Radiation-induced damage analysed by luminescence methods in retrospective dosimetry and emergency response

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Summary. The increasing risk of a mass casualty scenario following a large scale radiological accident or attack necessitates the development of appropriate dosimetric tools for emergency response. Luminescence dosimetry has been reliably applied for dose reconstruction in contaminated settlements for several decades and recent research into new materials carried close to the human body opens the possibility of estimating individual doses for accident and emergency dosimetry using the same technique. This paper reviews the luminescence research into materials useful for accident dosimetry and applications in retrospective dosimetry. The properties of the materials are critically discussed with regard to the requirements for population triage. It is concluded that electronic components found within portable electronic devices, such as *e.g.* mobile phones, are at present the most promising material to function as a fortuitous dosimeter in an emergency response.

Key words: optically stimulated luminescence, thermoluminescence, dosimetry, fortuitous dosimeters, population triage.

Riassunto (Il danno radioindotto analizzato con metodi di luminescenza nella dosimetria retrospettiva e nella risposta in emergenza). Il rischio crescente di uno scenario di ricovero urgente di massa in seguito a incidenti o attacchi radiologici su grande scala richiede lo sviluppo di metodi dosimetrici adatti alla risposta di emergenza. La dosimetria a luminescenza è applicata da decenni per una ricostruzione affidabile della dose in territori contaminati. Ricerche recenti hanno aperto la possibilità di stima della dose individuale per la dosimetria di incidente e di emergenza usando la stessa tecnica luminescente in materiali disponibili vicini al corpo umano. Questo lavoro esamina i dati disponibili in letteratura relativi alla misura con tecniche di luminescenza in materiali utili per la dosimetria di incidente e retrospettiva e illustra alcune applicazioni. Le proprietà dei materiali sono discusse criticamente riguardo alle esigenze di triage di popolazione. Si conclude che i componenti elettronici disponibili in molti dispositivi elettronici portatili, come per es. telefoni mobili, sono al momento i materiali più promettenti per funzionare come dosimetri fortuiti nella risposta in emergenza.

Parole chiave: luminescenza otticamente stimolata, termoluminescenza, dosimetria, dosimetri fortuiti, triage di popolazione.

INTRODUCTION

One of the important activities which have to be accomplished immediately after the occurrence of a mass casualty involving accidental or malicious use of radiation is the evaluation of doses absorbed by the individuals not carrying physical dosimeters, in order to carry out a *triage* of the population and to minimize the harmful or lethal effects of radiation [1]. An estimation of the dose absorbed by common use objects and biological tissues collected from ar-

eas and individuals may be performed through the identification of radio-induced defects with physical techniques such as electron paramagnetic resonance (EPR), optically stimulated luminescence (OSL) and thermoluminescence (TL).

Up to now, luminescence methods have been used in retrospective dosimetry of past exposures rather than in the response to emergency situations. The applications of the luminescence techniques in retrospective dosimetry are based on the utilization of

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the luminescent properties of some minerals, such as quartz and feldspars, allowing the assessment of the cumulative absorbed dose due to ionizing radiation. Unlike other physical and biological techniques, which are used for a more direct assessment of the individual dose through analysis on biological tissues, up to now luminescence measurements have been used only to assess absorbed doses in media of the environment, therefore leading to a precise evaluation of local doses rather than individual ones; in particular, they have been applied for the assessment of the external gamma radiation exposure of clay based ceramic materials containing feldspars and quartz, such as bricks, tiles and porcelains [2-4]. The method was implemented for the application to dose reconstruction at various sites such as Hiroshima and Nagasaki, the Chernobyl power plant, the Nevada and Semipalatinsk nuclear test sites, a house in Estonia in which an uncontrolled handling radioactive source was found and the settlements at the Techa River in Russia [5].

In order to obtain from local doses a reliable reconstruction of the dose absorbed by an individual or by a population group, it would be necessary to evaluate the radiation field in different places and, subsequently, the time spent by the individual in these sites of interest. Large uncertainties would be involved in such a complex process and data elaborations would be quite time-consuming. For these reasons, up to now luminescence techniques did not appear to be suitable for getting a rapid evaluation of the exposure level in emergency situations, when individual dose estimates need to be carried out very rapidly. Nevertheless, recent studies on the luminescent properties of new materials employed as resistor substrates for integrated circuits lead to the hypothesis of the utilization of commonly held personal objects as "fortuitous dosimeters"; in particular, chip cards and other electronic components found within portable devices (e.g. cell phones, portable computers, music and video players, camera and digital watches) have shown luminescence sensitivity, as well as glasses recovering monitor displays of electronic devices and watches. Some particular features of these personal effects make them suitable emergency individual dose indicators, e.g. they are usually positioned close to the body and are carried by a large percentage of the population.

In the following a review about the possible utilization of luminescence dosimetry is shown, concerning in particular its application on fired and unheated materials (such as building materials and household chemicals), mainly used for the assessment of local doses, and on the new materials potentially usable for the individual dose estimation in emergency situation.

