD materials and in TL readers (*e.g.*, automatization) [12] have led to increased use of this type of dosimeters in environmental dosimetry.

According to The European Radiation Dosimetry Group (EURADOS) survey conducted within dosimetry services in Europe, 83 % of the dosimetry systems are based on TL detectors (the most widely used TL materials are LiF:Mg, Ti and LiF:Mg,Cu,P). The RPLD is used by 7 %, OSLD by 3 %, followed by direct ion storage (DIS) dosimeters, CR-39 and fission track detectors by 3 %, 2 % and 2 %, respectively [5]. The basic demands for a TLD are good reproducibility, low hygroscopicity, and high sensitivity for very low dose measurements or good response at high doses and in mixed radiation fields [5, 13, 14].

Many environmental TLD are based on the LiF with different dopants, and as aforementioned, the two of which are most commonly used: LiF:Mg,Ti and LiF:Mg,Cu,P. The different TLD materials are analyzed elsewhere [15]. Initially, a LiF:Mg,Ti, was used, as probably the most studied and exploited TL material in radiation dosimetry. The progress in TLD environmental systems is mainly related to the implementation of the tissue equivalent high sensitivity material LiF:Mg,Cu,P [4, 16-18].

Although the choice of the TL material has a major influence on the measurement results, the design of the whole TL dosimeter has a significant role. The TL detector becomes a TLD after placing it in an adequate marked holder with dedicated filters the purpose of which is to establish the secondary charged particle equilibrium during irradiation. Filters usually absorb the low energy radiation and provide the appropriate measurement of the operational dosimetric quantity. Moreover, filters should minimize measurement result's dependency to the angle and energy of incident radiation. In environmental monitoring, where long-term field irradiation is sometimes carried out under extreme climatic conditions, the holder serves also to protect the detector against UV/sunlight, humidity, dirt and mechanical damage [15]. In many cases, dosimeters designed for individual monitoring are used for environmental monitoring without further method modification except for calibration.

The use of TLD for determining the dose from the penetrating components of environmental radiation requires extraordinary efforts in TLD measurement system calibration, dosimeters handling, readout and data interpretation without which the inferred average dose rates for the particular exposure period employed will be essentially meaningless [4, 5, 16, 19]. It is important, therefore, to obtain meaningful measurements and to distinguish increase in radiation dose (*i.e.*, the contribution from a nuclear facility) from that due to natural environmental radiation (background radiation) fluctuations. In many cases, the contribution from natural background is 10 times higher compared to dose from a nuclear installation that needs to be assessed, thus fluctuations in background must be determined accurately, in order to assess the contribution from man-made sources of radiation [20].

Over the decades, extensive investigations have been made of the different TLD systems' properties with an aim to reduce the overall uncertainty of dose measurement caused by environmental factors like temperature, humidity, dust contamination, *etc.* [17, 21]

The quantified requirements for TLD or other passive dosimetry measurement systems are outlined in the IEC standard 62387 [22] where the type test procedure is described together with guideline for the statistical analysis of the test results.

Apparently, national practices may significantly differ with respect to the period of monitoring and performance criteria for environmental dosimetry systems [5]. Moreover, general guidance or technical recommendations for the use of TLD method for environmental monitoring do not exist, nor the guidance on how to improve and adapt the individual monitoring method (adaptation of holders, filters, detectors and specific steps) to use as environmental monitoring method. Therefore, the procedures involved in selecting, testing and using a TLD system for environmental dose measurements and the interpretation of these measurements, discussed in this paper, are based on the extensive review of the available scientific data.

METHOD

The data presented in this paper are compiled from scientific papers and standards published over five decades, in the period from 1974 to 2021. In total, 63 references related to the subject of this paper were collected. The review is presented in the form of the narrative review, in which the collated scientific information is presented in a qualitative manner.

The relevant literature sources were identified by using available scientific and general search engines and research paper databases, including: ScienceDirect, Google Scholar, Scopus, Google Search *etc*. The key words that were used in the search include: thermoluminescent dosimetry, environmental dose monitoring, annealing, fading, readout, calibration, transit dose, preheating, fading *etc*. The key words were used alone and in different combinations and variations of the key words. Additional sources were identified by checking the literature section of the already identified papers, and also in other activities of the *Preparedness* project.

The information that is included in the following sections has been selected to give answers to the question which steps and measuring procedures are performed by measuring bodies and services for environmental monitoring using passive area dosimeters, in the first place in Europe but also in other parts of the world. Some of the specific topics that are discussed in the following sections are annealing, heating rate, calibration, fading correction, measurement uncertainty, comparisons *etc*. The findings were used to give a list of recommendations with the goal to help the standardization of measurement procedures.

SOLID STATE DOSIMETRY BASED ON LUMINESCENCE

The most often used solid state dosimeters for environmental monitoring are those based on thermoluminescence (TL), radiophotoluminescence (RPL), and optically stimulated luminescence (OSL) mechanisms. These three types of dosimeters are based on the principle that energy absorbed from ionizing radiation and stored in detectors' material is released in a form of light (luminescence) by exposing them to heat (TLD), ultraviolet or visible light (RPLD and OSLD) [23, 24]. This review is predominantly concerned with the TLD, whereas the OSL and RPL dosimeters are briefly discussed.

