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RESEARCH ARTICLE

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Overview of physical dosimetry methods for triage application integrated in the new European network RENEb

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ABSTRACT

Purpose: In the EC-funded project RENEb (Realizing the European Network in Biodosimetry), physical methods applied to fortuitous dosimetric materials are used to complement biological dosimetry, to increase dose assessment capacity for large-scale radiation/nuclear accidents. This paper describes the work performed to implement Optically Stimulated Luminescence (OSL) and Electron Paramagnetic Resonance (EPR) dosimetry techniques.

Materials and methods: OSL is applied to electronic components and EPR to touch-screen glass from mobile phones. To implement these new approaches, several blind tests and inter-laboratory comparisons (ILC) were organized for each assay.

Results: OSL systems have shown good performances. EPR systems also show good performance in controlled conditions, but ILC have also demonstrated that post-irradiation exposure to sunlight increases the complexity of the EPR signal analysis.

Conclusions: Physically-based dosimetry techniques present high capacity, new possibilities for accident dosimetry, especially in the case of large-scale events. Some of the techniques applied can be considered as operational (e.g. OSL on Surface Mounting Devices [SMD]) and provide a large increase of measurement capacity for existing networks. Other techniques and devices currently undergoing validation or development in Europe could lead to considerable increases in the capacity of the RENEb accident dosimetry network.

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Introduction

In the event of a large-scale radiological emergency, triage of victims according to their degree of exposure forms an important initial step. Retrospective/accident dosimetry techniques are considered by many institutions as essential tools in the management of radiological mass casualties, and can provide timely assessments of exposures to the general population (Blakely et al. 2005; Alexander et al. 2007; Dainiak et al. 2007). Retrospective dosimetry techniques commonly considered for this purpose are mainly based on biological assays and on cytogenetic approaches (Wilkins et al. 2008; Wojcik et al. 2010; Ainsbury et al. 2011, 2014; Di Giorgio et al. 2011; Romm et al. 2011, 2013; Sullivan et al. 2013). In addition to these 'gold standard approaches', the development of new approaches is also being considered. For example, Electron Paramagnetic Resonance (EPR) spectroscopy on nails (in vivo and ex vivo) and teeth (in vivo) was recently investigated in the context of NIH National Institute of Allergy and Infectious Disease (NIH/NIAD) and Biological Advanced Research and Development Authority (BARDA)

programs (Satyamitra et al. 2015), with the aim of developing field deployable EPR spectrometers for in situ dose evaluation (e.g. Williams et al. 2011, 2014; He et al. 2014). In Europe, a laboratory network approach was taken, which was founded on the biological dosimetry techniques already available in cytogenetic laboratories. Alternative techniques to biological dosimetry were also considered in EC programs: Multibiodose (Jaworska et al. 2015), RENEb (Kulka et al. 2012, 2015) and Booster (Robbe et al. 2014). In the Multibiodose and RENEb projects, luminescence and EPR analyses were implemented, respectively, on electronic components (resistors) and mineral glass found in mobile phones, though they were not considered as established techniques. Thermoluminescence (TL) on resistors was also considered in the course of the Booster project (Mesterházy et al. 2012). In RENEb, TL on resistors and on Liquid Crystal Display (LCD) glass were identified as possible new approaches to be implemented in the RENEb follow-up: RENEb-plus.

In recent decades, EPR dosimetry on mineral glass was used several times for the estimation of dose to persons who

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had been accidentally overexposed. Mineral glass from watches was used by Wu et al. (1998). More recently, the potential to use the mineral glass from LCD screens found in electronic devices such as mobile phones was explored by Trompier et al. (2011). Luminescence (TL and OSL) analyses of the dosimetric properties of different types of electronic components, such as resistors, inductors, and capacitors have been reported by different groups (Inrig et al. 2008; Beerten and Vanhavere 2008, 2010; Bassinet et al. 2010; Woda et al. 2010; Beerten and Vanhavere 2011; Fiedler and Woda 2011; Trompier et al. 2011; Pascu et al. 2013; Lee et al. 2015), and proposed as new tools for accident dosimetry. Among these Surface Mounted Devices (SMD), resistors are the most studied type, and OSL the technique the most used (e.g. Ekendahl and Judas 2012; Bassinet et al. 2014; Smith et al. 2015; Eakins et al. 2016).