FUNDAMENTALS OF LUMINESCENCE DOSIMETRY

Luminescence is the stimulated emission of light from an insulator or semiconductor following the previous absorption of energy from radiation [6]. If the stimulation is provided by heat, the emission is termed Thermoluminescence (TL), if it is provided by light it is termed optically stimulated luminescence (OSL) and if it is provided by infrared light it is termed infrared stimulated luminescence (IRSL). Luminescence may also be produced by other types of excitation than radiation (e.g. electrical, mechanical, chemical energy) but within the context of dosimetry, the excitation is always to be understood as nuclear radiation (i.e. alpha, beta, gamma radiation, X-rays etc.). The driving agents in producing TL or OSL in a material are defects in the material structure. In crystalline materials, the defects form metastable states in the forbidden zone between valence and conduction band and function as traps for free charge carriers (electrons/ holes), produced within the crystal by the ionizing radiation. If the energetic difference between defect ground state and conduction or valence band is sufficiently high, the trapped charge carriers will not be able to escape at room temperature and thus charges will accumulate within the defects as long as the radiation continues. Thus the amount of trapped charge carriers in a defect is directly proportional to the amount of absorbed dose of the material. In the laboratory the trapped charges are set free by supplying sufficient amount of energy either in the form of heat (TL) or light (OSL) and recombination with charge carriers of opposite signs can take place (electron-hole recombination). The recombination process can be accompanied by the emission of light which constitutes the luminescence signal (non-radiative recombinations, which will go undetected, are however also possible). In the simplest case of only one trap and one recombination center, the luminescence signal is proportional to the amount of trapped charges and thus directly proportional to the dose the material has received. For a quantitative determination the radiation sensitivity of the material has to be calibrated, by measuring the luminescence response to known laboratory doses.

Since the charge carriers (electrons and holes) are trapped in metastable states, there is a finite probability that they are thermally released from their trapping states to the conduction/valence band even at room temperature, leading to a loss in luminescence signal. Since this effect is strongly dependent on temperature, it is termed thermal fading. In order to avoid a systematic underestimation of the absorbed dose, the mean lifetime of the luminescence signal should be large compared to the time interval between exposure and measurement, in the case of a single event or the duration of irradiation, in the case of a continuous exposure. For some materials, however, a luminescence signal decay has been observed which is much faster than expected from pure thermal effects. This phenomenon is called *anoma*lous fading and it is due to tunneling of charge carriers from the trap to the recombination centre [6]. The main characteristic of the anomalous fading is

a rapid initial decay and then a gradual decrease of the decay rate for longer storage times.

When using fired or unheated building materials for dosimetry, sand-sized quartz grains or polymineral fine grains are often extracted as target materials. This requires sample preparation which usually takes several days. Personal objects like chip cards or portable electronic devices only require the isolation of the radiation sensitive components from the card or the circuit board (under dark room conditions), which can then be measured as they are. This can be accomplished within a few minutes.

The readout process is very similar for both TL and OSL measurements, except for the stimulation mechanism. During a TL measurement, the sample is placed in a stream of heated gas or on a thin metal heater, its temperature is linearly raised with time and the luminescence signal is detected by means of a photomultiplier (PM) tube, operating in photon counting mode. The light yield is then recorded as a function of the sample temperature in a glow curve. The quantity related to the total number of the emitted photons, and thus to the radiation exposure, is the area under the glow curve. For an OSL measurement, the sample is stimulated with a strong light source such as a laser or a high power light emitting diode and the signal is again detected with a PM tube. Generally, the stimulation is carried out with a continuous excitation source, like a continuous wave laser, illuminating the sample and recording the light yield simultaneously over a time period of many seconds. Other stimulation modes, such as linearly increasing the light intensity or pulsing the light source, are also in use [7, 8]. In order to discriminate the luminescence signal from the simulating light and to protect the PM tube, appropriate optical filters need to be inserted between sample and detection unit. Quartz grains from building materials are usually measured as multiple grain aliquots but instrumental developments in the last decade also allows the measurement of single sand-sized grains using a special sample disc and a focused laser [9]. In addition, modern research luminescence readers have the advantage of being equipped with a built-in radioactive sources (e.g. beta- or X-ray source) and being automated, so that calibration of the radiation sensitivity of a sample can be achieved efficiently.

An important advantage of optically stimulated luminescence lies in the possibility of reading out thermally deep traps in the material without the necessity of subjecting the sample to high temperatures. This can give an increase in sensitivity compared to TL, by avoiding the effect of thermal quenching, and can avoid strong changes in radiation sensitivity, which e.g. quartz displays after TL measurements above 300 °C [9]. The possibility of detecting OSL at room temperature also implies that more materials can be investigated than by TL, e.g. plastics. On the other hand, the need for optical filters, which must not necessarily encompass the wavelength range of the luminescence emission and

the need for an additional stimulation unit, which increases the distance between sample and PM tube, can also lead to a compensation of the increase or even to a decrease in sensitivity compared to TL. Only optically active defects can be measured with OSL, whereas a greater variety of defects will be detected by TL. This also implies that the sample must have been sufficiently shielded from any light after exposure and prior to measurement to avoid bleaching of the OSL signal. Which type of measurement is to be preferred for dose assessment will thus depend on the specific properties of each material.