There are materials that could be used both as TLD and OSLD. Aluminum oxide Al_2O_3:C has been proposed as a high sensitivity TL detector, showing a higher sensitivity than LiF:Mg,Cu,P, though it is not tissue-equivalent and presents some important drawbacks such as the noticeable light induced fading and sensitivity changes with heating rate. However, the greater potential of aluminum oxide is in its use as OSL dosimeter. Furthermore, considering that OSLD obviates the necessity to heat the detector, the instrumentation and control parameters are drastically simplified. This enables production of portable OSL readers. The published study [4] suggests that the Al_2O_3:C as an OSL environmental dosimeter can easily measure doses around 1 μGy and no measurable fading has been observed after 80 days at room temperature.

The principle of RPL is applied to the glass dosimeter. The silver activated phosphate glass irradiated with ionizing radiation emits luminescence when exposed to UV light. The main advantage that RPL and OSL dosimeters hold over the TLD is that while luminescence centers of TLD disappear by readout process making the repetition of measurement impossible, RPL and OSL dosimeters can be repeatedly read [7, 25, 26]. Additionally, RPL systems do not require any fading correction.

Instruments that are made to stimulate and collect luminescence from TL, RPL or OSL dosimeters, converting it to dose value are called readers. The whole process of acquiring signal from these dosimeters is called reading or readout. It is important to note that dose value can only be obtained if a system of the reader and dosimeters are working correctly, and consequently we refer to it as a dosimetry system [22].

PROPERTIES OF TLD SYSTEMS

Ideally, the selection of TL materials and the design of TLD systems should be made with the objective of optimizing specific properties of the system. Properties with description and influencing parameters are listed in tab. 1.

Detection threshold

The detection threshold is determined as the lowest dose that can be detected with an acceptable confidence level, which is defined as three times the standard deviation of the reading at zero dose [14]. This value measured in a research laboratory under controlled conditions may be different from the threshold encountered under field conditions.

Sensitivity

The most obvious advantage of LiF:Mg,Cu,P arises from its high sensitivity (up to 40 times more sensitive than LiF:Mg,Ti), in addition to the other particularly useful TL characteristics (*e. g.* good energy response, insignificant fading and lack of supra-linearity). Materials like this permit detecting doses in the μSv range and reducing the environmental exposure period to about one day. However, there is no ideal material, thus the drawbacks for LiF:Mg,Cu,P and high sensitivity TL materials are mainly derived from the practical temperature limit for heating or annealing that leads to the relatively high residual signal.

Dose resolution

The capability to distinguish dose resolution between two dose values, which are very close, is also an

Table 1. Basic properties of TLD system (adopted from [14, 22, 27])

Property	Description
Zero dose reading (background)	Background is the lowest reading obtainable from the material and is based on readout shortly after annealing (with the assumption that the contribution from ambient radiation is negligible). Infrared signal and reader's photomultiplier tube (PMT) dark current may be significant contributors. When expressed in units of a dose, this value will be largely a function of the sensitivity of the material.
Sensitivity	The amount of light output per unit mass per unit dose. The light output is usually expressed in terms of the charge collected by the photomultiplier. Sensitivity for a given material is influenced by the heating rates and the area of the glow curve that is integrated for dosimetric purposes. When only stable peaks are used for dose determination, and/or post-irradiation anneal treatments are used to reduce fading, the sensitivity may be significantly reduced.
Reproducibility	Expressed as the standard deviation of repeated readout values obtained from the same detector, when given the same dose. Reproducibility depends to a large extent on readout equipment and methods, but also on the dose level. It is generally better with linear heating and with non-contact heating, such as hot gas or infrared heating. Reproducibility at dose levels more than 10 times the zero dose will be substantially better than at dose levels at or near the zero dose. Monitoring period should be selected in such way to ensure that the signal is significantly higher than the zero dose reading.
Detection threshold	Dose level that is significantly different from the background and can be reliably detected with the given system.
Linearity	Degree to which the reference and measured dose values agree over the range of a dose for which system will be used.
Fading	Post-irradiation fading is loss of a signal with time after irradiation and pre-irradiation fading is loss of sensitivity before irradiation. Fading can be caused by exposure to light or heat, optical fading and thermal fading, respectively. With proper anneal treatment and readout methods, fading can be controlled, as well as with introduction of control dosimeters.
Energy dependence	The degree to which the thermoluminescence signal is directly proportional to the reference value over the range of radiation energies intended to be measured.
Light sensitivity	The degree to which the light influences the response of the TLD to the ionizing radiation. Light can influence the TLD response by producing its own signal or by reducing the signal caused by exposure to ionizing radiation.
Moisture sensitivity	The degree to which moisture influences the response of the TLD to the ionizing radiation or the degree to which the material responds directly to moisture.
Angular dependence	The degree to which the dosimeter response is dependent on the angle of incidence of the radiation. Angle of incidence for reference dosimeter orientation is by convention 0°
Dose resolution	The capability to distinguish two dose values which are very close

important feature. Dose resolution is obviously closely related to the total uncertainty of the system. Therefore, a good system should be able to detect environmental dose variations of 10 % in a consistent way. This property depends very much on thermal treatments and monitoring period. In this sense a fast-cooling rate after high temperature annealing (240 °C) has been suggested to reduce loss of sensitivity with repeated short monitoring periods (*e. g.*, one week) [18].