Two main factors have guided these EC projects to develop and implement EPR, TL and OSL techniques. First, whereas biological assays provide a whole body dose estimate, EPR/TL/OSL on mobile phone components give localized dose information. By combining localized and whole body dose data, it is possible to derive additional information that can help evaluate both the heterogeneity of the dose delivered and the time delay since exposure. Combination of results from different assays or methods to evaluate heterogeneity was implemented in the Multibiodose software (Ainsbury et al. 2014; Jaworska et al. 2015). The time delay can be estimated based on differences in results from EPR and OSL, since the OSL signal presents significant fading following irradiation, while the EPR signal is stable. Second, the measurement capacity of a network could be greatly increased by including laboratories able to implement these techniques. Multibiodose included three institutions for the development of EPR/OSL methods: protocols for sample preparation, measurements and data analysis were developed and evaluated through internal blind tests. At the end of the project, training followed by an ILC was conducted with members of Working Group 10 'Retrospective Dosimetry' of the European Radiation Dosimetry Group (EURADOS). These include most European laboratories with interests in retrospective dosimetry (13 laboratories for EPR and 12 for OSL), which already constitutes a research network. The exercises were designed to evaluate the Multibiodose protocols and the ability of the laboratories to apply the Multibiodose methodology. The results of the ILC were considered as very satisfactory for OSL (Bassinnet et al. 2014). For EPR on glass, the ILC has brought to light some difficulties in signal analysis, due to an unexpected effect of UV light. Nevertheless, it was demonstrated that an operational network could be constituted with a minimum of training, and that accurate dose estimation could be provided once the identified analytical issues are resolved (Fattibene et al. 2014).

The materials used for OSL and EPR are not biological, and the techniques are not yet considered as fully established, but they were shown to provide important information to complement biological dosimetry analyses. Given this, the results obtained in Multibiodose were considered sufficiently satisfactory that these two new approaches were

implemented in the RENE project, which aimed to establish an operational and sustainable network. RENE included three laboratories for EPR dosimetry and five for OSL dosimetry. The work performed during RENE consisted of training and ILC programs that have led to the improvement of the OSL and EPR protocols. Additional materials and techniques were investigated later in the RENE project, for example TL on LCD glass and on SMD, through an ILC co-organized by Working Group 10 of EURADOS. Preliminary results of these later ILC are presented in Ainsbury et al. (2016). The present paper aims to give an overview of the work achieved within the RENE project that focussed on EPR of glass and OSL of SMD, to discuss the lessons learnt and describe the advantages of including these physical dosimetry approaches in a biodosimetry network.

Materials and methods

OSL

OSL dosimetry principles

Luminescence signals used in dosimetry consist of light emitted under stimulation by a material able to store energy from radiation. Such materials include insulators and semiconductors. TL and OSL are linked to the presence of defects in the structure of the material under consideration. Irradiation induces free charge carriers (electrons and holes). Defects form spatially localized potentials in the energetically forbidden zone between valence and conduction bands, in which free charge carriers may become trapped. If the difference in energy between defect ground state and conduction or valence band is sufficiently high, the charge carriers remain (metastably) trapped at room temperature, and thus charges will be accumulated within these types of defect during irradiation. Under stimulation, transfer of sufficient energy to the electron allows it to escape from the trap, migrate in the conduction band, and recombine with trapped holes. Some hole traps act as luminescence centers, by emitting light as they de-excite following recombination. In the case of OSL, the stimulation is with light. 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 detected using a photomultiplier tube. An example of OSL signals recorded as a function of the stimulation time for irradiated resistors is given in Figure 1. Generally, the stimulation is carried out with a continuous excitation light source, such as a continuous wave laser. The sample is illuminated and the light yield recorded simultaneously, in different wavelength bands, over a time period of many seconds.

In the simplest case of one electron trap, one hole trap (recombination center), and doses sufficiently low that only a small proportion of the electron traps are filled, the luminescence signal is proportional to the amount of recombined trapped charges and thus directly proportional to the dose the material has received. In this case the dose absorbed during an accidental exposure may be determined by comparing the quantity of light emitted from the sample during stimulation, with that obtained following a known dose of ionizing radiation. If the reading of sample is not performed