REVIEW OF LUMINESCENCE FOR RETROSPECTIVE AND ACCIDENT DOSIMETRY

Fired building materials

Perhaps the most important contribution of luminescence techniques to dosimetry and certainly the most applications can be found in the field of retrospective assessment of external exposures of the population of a settlement [4]. The feasibility of using fired building material for luminescence dose reconstruction was shown over 40 years ago [11], and one of the first attempts at luminescence dose reconstruction was made using roof tiles in the Dosimetry System 86 for Hiroshima and Nagasaki [12]. In the following luminescence methods using bricks were successfully applied at the Nevada Test Site [13] and further methodological developments were achieved in the applications to areas affected by the Chernobyl accident [14] and the Semipalatinsk Test Site [15, 16]. Several luminescence studies have also been carried out in two former settlements of the Techa River valley, in the Southern Urals, which was contaminated by the release of radionuclides from the Mayak plutonium facility into the river during the period of 1949-1956 [17, 18]. Measured doses due to man-made source of radiation in the near surface layer of the bricks ranged from high values of 2-4 Gy for bricks from the highly contaminated former village of Metlino at the upper Techa River to comparatively low values of 100-300 mGy for bricks from settlements in the middle Techa River valley (Muslyumovo) and the fallout area of the Semipalatinsk Test Site. By measuring height profiles along building walls and dose-depth profiles into the brick, information on the source distribution and photon energy can be obtained [19]. These data can then be used as experimental constraints for refined methods of photon transport calculations using Monte Carlo methods [20]. By the Monte Carlo method coefficients are derived to convert the measured anthropogenic dose in brick to dose in air at a given reference location [21]. The air kerma values can then be directly compared with the calculated values of the respective dosimetry system used in epidemiological studies. In this way, e.g. for the former village of Metlino the value for the cumulative gamma dose at the Techa River

shoreline from 1949-1956 of 26.6 Gy, used in the Techa River Dosimetry System (TRDS-2000), was confirmed by luminescence methods [22].

When using bricks as natural solid-state dosemeters, the background dose to natural radionuclides occurring in the brick, mortar, plaster, the soil in front of the sampled wall of the building and the cosmic dose rate needs to be assessed and subtracted from the measured dose. The absolute uncertainty in the background dose is dependent on the age of the brick and limits the minimum detectable dose. From theoretical considerations a minimum detectable dose of less than 10 mGy was postulated for 20 year old bricks [23], but results from real-case applications put the detection limit more at 25 mGy for bricks with ages ranging from 30 to 100 years [15, 24, Woda et al. Radiat Environ Biophys, submitted].

Cement-based building materials are widely used in the construction of urban settlements and concrete in particular has become ubiquitous. The challenge in using unheated building materials for retrospective dosimetry lies in the highly heterogeneous degree of optical resetting of the sand grains in mortar or concrete during quarrying from geological deposits and use. Thus when applying multiple grain single aliquot techniques, large zero-dose signals will be observed. To overcome this problem, single grain techniques have been employed to identify and process only those grains, which had been sufficiently zeroed by exposure to light before fabrication [23, 25]. Due to the general low sensitivity to radiation of the quartz grains in concrete and mortar, a large number of grains (>104) has to be measured, resulting in extensive time-consuming measurements. Investigations on unexposed concrete blocks indicated that the single grain approach results in a minimum detection limit about 100 mGy. So far, however, the method has only been tested for doses of 2 to 10 Gy using mortar from a radioactive waste storage building or for doses from 0.5 to 5 Gy using a laboratory irradiated concrete block [26, 27]. An alternative approach has been suggested for mortar using the low temperature TL peak located at ~170 °C (5 °C s⁻¹) on extracted coarse grains of quartz [28]. As the lifetime of this peak is small compared to the geological age of the quartz, only a small "background" signal is found in this temperature region for unexposed mortar, corresponding to a dose of around 100 mGy. For a gamma irradiated block of render, the given dose of approx. 1.5 Gy could be recovered within error limits using a multiple aliquot regeneration protocol.

Next to concrete and mortar, household and workplace chemicals have also been investigated as potential fortuitous dosimeters in case of a radiological accident. Their advantage is that they can be measured as they are, without the necessity of lengthy sample preparation. Materials investigated using OSL comprise dish-washing powder, calcium-chloride in frost protection, common salt, glauber salt, washing powder and water softener

[29]. All these materials displayed a radiation induced OSL signal, with low or negligible zero dose signal. The dose response up to 16 Gy followed a single exponential saturation function, except for common salt, for which the growth curve seemed to contain two components. However, in a recent study the dose response of household salt was studied in more detail in the lower dose region and it was observed that the response was linear in the range of 1 mGy to about 100 mGy and became moderately supra-linear above that level, up to 9 Gy [30]. Estimated minimum detectable dose ranged from <1 mGy for common salt to 100 mGy for washing powder. Fading of the signal after two weeks of storage time was <7% for most materials. except for dish-washing powder and water softener, for which a signal fading of 40% and 25% was observed. It thus seems that the use of household and workplace chemicals in accident dosimetry should be seriously considered. As they generally can be found in only a few selected places in a household however, in contrast to the ubiquity of building materials and porcelain, they should be regarded more as a supplement to rather than a replacement of the existing materials.

Although showing very usable dosimetric properties and despite the fact that some have been reliably tested and used in numerous applications, all materials reported so far have the disadvantage of only being able to measure a local dose, albeit with high precision. Current research therefore focuses on identifying and characterizing new materials that are part of or carried close to the human body, are ubiquitously available and thus allow the estimation of individual doses.