Zero dose reading

Zero dose reading is the readout of results of unirradiated dosimeters, following annealing. The zero reading is related to signal due to detector itself and its previous irradiation and readout history as well as to the dark current from the photomultiplier. The typical values of zero reading for a range of different dosimetry systems indicate that zero dose reading ranges from 2 to 200 μSv for LiF materials and from 0.3 to 5 μSv for CaF_2 and $CaSO_4$ systems [20, 28]. The variability of zero reading components is quantified by corresponding standard deviation of readout for ten days, in which dosimeters are reused. It has been demonstrated that high values of zero reading are predominantly related to the quality of detectors [20, 28]. Therefore, improved dose measurement at low dose range can be achieved by selection of good quality dosimeters.

Monitoring period

The TLD system is composed of TLD material, a TLD reader, a dosimeter holder, a calibration and dose calculation method, and storage and/or deployment methods. If different TLD systems are used over the different monitoring periods at particular location, it is not appropriate to compare such systems without accounting for the potential differences in response [27].

Environmental TLD monitoring period, however, ranges normally from one month to one year and therefore other features such as self-dose, thermal stability or sensitivity changes with re-use are more important than threshold [18]. In fact, all steps of the measurement cycle, such as annealing, package and storage, transportation, irradiation, readout and mathematical evaluation influence properties and uncertainties of the dosimetry system [15].

An example of four different systems used for environmental monitoring over decades is presented in the tab. 2.

The analysis of variations in ambient dose observed during the decades revealed that trend does not

Table 2. Some parameters of environmental dosimetry systems (adopted from [27])

	Design 1	Design 2	Design 3	Design 4
TLD	CaF_2:Dy (TLD-200)	CaF_2:Mn (TLD-400)	^{7}LiF:Mg,Ti (TLD-700)	^{7}LiF:Mg,Ti (TLD-700)
Anneal procedure (pre-irradiation)	Oven anneal: 1 h at 400 °C 2 h at 100 °C	Oven anneal: 1 h at 400 °C 2 h at 100 °C	Reader anneal: 2 read cycles 20 s at 300 °C × 2 Oven anneal: 16 h at 80 °C	Reader anneal: 1 cycle 39 s (14 s at 300 °C) Oven anneal: 16 h at 80 °C
Anneal procedure (post-irradiation)	Oven anneal in capsule: 15 min at 80 °C	None	Oven anneal: 30 min at 80 °C	None
Readout procedure	Preheat pan to 90 °C for 10 s 90-275 °C/15 s	Linear heating: 25-150 °C/2 s 150-325 °C/18 s integrate glow: 150-280 °C	Oven anneal: 30 min at 80 °C Readout: 20 s at 300 °C Integrate glow 20 s	Linear heating: 50-300 °C/25 s 300 °C/8 s Integrate glow 15-30 s (150-300 °C)
Approximate detection threshold	0.01 mSv	0.001 mSv	0.01 mSv	0.01 mSv
Field cycle	Monthly	Monthly	Quarterly	Quarterly
Post irradiation fading	est. at ~12% in 4 weeks	20 % in 4 weeks	20 % in 4 weeks	15 % per year
Fade correction applied	NA	10 % monthly	None	5 % quarterly

reflect an actual change in dose rates in the environment, but rather a change in TL materials, shielding, or laboratory practices during these years.

Type-test

As a result of different TLD systems having different response, all passive integrated systems intended to measure dose equivalents $H^*(10)$, $H'(3)$ and $H'(0.07)$ for environmental monitoring should be type tested, according to [22]. Besides radiation performance and the design requirements, the standard also defines requirements for software, data and interface of the dosimetry system, as well as environmental, electromagnetic and mechanical performance requirements. Testing of 12 passive dosimetry systems, including the linearity, energy dependence, angular dependence, reproducibility, statistical uncertainty and response to natural radiation, was recently also performed within the Preparedness project [29].

ROUTINE PRACTICES IN TLD ENVIRONMENTAL MONITORING

The use of TLD systems in individual monitoring is well established and can be further modified to address the needs of environmental monitoring. The modification is based on the better control of the monitoring process and influencing factors that contribute to the overall uncertainty of dose measurements. The following elements of the monitoring process are considered: batch uniformity, annealing, fading reduction, dosimeter calibration, and system calibration. Furthermore, in routine monitoring procedure the following parameters have to be optimized in a performance test: temperature and time of preheat, heating rate during readout, temperature and time of readout. Preheat (sometimes called post-irradiation annealing) is used to eliminate the low temperature peaks of the glow curve *i. e.*, to reduce fading [15].

A choice of an adequate temperature-time profile (TTP) 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 TLD [30]. It has been observed that the kinetic parameters, E, and s in eq. (1) are strongly dependent on the cooling rate of the pre-irradiation high temperature annealing and the heating rate during the readout

$$p = s\mathrm{e}^{-E/kT} \tag{1}$$

where p is the probability of an electron release from the forbidden band to a conductive band per unit time, s – the so-called frequency factor, E – the activation energy, k – the Boltzmann's constant, and T – the absolute temperature.