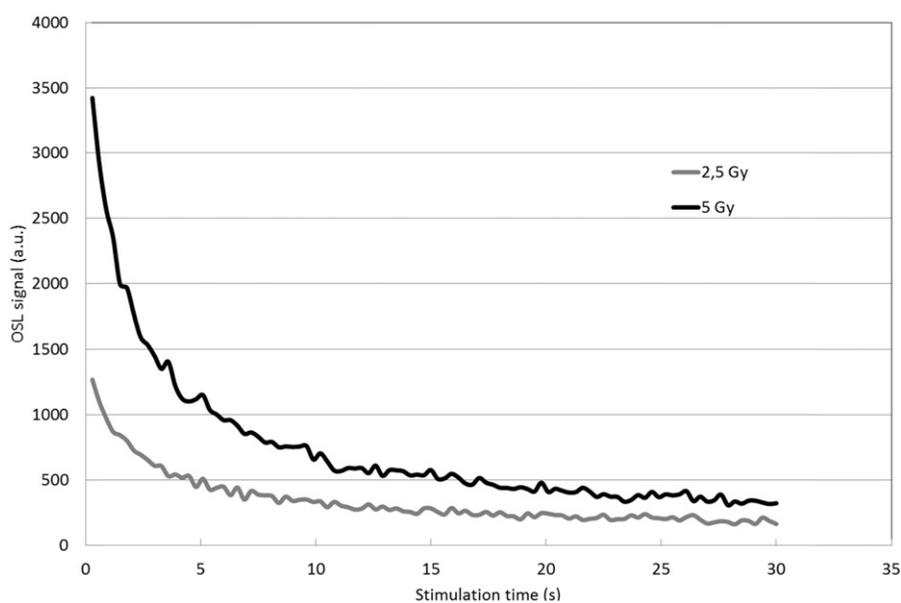


Figure 1. Example of Optically Stimulated Luminescence (OSL) curve for resistors irradiated respectively at 2.5 and 5 Gy.

immediately after irradiation, the quantity of trapped charges may decrease with time, even at room temperature. At a given temperature there is a finite probability that charge carriers (electrons and holes) will be thermally excited from their trapped states to the conduction/valence band, and the intensity of this phenomenon increases with temperature. This effect is termed thermal fading. For some materials, the luminescence signal decay rate can be different from the decay expected from simple thermodynamic considerations. This phenomenon is called anomalous fading. The origin of anomalous fading has been explained by tunnelling of charge carriers from the trap to the recombination centre. To avoid systematic underestimation of the absorbed dose, a correction factor is applied to the estimated dose to take into account the fading effect (of whatever origin). From a practical point of view, for accident dosimetry, it is therefore necessary to know the time elapsed between the accidental radiation exposure and the readout of the materials. The accuracy of dose estimation could therefore be affected by the uncertainty on the delay time.

OSL dosimetry on resistors

Resistors are composed of thin metal resistive elements mounted on alumina porcelain substrates. This kind of material is known to present adequate luminescent and dosimetric properties for accident dosimetry. The OSL signal of resistors was found to fade significantly in the days following irradiation, in a manner approximately proportional to $1/\log$ time (Bassiniet et al. 2014). After a delay of 10 days, the signal is reduced by a factor of two. The minimal detectable dose (MDD) is thus dependent on the time elapsed after irradiation. The predicted MDD at 10 days is estimated at 120 mGy, and at 300 mGy after 2 months. As noted above, uncertainty on the time between accidental irradiation and sample readout is important in evaluating the uncertainty on the dose estimate. When the date of the irradiation is not known, as for some insidious scenarios of exposure, the approach

remains valid for the lowest dose (<200 mGy) used in the dose categorization applied for triage (Ainsbury et al. 2014). In the case of high doses, where clinical signs would be the main means of identification of irradiated persons, accurate dose estimation is required at least in a second step to define the best approach for medical management. Depending on how well the date of exposure could be constrained, this scenario would be challenging in a dosimetric analysis based on OSL alone.

Sample preparation for OSL analysis

As the materials considered for OSL are sensitive to light, all the sample collection and preparation must be performed under subdued red light conditions. The necessity to use red light can complicate the disassembling of the phone and, the identification and collection of these small resistors for the inexperienced operator but is generally not found to be a major obstacle for well-trained personnel. It could, however, lead to faster tiring of operators in an actual mass casualty event and thus to the need for faster exchange of available staff. Identification of the resistors is a crucial step in the dose estimation process. Inaccurate dose assessments during the ILC were attributed to errors in the identification of the resistors. For example, resistors classed as medium and small in size have, respectively, the following dimensions: $1 \times 0.5 \times 0.35 \text{ mm}^3$ and $0.6 \times 0.3 \times 0.2 \text{ mm}^3$. Once collected, the resistors should be stored in the dark before measurements.

OSL measurements

In the luminescence readers used by the RENEB (and EURADOS) partners, light stimulation is provided by blue LED ($470 \pm 30 \text{ nm}$). Two measurement protocols were developed within the Multibiodose project:

- A fast mode protocol without preheat of the samples, which makes this protocol better adapted to a large-scale radiological emergency. The OSL signal acquisition is done at room temperature.
- A full mode protocol with preheat (10 s at 120 °C) of the samples to make the signal more stable, which can offer more accurate dose assessment. The OSL signal acquisition is done at 100 °C.

OSL curves analysis

The complete analysis of the OSL curves was performed using an analysis template provided to the participants.

Each participant was asked to insert the following data in the template:

- The initial OSL curve corresponding to the unknown dose;
- The calibration OSL signal curve, corresponding to the calibration dose of 5 Gy;
- The dose conversion factor obtained from the comparison between the OSL signal of the samples irradiated at IRSN and the one obtained from the same samples after re-irradiating them at the same dose using the laboratory's source. To minimize the impact of fading effects, the time delay between the exposure and the signal acquisition was controlled using;
- The exact time (date and the hour) of the initial irradiation;
- The exact time (date and the hour) of the calibration irradiation;
- The instrumental background.