Tooth enamel and dental ceramics

Tooth enamel hydroxyapatite has been extensively used as a biodosimeter with EPR in retrospective and accident dosimetry, with a lower detection limit of around 100 mGy for extracted teeth [31]. That it can potentially be also used as a biodosimetric material for OSL has been first shown by [32] using both IR and green stimulation on crushed teeth. However, with these stimulation modes only very large doses in excess of 120 Gy could be detected. With instrumental improvements in the last decade, it could recently be shown that the minimum detectable dose reduces to a value between 1.4 and 4 Gy for crushed teeth, when using blue stimulation with higher photon flux [33]. Linear growth of the OSL signal up to at least 414 Gy was observed. Fading of the OSL signal is quite serious at 37 °C (equal to body temperature) with an estimated, extrapolated total signal loss occurring 2 days after irradiation. Next to tooth enamel, the luminescence characteristics of dental ceramics have also been investigated [34]. Luminescence properties of three different types of ceramics studied were complex due to the nature of the material. Sensitivity to radiation is generally higher than for tooth enamel, with a linear

dose response of the TL, OSL and IRSL signal observed from <100 mGy to >5 Gy. Storage of the irradiated samples at room temperature and exposed to white light revealed a fast decay of the TL and OSL signal in the first two days (50% signal loss) and a slow decay for longer storage times up to approx. 30 days (additional 10% loss).

Although either not yet in a sensitivity range useful for emergency response (tooth enamel) or not well enough characterized for application (dental ceramics), both materials display potential for a possible future development of an in-vivo OSL dosimetry application. However, the unavoidable, highly variable exposure of human teeth or dental ceramics to daylight after an irradiation, which is difficult to quantify and therefore the large uncertainty in the degree of bleaching of the OSL signal prior to measurement poses a major challenge when using this approach.

Personal objects

One of the first materials identified, that are carried close to the human body and can be used as fortuitous dosimeters were certain type of telephone chip cards, that were in wide use in the 1990s before the advent of mobile phones [35]. Among these, the chip cards where the chip was covered with a transparent plastic cover could be measured using either TL or IRSL. However, in TL a very strong zero-dose signal was observed, corresponding to an equivalent dose of 3-5 Gy of the regenerated signal, rendering this mode of measurement unsuitable for dosimetric purposes. Using IRSL, the zero-dose signal corresponded to only 100 mGy, which marked the detection limit of this method. Measuring at 140 °C after preheating to 100 °C, the IRSL dose response was found to be linear in the dose region between 0.25 Gy and 5 Gy. A lifetime of the IRSL signal of 2.5 years at room temperature was determined from isothermal decay experiments, which would be sufficient for accident and emergency dosimetry. A similar study was later conducted on chip card modules used in civilian identification cards [36]. Using IRSL measured at room temperature, the dose response was linear from doses of 400 mGy up to 12 Gy. When measuring IRSL at 140 °C, the dose response became supralinear in the dose range between 0.15 and 0.63 Gy. In this work, the OSL signal using blue LEDs was also studied. With no preheating and measuring OSL at room temperature, the OSL signal was linear up to 10 Gy with an estimated minimum detectable dose of only 20 mGy. A fast decaying and a more stable component of the OSL signal was identified for delay times up to 10 hours after irradiation. Research was undertaken to identify the radiation sensitive components of the chip cards and it was found that the radiation induced luminescence signal originates from silica in the epoxy encapsulant, the latter protecting the chip and wiring from the environment and physical damage [37, 38]. Efforts were made to improve

the radiation sensitivity of the chip card, by adding various phosphor substances to the encapsulant before hardening [39]. The potential of OSL using blue LEDs for dosimetry was further investigated on chip card modules, which were manufactured using a world-wide spread UV-cured epoxy product for encapsulation and which find use in health insurance, cash and credit cards [40]. OSL properties of these types of chip card modules turned out to be complex due to the presumed thermo-optical release of electrons from the epoxy and transfer into the silica during stimulation. Best results and highest sensitivity were obtained by using no or only low preheat treatments and measuring the OSL at room temperature, using a single aliquot regeneration protocol with test dose normalization. The dose response of the OSL signal was linear up to approx. 7 Gy, with a minimum detectable dose of approx. 3 mGy immediately after irradiation. A high degree of fading of the OSL signal during storage at room temperature was observed, which was tentatively explained by the superposition of thermal decay of shallow OSL traps and athermal (anomalous) decay of deeper OSL traps. As a result, the minimum detectable dose increased to 20 mGy for dose assessment 10 days after exposure. Dose recovery tests demonstrated that given doses could be recovered within a deviation of $\pm 14\%$, if measured signals were corrected for fading.

Although chip cards thus show very promising OSL properties, which would make them suitable candidates for emergency response, a major drawback lies in the fact that only wire-bond chip cards with a translucent, either UV or heat cured encapsulation display a radiation sensitive signal. Opaque encapsulations are unsuitable for emergency dosimetry as well as chip cards produced using the newest flip-chip technology, which no longer requires any encapsulation [41].