For example, glow peaks of the LiF:Mg,Ti undergo an important modification following post-irradiation annealing at 165 °C, which depends on the preheat time [15, 31]. A similar shift in glow peaks of LiF:Mg,Cu,P with increasing heating rate to the temperature range where irreparable damage takes place has been demonstrated. Much the same effect was observed in Al_2O_3:C, and, therefore, these materials have limited use with environmental monitoring readers with fast heating rate [15, 32, 33].

For example, Omanware *et al*. [34] investigated the effect of heating rate by heating a sample made of LiF:Mg,Cu,P and LiCaAlF6:Eu at different heating rates, from 2 °C per second to 20 °C per second. Not much loss in TL intensity was observed at different heating rates. However, the main glow peak shifts from 220 °C to 232 °C gradually, with the increase in the heating rate [34].

Table 3. The example of procedure's parameters for different TL materials (adopted from [14])

	LiF:Mg, Cu, P	LiF:Mg, Ti, CaSO4:Dy
Preheating temperature [°C]	100	50
Preheating time [s]	12	5
Preheating speed [°Cs^{-1}]	8	10
Max heating temperature [°C]	240	350
Acquisition time [s]	20	30
Annealing temperature [°C]	240	350

In practice, a heating rate of 10 °C per second is considered to be adequate. In general, properties of the reading cycle depend on the type of detector. An example of parameters used during the readout process is given in tab. 3.

Batch uniformity – detector selection

Sensitivity of individual detectors within a batch is varying due to non-uniformity in TL crystal. The parameter – relative standard deviation from ten representative detectors irradiation to a selected dose may be used to characterize this effect. The results of assessment of this effect indicate that variation within a same batch is typically 10 %, but can reach even 18 % [20, 28]. The careful selection of detectors or application of individual sensitivity correction factors to each detector may reduce this variation to about 1 %. On the other hand, for RPL detectors (type FD-7) batch uniformity expressed as coefficient of variation of the indicated dose value varied between 0.013 and 0.037 in 10 measurement cycles [26]. The reproducibility of dosimeters in 10 measurement cycles was less than 2.7 % [26, 35]. According to [22] batch uniformity is expressed by coefficient of variation (CoV) and the acceptance values are max 15 % for doses lower than 0.1 mSv, and 5 % for doses higher or equal to 1.1 mSv.

Annealing

Annealing is an essential pre-irradiation heat treatment procedure utilized to achieve reproducible results. High temperature annealing is required in order to maintain the characteristics of a TLD (shape of the glow curve, sensitivity, background signal) after repeated irradiations. In particular, proper selection of annealing temperature is important in order to get the highest TL sensitivity and to eliminate the effects of the previous irradiations. It is also very much useful to avoid high temperatures that may result in permanent loss of TLD sensitivity [34].

The annealing procedure develops in the following way: the detector is heated and kept at a given temperature for a certain time, after which it is cooled with a constant cooling rate to ambient temperature. Factors as heating and cooling rate, as well as the annealing temperature have significant impact on the shape of the glow curve. Therefore, it is important that all detectors within a batch are annealed under the same conditions. There has been an extensive research of the heat treatment effect on the properties of LiF: Mg,Ti dosimeters that shows the importance of annealing [15, 35-37]. For example, the investigation of different annealing regimes of LiF:Mg,Ti detector, including heating at 400 °C followed by: slow cooling, fast cooling or 20 second readout process without high temperature annealing, revealed that fast cooled regime maintains the integrity in the best way, in terms of sensitivity. The regime without annealing demonstrated sensitivity change less than 10 % for 25 cycles. This indicated that in situation when sensitivity is the only parameter of interest, significant amount of time can be saved, using this regime [37]. In [34] it was demonstrated that annealing at 240 °C in the TL reader was sufficient to restore the original TL sensitivity of the LiF:Mg,Cu,P (MCP) and to remove the previous dose. Heating in the oven at any higher temperatures reduces the output of this type of detector.

Fading reduction

Fading, a spontaneous loss of the signal or the stability of dosimeters output signal under various climatic conditions as a function of time, is very important in the environmental monitoring since in the environment mostly long-term, outdoor exposures are the subject of the investigations [15, 26, 38]. The ambient temperature can reach 60 °C, which may cause a loss of signal within the TLD material. The fading of the TLD varies significantly for different TLD materials, as a function of temperature and irradiation time. It has been demonstrated that fading can be reduced through optimized readout process and by selection of suitable annealing and post-irradiation treatment [15, 20, 28, 38].

The fading depends 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), and on the climatic and light conditions during exposure. As temperature is of utmost importance in thermoluminescent dosimetry, the ambient temperature of the environment in which the TLD is deployed has a profound effect on its response, with higher temperatures resulting in higher pre- and post-irradiation fading rates. Accordingly, it has been observed that fading is more pronounced in summer months [39]. Accurate correction for temperature effects is difficult to make. In practice, high temperature fading during summer months can be minimized by proper design of dosimeters, whereas knowledge of fading dependence on the annealing procedures can further reduce the uncertainty of measurement [39]. It has been demonstrated that accuracy of measurement can be improved by correcting the actual field dosimeters' response due to fading, during the monitoring period, by using control dosimeters at representative point in the environment. These dosimeters should be exposed to the same environmental conditions and ambient temperature profile during monitoring period.