The analysis template first evaluated the net OSL signals corresponding to the unknown dose and to a calibration dose of 5 Gy. Most OSL readers were equipped with an internal beta $^{90}\text{Sr}/^{90}\text{Y}$ source but some participants of the ILC have used an external gamma source (^{137}Cs or ^{60}Co) to deliver the calibration dose. Dose rate in terms of air kerma are usually of the order of few tens of mGy s^{-1} . The signal analysis was done using integration windows of 0–6 s for signal and 6–12 s for background, respectively. Then, the value of the calibration dose, equal to 5 Gy, was corrected using the dose conversion factor. The uncorrected dose was calculated applying the linear proportional relation between the initial net OSL signal, with the ratio between the corrected calibration dose and the calibration signal used as the calibration coefficient. The uncorrected dose obtained was then corrected for fading. The fading correction factors were derived from two fading curves (one for each protocol) built by three partners (HMGU, IRSN and ISS) within the MULTIBIDOSE project. Uncertainties were calculated from the combination of the error estimation due to counting statistics for the instrumental background and the uncertainty resulting from fitting of the fading curves to the experimental data. As an example, in Bassinet et al. (2014) uncertainties range from 25–40% ($k=1$) for doses above 1 Gy and from 30–90% at 0.3 Gy. For the purpose of triage categorization, only the numerical value of the measured dose, without the

uncertainties, was considered to see whether the result falls into the correct category. Uncertainties were nonetheless taken into account in the evaluation of the dose assessment capabilities of the method.

Dose estimation method and calibration

After the read out of the OSL signal of the resistors, they were irradiated with a dose of 5 Gy and read out again. The sensitivity to dose is therefore assessed for each cup of resistors. The irradiation is usually performed by an internal calibration source installed in the reader. The type of source used by most of laboratories was a $^{90}\text{Sr}/^{90}\text{Y}$ beta source. It has been verified that doses delivered by gamma irradiation with a ^{60}Co source calibrated in units of air kerma, generated identical OSL signal intensity in resistors to an identical dose from the internal beta source: the OSL readers of some laboratories did not contain a beta source. In some other cases the calibration was done using a MV X-ray beam or ^{60}Co sources calibrated in air kerma. Air kerma calibration with photons above 1 MeV is almost equivalent to a calibration in alumina kerma (National Institute of Standards and Technology [NIST] 2016). At 1.25 MeV, the ratio of mass energy absorption coefficients between air and alumina is 1.02.

EPR spectroscopy

EPR dosimetry principles

EPR dosimetry is based on the quantification by EPR spectroscopy of dose-dependent changes in the concentrations of free radicals, defects or any species with paramagnetic properties that is formed in a given material under exposure to ionizing radiation. These include, but are not limited to, electrons and holes trapped in states similar to those that participate in TL and OSL. At the dose levels of interest for evaluation of human exposures, the quantity of radio-induced species is approximately proportional to the dose absorbed in the considered materials, but use of a calibration curve is required for improved accuracy. Most of the EPR dosimetry laboratories perform the measurements using continuous wave spectroscopy and instrumentation working in X-band. The recording parameters selected for the measurements have to be optimized according of the type of resonant cavity and the type of samples measured. Therefore, each laboratory has its own set of recording parameters defined according its own criteria (International Organization for Standardization [ISO] 2013).

EPR dosimetry on glass

Mineral glasses have been investigated for different types of application in dosimetry for many years, including for accident dosimetry. Glass found in wrist watches (Wu et al. 1995; Marralle et al. 2011) and eye glasses (Bassinnet et al. 2010) have been investigated, but the use of mineral glasses in the fabrication of these kinds of objects is declining. However, in personal electronic devices such as mobile phones, mineral

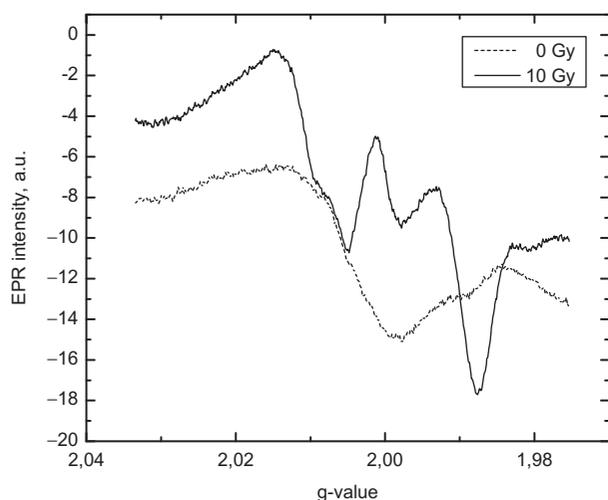


Figure 2. Electron Paramagnetic Resonance (EPR) spectra of unexposed and 10 Gy exposed Gorilla Glass[®] samples.