Materials that are widely spread among the population and thus have received high scientific interest in the last years are electronic components found within portable electronic devices, such as mobile phones, portable computers, music and video players, USB flash drives, digital cameras, personal digital assistants and digital watches. Of the electronic components investigated, surface-mount resistors with alumina porcelain substrates seem to be the most promising material, consistently producing an OSL signal following irradiation, with minimum detectable doses on the order of 10 mGy for a typical sample [42]. The dose response of the OSL signal of alumina-rich resistors was found to be linear in the dose range of 10 mGy up to almost 100 Gy. Comparative OSL and TL measurements indicated that the OSL signal originates from the 190 °C TL peak, similar to the well characterized dosimeter material Al2O3:C. To eliminate a thermally unstable low temperature TL peak prior to OSL measurement, a preheat of 10 s at 120 °C was applied and the OSL output was maximized by

measuring at 90 °C. A problematic issue of this material is that the OSL (and correspondingly the 190 °C TL peak) is prone to anomalous fading, with a signal loss of 50% occurring in the first 10 days following irradiation. The fading rate was determined for a specific type of resistor sample but variability for different types of alumina substrates in various electronic devices is conceivable. Trial irradiations of four different brands of mobile phones in the dose range between 0.1 and 0.6 Gy were performed and the dose assessed between 4 and 21 days after irradiation using the same fading rate for signal correction. The fact that the recovered doses were within 150 mGy of the dose recorded by an electronic dosimeter indicates that the systematic error introduced by varying fading rates might be acceptable for emergency response. An indication for this has also been shown in a comparative TL and OSL study of an alumina-rich ceramic resonator in a USB flash drive, for which an only 10% higher fading rate was observed [43]. The OSL decay curve of the ceramic resonator showed a complex shape with a dominating "fast component" but also marked "slow components" and a constant background signal was not reached even after a stimulation time of 900 s. When using an OSL readout time of 150 s for constructing a dose response curve, a small OSL integration interval and early background subtraction had to be applied to avoid apparent sensitivity changes and signal recuperation, which were shown to be an artefact of the data processing procedure. In a trial irradiation of the USB flash drive, a dose of 0.98 Gy could be recovered when using an OSL readout time of 450 s.

Two other types of electronic components found on the circuit board of mobile phones have been studied by OSL for use in accident dosimetry: capacitors and integrated circuits [44]. In comparison to alumina-rich resistors the radiation sensitivity of these components is about two orders of magnitude smaller, with minimum detectable doses < 0.7 Gy for capacitors and integrated circuits. The dose response curve for these components was linear for doses from 0.7 Gy up to 160 Gy. The fading rate for capacitors and integrated circuits (50% after 10 hours) was smaller than for resistors (90% after 10 hours), for measurements at room temperature and without preheat. The ability of capacitors and integrated circuits to recover a known laboratory-delivered dose of 15 Gy has been tested. A single-aliquot regenerative-dose protocol with test dose normalization was used immediately after irradiation. For resistors and capacitors, recovered dose was compatible with applied dose (15.5 \pm 0.1 and 15.2 \pm 0.4 respectively), while an underestimation of 18% was observed for integrated circuits.

Thermoluminescence properties of glass from a large number of display windows of mobile phone and wrist watches have also been studied [45]. For some samples, zero-dose TL signals were observed. They varied in shape and in intensity from a glass to another, which could be a serious issue in retrospective dosimetry. When existing, they were much more intense in watch glasses than in display windows of mobile phones. UV exposure was suggested to explain their origin. Different kinds of radiation induced TL signals were observed, but a glow curve with a main TL peak at around 210 °C was the most common feature found. The dose response curve was linear in the range between 1 and 200 Gy. Concerning fading, the signal loss was around 40% in the first 24 h after which the signal decayed at a much slower rate. The TL signal was only weakly bleached by light, but because of this property, OSL measurements were not considered.

Exemplary dose response curves of components of personal objects are shown in *Figure 1*, the main

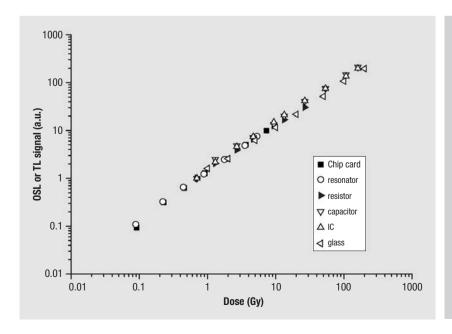


Fig. 1 | Exemplary luminescence dose response curves of UV-cured wire-bond chip cards, an alumina rich resonator from a USB flash drive, alumina-rich resistors, capacitors and integrated circuits (IC) from circuit boards of mobile phones (all OSL) and glass from display windows (TL). Data points are normalized to respective luminescence response at 0.7 Gy (measured or estimated). Data taken from [40, 43-45].

Table 1 | Compilation of the main characteristics of all materials usable for retrospective and/or accident dosimetry, using luminescence techniques. The fading rates of capacitors and integrated circuits are reported here for measurements at room temperature without preheat. Tooth enamel and dental ceramics are not listed in this table, as only an (not yet existing) in vivo application would be useful for these materials

Materials	Zero dose signal	Dose response	Fading	Minimum detectable dose (mGy)	Type of dose	Ubiquity	Processing time
Bricks, tiles, porcelain	No	Linear up to several Gy	No	25	local	high	Days to weeks
Cement, mortar	No	Linear up to several Gy	No	> 100	local	high	Days to weeks
Chemicals	< 10 mGy	region of linearity dependent on material	Negligible for most materials	1-20	local	moderate	< 1 h
chip cards with translucent encapsulation	Yes (TL) No (OSL)	Linear up to 7 Gy	70-80% in 10 days	3-20	individual	low	< 1 h
Alumina rich resistors	No	Linear up to 90 Gy	50% in 10 days	< 10	individual	high	< 1 h
Capacitors	No	Linear up to 160 Gy	50% after 10 h	< 700	individual	moderate	< 1 h
Integrated circuits	No	Linear up to 160 Gy	50% after 10 h	< 700	individual	high	< 1 h
Glass in monitor displays	Yes/No	Linear up to 200 Gy	40% in 1 day, slow decay after	< 1000	individual	high	< 1 h
Watch glasses	Yes/No	Linear up to 200 Gy	40% in 1 day, slow decay after	< 1000	individual	high	< 1 h

characteristics of all materials reviewed in this paper are summarized in *Table 1*.