The fading correction can be carried out in different ways. One possibility is a 24 hours storage before reading of calibration and field dosimeters, during calibration procedures. In most cases the fading curves decrease exponentially in the first 24 (or 48) hours and later become linear. Another method is the thermal cleaning procedure, *i. e.*, preheat to eliminate the low temperature peaks [15]. An absence of fading is reported for the LiF:Mg,Cu,P detectors over a period of 15 days [34]. Table 4 presents the fading of various dosimeters previously stored 24 hours after irradiation.

Furthermore, the effect of readout and post irradiation treatments are available in the literature [15, 20, 28, 40-42]. The post irradiation treatment may have an impact on the fading properties of the TLD. It has been demonstrated that temperature-dependent fading, in particular for LiF, is independent of the time of irradiation within the monitoring period of 100 days. The simple glow curve structure of LiF:Mg, Cu,P, when read with pre-heat at 165 °C, offers a negligible fading in an extended period up to four months and great reusability of over 1000 uses [16]. Therefore, special attention should be paid to the handling of this type and dosimeters alike [4, 16, 18].

Several well-conducted studies [43-46] have investigated the pre-irradiation and post-irradiation fading rates of LiF:Cu,Mg,P. These studies have reported the exponential decay of third glow curve peak both before and after an irradiation and an increase in the total area of fourth peak during pre- and post-irradiation fading [43, 45]. The peak four has been shown to have a high thermal stability and fades independently of the temperature whereas the low temperature peaks decay exponentially and are strongly dependent on temperature [43]. The total TL output has been reported to post-irradiation fade up to 26 % [46]. The peak area ratios involving lower temperature peaks were found to decrease more quickly than those involving higher temperature peaks. These results indicate that pre-irradiation fading represents a true TLD sensitivity change before irradiation, which occurs as independent of any background radiation dose that may be present during storage before irradiation.

The individual peak fading of CaF2:Dy was estimated to be 25 % in one month without any post-irradiation treatments. Applying a post-irradiation annealing (preheating) of 80 °C for 10 minutes removed the lower temperature peaks, reducing the fading to 13 % in the first month. When the glow curve for CaF2:Dy was treated as being comprised of two main peaks, 45 % and 12 % fading in the peak areas in 35 days at room temperature was reported [47].

A simple method that avoids the need of a fading correction for LiF:Mg,Ti was proposed by Weinstein *et al.* [40]. The method is based on locating the channel with the peak maximum and integrating the counts in a defined region around this channel (not the whole peak area). Sachar and Horowitz [41] showed an exquisitely complex time-dependent response of the peak 4 and 5 in TLD-100. Peak 4 grows at the beginning of storage and begins to decay only after several months, while peak 5 decays rapidly from the beginning. Peaks 4 and 5 have a yearly fading of about 8 % when not measured separately. This indicates the very complex background of the fading, requiring a more detailed analysis in order to select an adequate fading correction among many proposed. The differences in the stability of the various peaks could be useful in estimating the time between exposure and readout [15].

Another study [43] investigated the thermal stability of LiF:Mg,Cu,P compared to the more traditional LiF:Mg,Ti at 40 °C and 70 °C, to evaluate the relative importance of the temperature/storage induced effects on either traps or trapped charges. The obtained results confirm that, for both varieties of LiF phosphor, the process affecting their respective main dosimetric peaks is not fading. On the contrary, the observed variations in the TLD response should be addressed to the modifications experienced by the trap system during storage. In the case of LiF:Mg,Cu,P the higher thermal stability exhibited by this material at 40 °C and 70 °C suggest that the sensitivity correction, so important for LiF:Mg,Ti, may not be needed. This fact, along with the use of unannealed dosimeters, is feasible with this material. The long-term measurements of fading showed that the fading effect for RPL dosimeters is less than 1 % for period of 30 days. The

Table 4. Fading of various dosimeters previously stored 24 hours after irradiation (adopted from [15])

		Detector type				
		LiF:Mg,Ti (TLD-100)	^{7}LiF:Mg,Ti (TLD-700)	^{7}LiF:Mg,Cu,P (TLD-700H)	MgB_4O_7:Dy	CaF_2:Mn
Post-irradiation period [d]	Dose [mGy]	Fading [%]				
14	2	0*	1*	1*	–	–
21	200	–	–	–	10*	8
30	0.2	–	–	–	18	5

* indicate post-irradiation annealing at 100 °C for 20 minutes

RPL dosimeter systems are insensitive to ambient temperature and humidity and do not require any fading correction [7, 25, 28, 35]. On the other hand, similarly to TLD, OSLD also exhibits fading that could be nearly as much as 70 % after 30 days from the irradiation [48], material dependent. This can be corrected by optical pre-reading treatment and minimized to a few percent.