glasses are now widely used. Several types of glass can be encountered in mobile phones. The glasses used in the Liquid Crystal Display (LCD) were initially considered in Multibiodose. In LCD, various types of glass are used depending on LCD technology. Up to six families of glass were identified (Trompier et al. 2012). Only one of the types of glass does not present dose-dependent changes in EPR signals. Specific procedures have to be developed for the sample preparation, measurement and signal analysis of each type of glass, which makes the management of large number of samples more difficult. It was observed that glass used in touch screens does not present such variability. Moreover, touch screens can be dismantled and replaced in a much easier way than LCD screens, and smartphones are now widely used. For these reasons, it was decided to consider only touch screen glass in RENEb. Touch screens are made of alkali-alumino silicate glass: the radiation-induced EPR signal from this type of glass lies in a spectral region that also contains non-radiation-induced signals (Figure 2): separating these signal components adds complexity and limits precision in the analysis of spectra, especially at low dose levels (<1 Gy).

The natures of the non-radiation-induced signal, of the radio-induced defects present in this type of glass, and of the parameters influencing signal stability are not yet fully understood. As for sodalime and boro-silicate glass, radiation-induced species with long thermal lifetimes are also present in alkali-alumino silicate glass, which is one of the main advantages of this approach. In controlled laboratory conditions, the minimum dose level for detection has been estimated at 0.75 Gy (Fattibene et al. 2014). UV light induces species with signatures similar to those that are radiation-induced, and also stimulates chemical reactions that lead to the decay of some of the radio-induced defects, including recombination of electrons and holes (Fattibene et al. 2014).

Sample preparation for EPR analysis

The sample preparation aims to remove all materials that could produce parasitic signals, and to give the sample material a form that can be measured using EPR

spectroscopy. The part of the touch screen glass that is analyzed is the external slice of glass, which has to be separated from the other slices of the touch screen. It is important to remove all paint, glue, and other materials that could be applied on the surface of the glass sheet, to eliminate all possible parasitic signals. The cleaning can be performed with ethanol or acetone. The use of an ultrasound bath is not recommended unless it is established that the frequency, power and duration of the bath has no influence on the signal of interest. The glass can be cut or crushed to obtain grains or fragments that would fit in the EPR tubes. The use of pincers to break off fragments does not mechanically induce signals. Whatever the method used, it should be checked that it does not induce an additional signal component. One of the advantages of using touch screens is that the sample preparation can be made quickly.

EPR spectra measurement

The measurements are performed in X-band and in continuous wave mode. For the recording of the spectra, it is recommended to use very high microwave-power to achieve saturation of the signal of interest. For example, microwave power greater than 20 mW is appropriate with a high Q resonator. Recording parameters that have an influence on the signal to noise ratio, on the reproducibility of the measurements, and on the measurement time, need to be optimized by adjustment of at minimum the microwave power, modulation amplitude, conversion time, time constant, number of accumulation, and sweep width. The number of repeated measurements to be made depends on the best compromise between accuracy and rapidness to address the dosimetric question.

EPR spectra analysis

Simple EPR spectra analysis, as done, for example, for alanine dosimetry, is not sufficiently robust for fortuitous dosimeters. In laboratory conditions, with homogenized samples, it gives very good results, but with samples of different origins that were exposed to sunlight, the dose estimation was much less successful, due to variability in response to dose of signals from the different samples, as well as additional effects from UV. On the basis of the results of the ILC, it is expected that a more sophisticated analysis could provide robust dose estimation whatever the history of the glass. Such analysis could be based on the fitting of the different signal components to a measured spectrum to extract the characteristic of the signal used for dosimetry, as is done for tooth enamel dosimetry (Wieser et al. 2006; Ivannikov et al. 2007; Fattibene et al. 2011). Therefore, to develop such analysis, it is necessary to have a very good description of the different signal components. The EPR-DOSIMETRY software (Koshta et al. 2000) was used to evaluate the performance of this approach.

Dose estimation method and calibration

Whichever method is selected for dose estimation, it needs to be validated through an ILC. For a large number of

samples that must be evaluated rapidly, use of common or shared calibration curve(s) to estimate the dose is recommended. The calibration curve should take into account the variability among samples to be analysed. Pre-establishment of curves for the main types of touch screens is an option for rapid assessment. Conversely, for precise dosimetry with a small number of samples, it is preferable to use a calibration curve established using glass of the same origin as the samples to be evaluated.

Although the radiation-induced EPR signals from touch screen glass are considered as stable over a period of weeks, the delay between calibration irradiation and EPR measurement should be made as similar as possible to the delay between accidental exposure and EPR measurement. It is the responsibility of each laboratory to select the most appropriate calibration curve for their analyses.