APPLICATION FOR POPULATION TRIAGE

Effective triage requires the availability of methods to assess absorbed dose rapidly for a large number of possible casualties, and to identify individuals who have minimal or no exposure. In addition to triage capacity based on biological dosimetry, physical dosimetry techniques are nowadays also considered for this specific application [46].

It is worth noting that, compared to EPR [31], luminescence techniques were rarely used for dose estimation to individuals in the case of an acute radiation accident. Materials such as cement, mortar, ceramics or bricks need complex and long investigation to reconstruct the dose in the materials and to relate it to the dose to the victims. Additional calculations or simulations with Monte Carlo methods and a very good knowledge of the circumstances and scenario of the radiation accident are needed. When considering that most of accidental exposures are localized or highly heterogeneous, one can easily understand the difficulties in converting the dose measured in the materials to the dose distribution of the victim's organism. This also applies to chemical products, such as common salt or washing powder, even though they are easier to collect, prepare and measure.

Recent investigations on new types of materials such as chips cards, electronic components in and glass of display windows from portable electronic devices make luminescence dosimetry more attractive for radiation accident and triage applications. As those objects are usually worn close to the body (e.g. in pockets or handbags), the dose measured in the materials is obviously more pertinent and can be related to the victims.

For triage application, the characteristics of these new materials should meet specific criteria, which are not mandatory for common radiation accident situations.

- Materials should be available and easily collectable on most of the potential victims. Chip cards that show a radiation induced signal (i.e. with a translucent encapsulation) have a relatively low occurrence, whereas portable electronic devices such as mobile phones or MP3 players are very popular and can thus be found on a large percentage of the population. From the reported studies, it seems that radiation sensitive electronic components (resistors, capacitors, integrated circuits) can be found in most of the modern electronic personal devices. Display windows of these devices are usually made with two slices of glass. Nevertheless, as the technology is changing very fast in this field (miniaturization of components and change in substrates used, new types of screen for which plastic is often preferred) permanent control of technical advances is needed.

- The presence of zero dose (background) signals can impede the identification and quantification of the luminescence signals from ionizing radiation and increase the minimum detectable dose. Electronic components investigated by OSL do not show any zero dose signals, while glasses measured by TL present very intense background signals, with varying shapes of glow curves.
- The requirement for the minimum detectable dose for population triage is about 1 Gy, which is fulfilled by all materials investigated by luminescence (*Table 1*).
- Signal stability is a critical point for dose evaluation. When the radiation-induced signals decay with time after irradiation (fading), several difficulties may arise. Time span between the accidental exposure and the measurement can range from days to weeks, in the worst case. Decay rates should then be sufficiently slow in order not to raise the minimum detectable dose significantly above the threshold value of 1 Gy. The decay rate should not vary significantly for the same type of component in order to apply a common correction factor. Considering the reported fading rates of the OSL signal of electronic components, dose determination within the mentioned delay seems a priori possible. Nevertheless, further studies on the variability of fading rates for the different types of components and among the same type of component are needed. The last difficulty lies in the fact that, in order to be able to correct the measured dose, the time of the exposure has to be known, which is not always the case. By measuring different types of materials with different fading rates, it should theoretically be possible to estimate the time elapsed since irradiation and thus to correct the measured dose for fading.
- For an efficient triage, the applied methodologies (including sample preparation, measurement and data analysis) should be rapid and with high instrumental capacity in terms of analyzable samples. For glasses and electronic components, the sample preparation step is simple and can be done in little time [42, 44, 45]. A single luminescence measurement takes less than 5 minutes for one sample. The need for individual calibration of the radiation sensitivity and for possible changes in sensitivity implies that at least two measurements may be needed per sample, which would still result

in a comparatively short measurement time span. Moreover, practically all modern research luminescence readers present the advantage of being automated.

Regarding all of the criteria mentioned above and the available data on the different materials investigated by luminescence techniques, OSL measurements on electronic components seem to be the most promising method for population triage.

CONCLUSIONS AND OUTLOOK

A number of materials have been reported on in this paper that show useful luminescence properties to be potentially used for population triage. Unfortunately the only materials not showing any signal fading are either building materials or chemicals, which however only allow the estimation of a local dose and for some also require lengthy sample preparation techniques, making them unsuitable for population triage. Personal objects, carried close to the human body, seem to be much more promising for such an application, however all components studied within such objects have the common feature of showing more or less pronounced signal fading with storage time. This implies that either the time and duration of exposure of the individuals has to be known with reasonable accuracy, or that at least two suitable components with different fading rates can be found on an individual and a suitable methodology can be developed to infer the dose, independent of an a-priori knowledge of time and duration of exposure. Ubiquity of the materials is an essential prerequisite, making chip cards not one of the prime candidates but electronic components and glass found within portable electronic devices. So far all laboratories involved in luminescence accident dosimetry use their own developed and preferred measurement protocols and a harmonization of the measurement procedure and the development of a unified approach to uncertainty estimation is a desirable future activity for quality assurance. Clearly more investigations into the luminescence properties are necessary, in particular the variability of fading rates to establish personal objects as a reliable fortuitous dosimeter for emergency response.