Dosimeter batch calibration

The calibration of dosimeters is performed using reference dosimeters, for example after the field cycle and before readout [28]. Dosimeters that were stored or used can be annealed before calibration. The transfer of trapped electrons from deep to shallow traps results in an increased response of annealed dosimeters, followed by temperature dependent fading. Nevertheless, the application of annealing technique allows response relatively independent of the storage time. For the calibration of dosimeters without annealing, one should use the reference dosimeters with the same annealing and thermal treatment as field dosimeters. These reference dosimeters should be also stored at measurement locations, during monitoring period under same conditions as the field dosimeters [20]. If field dosimeters are annealed, the reference dosimeters may be annealed at any time before the readout, provided that annealing technique is well established. Both approaches ensure that the reference dosimeters have same fading properties as field dosimeters. This eliminates need for fading correction. Appropriate calibration technique should be established for each dosimetry system used for environmental monitoring owing to the differences in detector types and readout procedure [20]. According to [15], as previously noted, the annealing is very important and should be adjusted to various detectors. The pre-irradiation annealing parameters and effect on TLD response and long-term stability for LiF:Mg,Ti, CaF_2:Tm, $CaSO_4$:Dy, Al_2O_3:C, LiF:Mg,Cu,P, LiF:Mg,Cu,Na,Si are given elsewhere [15, 49-55].

System calibration procedure

A dosimetry system can be generally used for environmental monitoring only if combination of dosimeter, reader and monitoring procedure has been optimized. In case of the long term exposure in the environment [28], it is suggested that the repeated calibrations of the system and assessment of the related uncertainty budget should be carried out for each field cycle. A dosimeters batch is divided into several different sets for the calibration and control of the system, as shown by the example in tab. 5. These sets are used for assessment of reader's calibration factors, zero dose reading of the batch, the actual fading, transit exposure and field measurement.

Both reference (control) and field dosimeters are prepared or annealed together before use and later, evaluated after monitoring period.

Table 5. Sets of TLD and RPL dosimeters of the same batch for the calibration of dosimetry system for the readout of field dosimeters (adopted from [28])

	Number of dosimeters		Purpose
	TLD	RPL	
Reference/control dosimeters (5 sets)	10	3	Calibration
	10	3	Zero dose reading
	3	–	Zero dose correction for calibration factor
	3	–	Fading (exposed at field location)
	3	3	Transit dose
Field dosimeters	3	2	Dose measurement at locations (1, 2, 3,… *n*)
	2	2	
	⋮	⋮	
	2	2	

Irradiations of calibration dosimeters should be performed in reference fields established according to ISO 4037:2019 standard [56]. The most frequently used radiation qualities are S-Cs and S-Co. If the field is known at the place of the measurement, the calibration could be done with the quality that has similar mean spectrum energy. Thus, it is recommended in [57, 58] that calibration should be done in photon fields, preferably in a sealed ^{226}Ra source.

Although [28] suggests frequent calibration procedures, this is not feasible for many laboratories due to high cost of dosimeter irradiations in facilities that are traceable to primary dosimetry standards. Besides the cost issue, the logistics of sending/receiving the dosimeter to/from irradiation facilities could be demanding, and even could introduce new influencing factors for dose estimation (*i. e.*, transport dose for control dosimeter). The technical recommendations published for personal monitoring [59] suggest that reader calibration should be done regularly, every two years, and dosimeter calibration should be done also in every two years or every ten readouts (whichever comes first). It is suggested that if this calibration period is applied, more frequent periodic performance test should be carried out, not necessarily by utilizing reference irradiation fields.

MEASUREMENT MODEL

The mathematical, measurement, model is a function between input and influence quantities, and the output quantity. The model can vary depending on the TLD system and the dosimetrist's knowledge about its properties and influencing quantities. An example of a measurement model is given below.

The field dose, D_1, is calculated from the results of readout of field dosimeters $A(F_1)$ [28], as

$$\begin{array}{c} D_1 \quad NC_E C_\beta C_\mathrm{f} \\ A(F_1) \quad A(T) \quad C_\mathrm{tu} A_\mathrm{u}(F_1) \quad A_\mathrm{u}(T) \end{array} \tag{2}$$

where D_1 is accumulated dose from the set of field dosimeters F_1 at location *1* after the monitoring period, $A(F_1)$ and $A(T)$ – the mean readings of the field dosimeters and transit dosimeters after the monitoring period, respectively. $A_u(F_1)$ and $A_u(T)$ – the mean readings before the monitoring period (zero dose readings), and C_E, C_β, C_f, and C_{tu}, correction factors for the photon energy – E, angular response – β, fading – f, and time dependent correction of the calibration factor N for the reading of the unirradiated dosimeter – tu. These corrections account for any variations compared to irradiation parameters during calibration. The calibration factor, N, is related to the mean readings of the control sets for calibration, s, and zero reading, u, after irradiation with reference dose, D_r, according to

$$N = \frac{D_r}{A(s) - A(u)} \tag{3}$$

The measurement uncertainty is derived from the measurement model equation evaluation.

Factors contributing to the uncertainty budget

Overall factors contributing to the uncertainty budget are field dose reading, system calibration, individual detectors sensitivity, fading correction, reader stability, transport/transit dose, zero dose, energy dependence, angular dependence, light and humidity impact, long-term stability, *etc*. The majority of them, as influencing quantities, are shown in eqs. (2) and (3) [28]. The influence of appropriate calibration quality is underlined in [58]. In the most recent study [60], a detailed analysis of measurement uncertainty for passive dosimetry systems was performed and a few conclusions were drawn:

– detection limits were calculated to vary between 51 μSv per month and 86 μSv per month,

– the four participating laboratories had substantial differences and only a few conformances in their methodologies including that no one from participants used correction for non-linearity, signal fading and environmental influences, and

– the uncertainty of measurements in emergency situations (short measurement period) is relatively high at low dose rate levels and the use of more detectors for each dosimeter would reduce the final uncertainty.