Calibration coefficients, from signal intensity to absorbed dose, are usually established using calibration curves obtained from the measurement of samples irradiated with known doses in reference conditions in a calibrated facility. One important parameter is the choice of the beam type used for calibration. In accident dosimetry, the practice is to report the absorbed dose in the measured materials using ISO 13304-1:2013 (ISO 2013). For example, for retrospective dosimetry based on tooth enamel, EPR signal intensity is converted into absorbed dose in enamel.

Reference facilities are calibrated in air kerma or absorbed dose in water, so conversion coefficients must be applied to convert the reference quantities into the absorbed dose in the considered medium. It is recommended to use photon beams with energies above 1 MeV, because in this energy range the conversion coefficients are not significantly affected by the atomic number, and therefore the difference in terms of composition between real samples and composition used to calculate mass energy absorption coefficient is minimized. As an example, the calibration in terms of absorbed dose in enamel is commonly done by irradiating samples with Co-60 gamma-rays in terms of air kerma. In these conditions, with photon of mean energy of 1.25 MeV, the ratio between mass energy absorption coefficients of air and enamel is less than 1%. For glass, a similar approach is proposed. As an example, the difference between mass energy coefficients of air and alkali alumina silicate glass is 0.9% (NIST 2016).

Work performed and lessons learnt

OSL dosimetry on resistors

For OSL on SMD, the work achieved during RENEB has mainly consisted of an ILC program to evaluate and improve the protocols developed during the Multibiodose project (Wojcik et al. 2014). In Multibiodose, two protocols were developed: one simpler and faster but supposedly less accurate, the other with additional measurement steps designed to isolate signals with greater thermal stability and better defined fading characteristics. During the ILC, the 'fast mode' and the 'full mode' protocols were evaluated and compared. The first ILC was jointly organized with the Multibiodose project and EURADOS. The overall results of this joint ILC can be

found in Bassinet et al. (2014). Blind doses delivered to cell phones were 0.3, 1.7 and 3.3 Gy. The doses fall within three triage dose ranges: <1 Gy (low dose), 1–2 Gy (medium/intermediate dose) and >2 Gy (high dose). Irradiations were performed in term of air kerma with Co-60 gamma rays. With both protocols and for all dose ranges the mean of the doses measured by the labs fell in the correct range and correctly estimated the nominal dose within 1 standard deviation. Considering fast- and full-mode separately, there were slightly more correct categorizations in the case of the pre-heated samples (91%) compared to non-preheated ones (88%), but it was not significant. From this ILC, it can therefore not be concluded that the full -mode protocol with pre-heat may show a small improvement in terms of accuracy compared to the fast mode protocol. The biggest difficulties encountered by participants came from possible misidentifications of resistors on the circuit board. Spending more time on a training process, possibly involving more people from each lab, may help to solve this problem.

The second RENEB ILC aimed to test the OSL protocols in a more realistic configuration of irradiation. The irradiations were performed during an exercise organized within the FP7 security research project CATO. Within CATO, a field experiment based on real accident scenario was carried out, to evaluate different dose reconstruction techniques. The scenario and the irradiation configurations were based on the accident that occurred in 2002 in Cochabamba, Bolivia. A total of 55 passengers were exposed to ionizing radiation due to a malfunction of an industrial Ir-192 γ -radiography source, which was transported in a passenger bus (International Atomic Energy Agency [IAEA] 2004). Description of the exercise and the overall results of this ILC will be published in a future publication.

EPR dosimetry on glass from touch screen of mobile phone

The first RENEB ILC on touch screen glass was actually organized jointly with EURADOS and Multibiodose. Samples used were from a same batch of touch screens made from Gorilla Glass®. A two days training was provided to the 13 participants. Samples irradiated at known doses (0, 0.8, 2, 4 and 10 Gy) in terms of air kerma using Co-60 gamma rays to establish a calibration curve were distributed with samples irradiated at unknown doses (to be evaluated by participants). The blind doses were 0, 0.9, 1.3 and 3.3 Gy. The overview of the results and a detailed analysis can be found in Fattibene et al. (2014). Two groups of samples were distributed: (A) samples taken from a homogenized mixture of glass pieces collected from five different sheets of glass; and (B) samples from nine different sheets of glass irradiated at the different doses for blind and calibration samples. The blind doses of samples of group A was evaluated with a difference to the nominal dose lower than 5% for the highest blind doses (1.3 and 3.3 Gy) and lower than 10% for the lowest blind dose (0.9 Gy). Therefore, with optimum conditions, this first set of results has shown that EPR on touch screen glass could be very accurate and relatively sensitive, with a MDD estimated at 0.75 Gy. For

Table 1. Comparison of the whole capacity of RENEb for OSL, EPR and dicentric assay (Brzozowska et al. 2016).