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References

- Jaworska A. Types of mass casualties and their management. Ann Ist Super Sanità 2009;45(3):246-50.
- Haskell EH. Retrospective accident dosimetry using environmental materials: the role of thermoluminescence. *Nucl Tracks Radiat Meas* 1993;21:87-93.
- Bailiff IK. Retrospective dosimetry with ceramics. Radiat Meas 1997:27:923-41.
- 4. Göksu HY, Bailiff IK. Luminescence dosimetry using building ma-
- terials and personal objects. Radiat Prot Dosim 2006; 119:413-20.
- International Commission on Radiation Units and Measurements. Retrospective assessment of exposures to ionising radiations. ICRU; 2002. (Report 68). J ICRU 2(2), p. 144.
- McKeever SWS. Thermoluminescence of solids. Cambridge: Cambridge University Press; 1985.
- Bulur E. An alternative technique for optically stimulated luminescence (OSL) experiment. *Radiat Meas* 1996;26:701-9.

- Thomsen KJ, Bøtter-Jensen L, Jain M, Denbya PM, Murray AS. Recent instrumental developments for trapped electron dosimetry. *Radiat Meas* 2008;43:414-21.
- Bøtter-Jensen L, Bulur E, Duller GAT, Murray AS. Advances in luminescence instrument systems Radiat Meas 2000;32:523-8.
- Wintle AG, Murray AS. Quartz OSL: Effects of thermal treatment and their relevance to laboratory dating procedures. *Radiat Meas* 2000;32:387-400.
- Higashimura T, Ichikawa Y, Sidei T. Dosimetry of atomic bomb radiation in Hiroshima by thermoluminescence of roof tiles. *Science* 1963;139:1284-5.
- 12. Roesch WC. US Japan joint reassessment of atomic bomb radiation dosimetry in Hiroshima and Nagasaki, final report. DS86 Dosimetry System 1986. In: Roesch WC (Ed.). Radiation measurements Vols 1 and 2. Hiroshima, Japan: The Radiation Effects Research Foundation; 1987.
- Haskell EH, Bailiff IK, Kenner GH, Kaipa PL, Wrenn ME. Thermoluminescence measurements of gamma-ray doses attributable to fallout from the Nevada Test Site using building bricks as natural dosemeters. *Health Phys* 1994;66:380-91.
- 14. Bailiff IK, Stepanenko VF, Göksu HY, Bøtter-Jensen L, Brodski L, Chumak V, Correcher V, Delgado A, Golikov V, Jungner H, Khamidova LG, Kolizshenkov TV, Likhtarev I, Meckbach R, Petrov SA, Sholom S. Comparison of retrospective luminescence dosimetry with computational modelling in two highly contaminated settlements downwind of the Chernobyl NPP. Health Phys 2004;86:25-41.
- 15. Bailiff IK, Stepanenko VF, Göksu HY, Jungner H, Balmukhanov SB, Balmukhanov TS, Khamidova LG, Kisilev VI, Kolyado IB, Kolizshenkov TV, Shoikhet YN, Tsyb AF. The application of retrospective luminescence dosimetry in areas affected by fallout from the Semipalatinsk nuclear test site: an evaluation of potential. Health Phys 2004;87:625-4.
- Göksu HY, Stepanenko, VF, Bailiff IK, Jungner H. Intercomparison of luminescence measurements of bricks from Dolon' Village: Experimental methodology and results of European Study Group. J Rad Res 2006;47:(Suppl: A29-A37).
- Bougrov NG, Baturin VA, Göksu HY, Degteva MO, Jacob P. Investigations of thermoluminescence dosimetry in the Techa river floodplain: analysis of the new results. *Radiat Prot Dosim* 2002;101:225-8.
- Göksu HY, Degteva MO, Bougrov NG, Meckbach R, Haskell EH, Bailiff IK, Bøtter-Jensen L, Jungner H, Jacob P. First international intercomparison of luminescence techniques using samples from the Techa river valley. *Health Phys* 2002;82:94-101.
- Meckbach R, Bailiff IK, Göksu HY, Jacob P, Stoneham D. Calculation and measurement of dose-depth distribution in bricks. *Radiat Prot Dosim* 1996;66:183-6.
- Göksu HY, Heide LM, Bougrov NG, Dalheimer AR, Meckbach R, Jacob P. Depth-dose distribution in bricks determined by thermoluminescence and by Monte-Carlo calculation for external γ-dose reconstruction. Appl Radiat Isot 1996;47:433-40.
- Taranenko V, Meckbach R, Degteva MO, Bougrov NG, Göksu Y, Vorobiova MI, Jacob P. Verification of external exposure assessment for the upper Techa riverside with luminescence measurements and Monte Carlo photon transport modeling. *Radiat Environ Biophys* 2003;42:17-26.
- Jacob P, Göksu HY, Taranenko V, Meckbach R, Bougrov NG, Degteva MO, Vorobiova MI. On an evaluation of external values in the Techa River Dosimetry System (TRDS) 2000. Radiat Environ Biophys 2003;42:169-74.