Failing to identify one or more significant sources of uncertainty will cause the underestimation of the combined uncertainty of measurement. Therefore, a thorough analysis of the measurement system and procedure needs to be performed. For an adequate uncertainty analysis, data on metrological properties of the detectors are also needed, such as the data available in the recent study on twelve passive dosimetry systems [29]. Of course, only the data for the specific passive dosimetry system can be used for uncertainty evaluation.

Relative empirical standard deviation of a single readout

The calculation of relative empirical standard deviation of a single readout is explained elsewhere [20, 28]. With respect to the precision of measurements in the low dose range, one can use relationship between empirical relative standard deviation and dose curve to indicate which parameters are significant for the uncertainty of the dosimetry system. The relative empirical standard deviation depends on dose differently, depending if individual detector calibration or batch calibration was used, which TTP was used for the readout or if zero dose correction was used.

INTERPRETATION OF THE MEASUREMENT RESULTS

As mentioned in the *Introduction*, the total dose measured in the environment depends on many factors and has several contributors. In addition to natural radiation, the contribution from man-made ionizing radiation sources should not be neglected [15], or the transit dose.

Natural background

Environmental monitoring aim is to estimate variation in natural background and assess any additional dose from man-made sources of radiation, during a time period and at different locations. Hence, the precise measurement of natural dose rate, $\dot{D}_{nat}$, is required to interpret the measurement results. The minimum and maximum monitoring period should be found in order to precisely measure $\dot{D}_{nat}$.

A minimum monitoring period, t_{min}, can be found for each system [28] from

$$D_L = \dot{D}_{nat} t_{min} \tag{4}$$

where D_L is minimal dose measured with standard deviation of 10 %, and $\dot{D}_{nat}$ – the natural background dose rate.

The maximum monitoring period is limited by finding properties of a dosimeter. This period can be found by type-test procedure given in [22]. The monitoring period should satisfy the criteria $t_{min} \le t \le t_{max}$. If this is fulfilled, zero dose reading variations are minimized or more than two dosimeters are used on site, the $\dot{D}_{nat}$ should be precisely measured with standard deviation of not more than 10 %.

Transit (transport) dose

Transit dose contributions may vary in different ways, depending on distance, transportation means and potential additional exposures. The contribution from transit dose is more significant in short monitoring periods and can even exceed the contribution from natural background. Since high transit dose may result in higher measurement uncertainty special attention should be made to reduce any unnecessary exposure by:

- shielding during transport and storage of dosimeters,
- avoiding long distances between a laboratory and the point of measurement,
- avoiding long storage periods at the point of measurement before use, or
- avoiding long storage periods before evaluation of dosimeters.

Dose due to man-made sources

For the assessment of the dose contribution of man-made sources, it is necessary to consider significant fluctuations in the natural background. These variations are related to the time and space (distribution of natural radionuclides) as well as to the climatic and seasonal effects. Natural background is estimated using a set of reference dosimeters, from several cycles in order to reduce uncertainty. For several short monitoring periods, uncertainty of mean value decreases with increased number of measurements, as $1/\sqrt{m}$, where m is number of monitoring periods per year. For a dose of 0.01 mSv, uncertainty is 42 % for one month, but 12 % after 12 months, when accumulated dose is 0.12 mSv. In general, the monitoring period may vary between 2 and 12 weeks for short term variation monitoring and from 3 to 12 months if long/term changes are to be monitored [28].

The EURADOS WG 3 survey on use of passive dosimeters for environmental monitoring in 2013/14 [5] investigated the dose calculation methods in different laboratories, taking into account that a dose can be influenced by different contributions, key elements of the dose assessment methodology are the contributions of the background dose and transit (transport) dose, *e. g.*, the transit dose can account for up to 35 % of the measured dose if the transit period is long compared to the monitoring period [5]. In addition, detector readings are usually multiplied by many correction factors. Therefore, the survey was focused on the following elements: net dose calculation and applied methodology to measure/estimate the background dose, transit dose correction and applied methodology to measure/estimate the dose contributions not related to the exposure at the measuring location, fading or climate correction methods, other applied corrections and overall measurement uncertainty.

The participating laboratories reported calculation of net doses (*i. e.*, they subtract the natural background from the dosimeter results) in about (30-50) %. Transit corrections are applied in about 50 % of the cases. About 40 % of dosimetry services applied fading corrections. Among services which apply methods for fading corrections, about half of them apply a fading or climate correction based on estimated values, while additional control dosimeters are used in about 40 % of cases. Other corrections are applied in 28 % of the cases, specifically individual correction factors for single detectors and other corrections taking into account linearity and energy dependence [5].

PERFORMANCE TESTS

The dosimetry system reliability and consistency can be checked by regular external performance tests [59]. There are several types of performance tests: blind, surprise and announced as shown in tab. 6.

Table 6. Performance tests

	Test	
Worker	Announced	Not announced
Aware	Intercomparison	Surprise
Unaware	/	Blind

In a blind test, a dosimetry worker is unaware of the test, and thus cannot choose better dosimeters or take special measures during preparation and readout. During a surprise test a dosimetry worker is aware of the test in terms that he can choose the dosimeters but cannot take extra special measures during dosimeters readout, because he is supervised.