	OSL	EPR	Dic Assay manual mode	Dic Assay automatic mode
Number of lab	5	3	21	8
Average capacity per week	500 ± 100	770 ± 60	510 ± 70	400 ± 70
Average capacity per week and per lab	100	257	24	50

RENEb: Realizing the European Network in Biodosimetry; OSL: Optically Stimulated Luminescence; EPR: Electron Paramagnetic Resonance.

samples from group B, the results were less satisfactory mainly due to the variability of the response of the different glass sheets. The scattering of the calibration data points was large and the calibration curve best fits were affected by large errors. The maximum variation was observed with the calibration point at 0.8 Gy. For some participants the signal intensity reported for the 0.8 Gy calibration sample was almost equal or above the intensity of the 4 Gy calibration sample. The square of the sample correlation coefficients (R^2) of the linear regression of the calibration curves reported by the participants of group B consequently vary from 0.96–0.56. Thus, the MDD ranged from 2.6–11.9 Gy. The second part of the exercise has shown that different parameters could affect the performance of the technique. An apparent variability of glass behaviour was observed, which could be caused by intrinsic variability of glass response and the different storage conditions of samples (exposition to sunlight) used by the participants. Nevertheless, even with non-uniform samples, it was possible to discriminate between irradiated and non-irradiated samples and between high and low doses: these results encourage further investigation. Another important outcome of this exercise is that, with a minimum of training, it is possible to form a functioning network based on EPR expertise in dosimetry. If more robust protocols could be proposed, a network with large capacity could be easily set up in Europe and possibly worldwide.

During the RENEb project, in close collaboration with EURADOS Working Group 10, samples of the first ILC were re-evaluated with different approaches (EPR measurements in second harmonic mode, improved model for fitting) and various investigations (inter-sample variability of EPR response and the presence of radicals induced by solar light, gamma-rays or 254 nm UV). A summary of this work was presented at the EPRbiodose conference held in October 2015 in Hannover, NH, USA (<http://iaber.org/>) and papers are in preparation. Preliminary outcomes have shown that: (a) 95% of the samples showed similar dose response and time stability within ±7%, which was not sufficient to explain the scattering of the calibration data points for some participating laboratories (De Angelis et al. 2015); (b) solar light was confirmed to be a source of confounding EPR signals (Fattibene et al. 2015); (c) at least six signals (native, radiation-induced, light-induced) were identified in the EPR spectrum (Wieser et al. 2015). At the end of the project, it can be concluded that new protocol improvement may be possible. Thus, more investigations are still needed to fully establish the technique.

Quality Assurance (QA) and Quality Management (QM) program

Within RENEb, a work package was dedicated to the elaboration of a QA and QM programme (Gregoire et al. 2016).

Specific recommendations and guidelines were implemented in a manual for EPR and OSL techniques. Besides the recommendations made to establish protocols for sample preparation and measurements, and signal analysis, a minimum number of exigencies were listed regarding the control of the instrumentation (stability, sensitivity, parasitic signals), the traceability of these controls and any operation on the instrumentation (maintenance, reparation, upgraded, modification), the storage and the traceability of raw data and of the data analysis, and the storage of the samples. For EPR, the list of quality controls requested is based on the recommendations described in the ISO 13304-1:2013 (ISO 2013) relative to the minimum criteria of EPR dosimetry. The procedures implemented in the laboratory should describe the quality control practices. The minimum documents required for QA and QC are also listed in ISO 13304-1:2013, as well as the information on samples, measurements and dose estimation that have to be recorded.

There is not yet any standard for the use of OSL in retrospective dosimetry, so an approach similar to EPR was followed to elaborate recommendations and requests. Recommendations are made to elaborate the different protocols for the different steps of the dose estimation from SMD extraction from a phone to dose reporting. As for EPR, it is also necessary to elaborate a minimum level of documentation to describe the quality control practices (source calibration, control of the photomultiplier performances and control of background level), the traceability of these controls and the data storage.

Capacity of the network

The capacity of the network is based on the sample measurement capacity provided by RENEb partners during the virtual exercise organized at the end of the project. The exercise is described in this special issue in Brzozowska et al. (2016). After 27 weeks, partners were asked to categorize virtual victims based on the results of different assays. In addition, partners have to report each week on the available capacity in their laboratory for the different assays. The averaged capacity on the whole exercise duration is reported for EPR and OSL in the Table 1, and compared with dicentric assay capacity in manual and automatic mode.

In the RENEb network, the EPR and OSL capacity are similar to the dicentric assay capacity in manual or automatic mode, but the number of laboratories involved is very different. Normalized to the number of involved laboratories, one can observe that EPR and OSL present much larger capacity. Recent ILC have shown that it could be possible to use the EURADOS network in case of an emergency situation. Between 10 and 15 laboratories for each method (OSL and

EPR) have sufficient skills, experience and competences to perform the requested analysis. It is worth noting that the EURADOS network also involves extra-European laboratories, from South Korea and USA. The capacity of the EURADOS laboratories was not investigated. Some of the laboratories belong to universities with less manpower to perform the analyses. Assuming the lower average capacity values (400 for OSL and 710 for EPR) and extrapolating to the number of laboratories involved in EURADOS, the total capacity for an enlarged network could range between 800 and 1200 samples per week for OSL, and between 2370 and 3550 samples per week for EPR. For comparison, the present total capacity of RENEB for all assays was estimated at 4522 per week. This number includes the γ H2AX assay that represents 40% of the RENEB capacity, but whose applicability is limited in terms of scenario because the assay is only pertinent in the first 24 h after the irradiation.