- Bøtter-Jensen L, Murray AS. Optically Stimulated Luminescence in retrospective dosimetry. *Radiat Prot Dosim* 2002;101:309-14.
- 24. Ramzaev V, Bøtter-Jensen L, Thomsen KJ, Andersson KG and Murray AS. An assessment of cumulative external doses from Chernobyl fallout for a forested area in Russia using the optically stimulated luminescence form quartz inclusions in bricks. J Environ Radioact 2008;99:1154-64.
- Thomson KJ, Bøtter-Jensen L, Murray AS, Solongo S. Retrospective dosimetry using unheated quartz: a feasibility study. *Radiat Prot Dosim* 2002;101:345-8.
- Jain M, Bøtter-Jensen L, Murray As, Jungner H. Retrospective dosimetry: dose evaluation using unheated and heated quartz from a radioactive waste storage building. Radiat Prot Dosim 2002;101:525-30.
- Thomson KJ, Jain M, Bøtter-Jensen L, Murray AS, Jungner H. Variation with depth of dose distributions in single grains of quartz extracted from an irradiated concrete block. *Radiat Meas* 2003;37:315-21.
- 28. Göksu HY, Bailiff IK, Mikhailik VB. New approaches to retrospective dosimtery using cementitous building materials. *Radiat Meas* 2003;37:323-7.
- Thomson KJ, Bøtter-Jensen L, Murray AS. Household and workplace chemicals as retrospective luminescence dosemeters. *Radiat Prot Dosim* 2002;101:515-8.
- Bernhardsson C, Christiansson M, Mattson S, Rääf C. Household salt as a retrospective dosemeter using optically stimulated luminescence. *Radiat Environ Biophys* 2009; 48:21-8.
- 31. Trompier F, Bassinet C, Wieser A, De Angelis C, Viscomi D, Fattibene P. Radiation-induced signals analysed by EPR spectrometry applied to fortuitous dosimetry. *Ann Ist Sup Sanità* 2009;45(3)287-96.
- 32. Godfrey-Smith DI, Pass B. Detection of gamma radiation absorbed by human tooth enamel using optically stimulated luminescence. *Health Phys* 1997;72:1-5.
- 33. Godrey-Smith DI. Toward *in vivo* OSL dosimetry of human tooth enamel. *Radiat Meas* 2008;43:854-8.
- Bailiff IK, Correcher V, Delgado A, Göksu Y, Hübner S. Luminescence characteristics of dental ceramics for retrospective dosimetry: a preliminary study. *Radiat Prot Dosim* 2002;101:519-24.
- Göksu HY. Telephone chip cards as individual dosimeters. Radiat Meas 2003:37:617-20.
- Mathur VK, Barkyoumb JH, Yukihara EG, Göksu HY. Radiation sensitivity of memory chip module of an ID card. *Radiat Meas* 2007;42:43-8.
- Barkyoumb JH, Mathur VK. Epoxy encapsulant as serendipitous dosimeters during radiological/nuclear events. *Radiat Meas* 2008;43:841-4.
- Göksu HY, Jacob P. Dosimeter with phosphor substance in the cover layer of an electronic chip card. International and European Patent Applications WO/2006/108571 2006, Publication no: EP1712932A1.
- Göksu HY, Spöttl T. Improving the radiation sensitivity of chipcards. In: 10th international conference on defects in insulating materials, conference program and book of abstracts. July 10-14, University of Milano-Bicocca, Milano: Aracne editrice S.r.l.; 2006. p. 159.
- Woda C, Spöttl T. On the use of OSL of wire-bond chip card modules for retrospective and accident dosimetry. *Radiat Meas* (in press.) doi:10.1016/j.radmeas.2009.02.016.
- Göksu HY, Wieser A, Ulanovsky A. Retrospective individual dosimetry using luminescence and EPR after radiation accidents. BMU 2007-696 Reihe Umwelt udn Politik ISSN 1612-6386. Bundesministerium für Umwelt, Naturschutz und Reaktorischerheit: 2007.

- 42. Inrig EL, Godfrey-Smith DI, Khanna S. Optically stimulated luminescence of electronic components for forensic, retrospective and accident dosimetry. *Radiat Meas* 2008;43:726-30.
- 43. Beerten K, Woda C, Vanhavere F. Thermoluminescence dosimetry of electronic components from personal objects. *Radiat Meas*; (in press.). doi:10.1016/j.radmeas.2009.02.003.
- 44. Bassinet C, Trompier F, Clairand I. Radiation accident dosimetry on electronic components by OSL. *Health Phys* (accepted for publication).
- Bassinet C, Trompier F, Clairand I. Radiation accident dosimetry on glass by TL and EPR spectrometry. *Health Phys* (accepted fo publication).
- 46. Alexander GA, Swartz H M, Amundson S A, Blakely W F, Buddemeier B, Gallez B, Dainiak N, Goans R E, Hayes R B, Lowry P C, Noska M A, Okunieff P, Salner A L, Schauer DA, Trompier F, Turteltaub K W, Voisin P, Wiley Jr A L and Wilkins R. BiodosEPR-2006 meeting: Acute dosimetry consensus committee recommendations on biodosimetry applications in events involving uses of radiation by terrorists and radiation accidents. *Radiat Meas* 2007;42:948-71.