Well prepared performance tests are necessary to ensure accuracy, to reach international standards and to improve environmental dosimetry methods. This can also be achieved through international intercomparisons [57, 58, 61] as well through interlaboratory intercomparisons [62, 63]. Intercomparisons provide unique opportunities to participants to check their calibration, the use of different corrections and the methods of measurements [15]. The establishment of these international intercomparisons allows a broad exchange of experience and review of available dosimetric systems. As a consequence, the calibration and measuring procedures used in particular laboratories, as well as the quality of the dosimetry systems employed could be improved step by step. In order to achieve realistic results, it is important that these intercomparisons are organized in the environmental conditions. An example of environmental dosimetry intercomparison during project PREPAREDNESS is given in [57], in which reference sites for environmental radiation and secondary cosmic radiation are described in detail.

CONCLUSION AND RECOMMENDATIONS

Various passive dosimeters, mainly TLD, are used for environmental monitoring in Europe. There is not much data about specific steps in the whole process of the environmental monitoring by using passive dosimetry systems. The published surveys and available data represent the main sources of information about different measurement systems that are in use, as well as the statistics of the applied dose correction methods. The monitoring process is very complex due to many influence quantities, some of which are the intrinsic properties of the system, and the others are consequences of the chosen monitoring procedures and steps.

In order to overcome the complexity of problem related to environmental monitoring, several actions can be implemented, namely:

- select/use dosimetry systems with proven intrinsic properties and with detection limit appropriate for the intended use,
- conduct type tests for dosimetry systems that ensure that the requirements set by environmental monitoring international standards are met,
- irradiations of dosimeters for system calibration, control or type-test should be done in irradiation conditions compliant with ISO 4037 standard series,
- minimum and maximum monitoring period should be defined and appropriate for the dosimetry system that is chosen,
- use a representative set of control dosimeters (to correct for fading and additional transit exposures),
- use more than two dosimeters at each measurement site,
- keep transit dose to a minimum, by reducing transit time, storage before use and storage before evaluation,
- identify all significant contributions to measurement uncertainty, based on the specific measurement procedure that is used and based on the properties of the TLD system,
- estimate the measurement uncertainty as a function of dose, and continuously improve the knowledge about the system that is in use, and
- regularly participate in performance tests and intercomparisons.

With appropriate choice of the dosimetry system, including the detector itself and readout process a wide dose range can be measured, including those relevant for environmental radiation dosimetry, where low doses are of interest.

Further work, based on the good understanding of the influence factors on the accuracy of dose measurements, is required to optimize and harmonize TLD environmental monitoring procedures.

ACKNOWLEDGMENT

This project (16ENV04 Preparedness) has received funding from the EMPIR programme co-financed by the participating states and from the European Union's Horizon 2020 research and innovation programme. The research has been additionally funded by the Ministry of Education, Science and Technological Development of the Republic of Serbia.

AUTHORS' CONTRIBUTIONS

M. Z. Živanović was responsible for the work planning and management. O. F. Ciraj-Bjelac collected the majority of data and made a structural organization of the findings. Z. I. Knezević and M. C. Majer gave substantial feedback, in addition to providing the data about RPLD. J. S. Stanković Petrović and N. Lj. Kržanović prepared and completed the manuscript. All authors participated in shaping the manuscript by providing valuable comments and suggestions.

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Received on January 26, 2021
Accepted on August 23, 2021

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ПРЕГЛЕД ТЕРМОЛУМИНЕСЦЕНТНОГ ДОЗИМЕТРИЈСКОГ МЕТОДА У МОНИТОРИНГУ ЖИВОТНЕ СРЕДИНЕ

Пасивни дозиметри чврстог стања, као што су термолуминесцентни дозиметри, дају интегрисано мерење укупне дозе, и често се користе приликом мониторинга животне средине. Циљ овог рада је пружање свеобухватног прегледа употребе методе термолуминисцентне дозиметрије у мониторингу дозе зрачења у животној средини. У раду је представљен део резултата истраживања у оквиру пројекта *PREPAREDNESS* (EMPIR позив 2016 за заједничке истраживачке пројекте *Метрологија за животну средину*), са специфичним циљем хармонизације процедура које користе дозиметријски сервиси, релевантна регулаторна тела и институти широм Европе. Да би се то постигло истражене су различите мониторинг рутине засноване на пасивним методама дозиметрије. Анализиране су разлике у извођењу појединих корака при мониторингу као што су предгревање, очитавање, брисање, спонтано губљење сигнала и други. Истраживање је спроведено квалитативним прегледом литературе који је показао недостатак информација о специфичним корацима. Закључак је да мерни систем базиран на термолуминесцентној дозиметрији, колико год био сложен, мора имати испитивање типа, контролни дозиметри треба да се редовно користе, током калибрације треба да се прате стандарди за озрачивање Интернационалне организације за стандардизацију, и поред осталог, параметри који утичу на мерну несигурност морају бити познати и добро разјашњени како би се добили тачни резултати мерења дозе.

Кључне речи: термолуминесцентна дозиметрија, мониторинг у животној средини, брисање, спонтано губљење сигнала, очитавање, калибрација, транспортна доза