It is worth noting that the follow-up of RENEB will aim to consider the implementation of TL techniques (Ainsbury et al. 2016). The EURADOS laboratories generally have the capability to perform either TL or OSL, although usually the same people perform the analysis, so their capacity will remain similar to that of the existing network. TL is a technique that is more widely used than OSL, so it is hoped that new members can be recruited.

Discussion

Programmes of research and development aiming to increase the capacity of dose evaluation for large-scale events have mainly been based on bioassay, particularly cytogenetic techniques, however, the so-called 'physical dosimetry' approaches are now being actively considered. These approaches are still under development and do not have the same level of standardization as biological assays, but they nevertheless present some key advantages:

- Signal specific to an irradiation;
- No specific conditions for transportation;
- No imposed delay for starting measurements at sample reception;
- A large laboratory Network is already constituted;
- Large measurement capacity is available;
- The methods have complementarity with biological assay (Multi-parametric approach);
- Signal stability (EPR);
- Dating of irradiation (OSL + EPR), (pertinent for insidious scenarios).

There are also some weaknesses in this approach. As it is based on materials collected from technological devices, the future applicability of the relevant dosimetric methods and techniques will be completely dependent on technological evolution. The composition of the materials considered for dosimetry may change, these materials may disappear in future generations of mobile phones and there are already moves towards smaller resistors with accompanying reduced signal and greater handling

difficulty. For example, LCD with a passive matrix was largely used in the first generation of electronic devices. It was considered as a good candidate for dosimetry 10 years ago, but is now found in only a very small number of devices. Technological development will require that dosimetric tests are performed regularly on the new models of smartphones, and other devices that may become available. It will also be important to identify possible new materials present in phones, with better dosimetric properties. However, major effort needs to be devoted to the development of protocols and readers for physical dosimetry of biological samples such as nails, hair, or tooth enamel, since these will not change technologically.

The other problem is that present protocols for both EPR and OSL require collection, transport to the laboratory, dismantling and destruction of the phones. After collection of SMD from the electronic board for OSL, the phone will not be repairable, though if a screen is properly dismantled it can be replaced: modern touchscreens are easier to dismantle and replace than older LCD versions. Nevertheless, in case of large-scale events, spending time on screen dismantling will create a bottleneck in the process of dose estimation that will considerably affect the speed and capacity of measurements. As smartphones are relatively expensive devices, it is not obvious that people will willingly provide them for a destructive analysis, especially if no phone replacement or data recovery is envisaged. Therefore, it may be wise to concentrate effort on approaches that allow cheap and easy repair of the phone, or avoid removing it to a remote laboratory by developing on site measurements with non-destructive methods. Therefore, it makes sense to continue effort of research for TL and EPR analysis on touch screen, even if dosimetry techniques (TL and OSL) on SMD are qualified as operational.

ILC performed in association with the EURADOS network has demonstrated that with a minimum of effort and investment, a large functional network could be established. The strength of this approach is to use the spectrometers and readers available in research laboratories, without the need for additional technical developments and investments such as are required for in vivo approaches.

In the follow-up of RENEB, RENEB plus, to develop the capacity of measurements with physically based dosimetry techniques, it is envisaged to welcome new partners and to implement additional techniques in the network, such as TL on glass and on SMD. To implement EPR techniques on glass or on other types of materials, more work is needed to understand sample behaviour and develop approaches to overcome the identified problems. The longevity of the studied approaches depends on the presence of the analyzed materials in future generations of electronic devices. Even if some of these techniques can be considered as already applicable for triage, and some others as promising, in parallel with the work need to fully established or improve these techniques, other techniques applicable to biological samples should also be investigated.

Ex vivo EPR dosimetry on nails and hair remains very attractive, but it is not yet possible to detect with accuracy

doses below 5 Gy (Romanyukha et al. 2014; Trompier et al. 2014). Ex vivo EPR on enamel with mini-biopsy has been used for radiation accident dosimetry with success and can be considered as a valuable tool for triage, but it required specific instrumentation not available in most of EPR laboratories (Romanyukha et al. 2014).

Conclusions

Physically-based dosimetry techniques present large and new possibilities for accident dosimetry, especially in the case of large-scale events. Some of the considered techniques can be regarded as operational (OSL on SMD) and provide a large increase of measurement capacity of existing network. Other techniques and devices currently undergoing validation or development in Europe could lead to considerable increases in the capacity of RENEB accident dosimetry network

Disclosure statement

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