Retrospective Dosimetry Using OSL and EPR of Fortuitous Objects
Advantages and Challenges of Using Physical Biodosimetry to Assess Individual Exposure in a Large Scale Unplanned Event

Although the primary intent of our review was to highlight physical biodosimetry, i. e., techniques that measure physical changes in the body following exposure to ionizing radiation, EPR, like OSL, has also been proposed as a means to assess personal exposure by measuring objects found on or nearby the individual at the time of exposure. Since both OSL and EPR techniques assess dose at the specific location where the object was found on the body, they have the potential to assess whether the exposure was non-uniform, i. e., implying that the person received partial body exposure, which has important implications for medical response.

While Optically Stimulated Luminescence (OSL) was initially expected to be developed as an alternative physical biodosimetric method for informing medical response to unplanned largescale radiation events, the principal disadvantages of signal sensitivity to the presence of environmental light and fading over time proved to be intractable for this use.

Retrospective dosimetry using OSL

Sergey Sholom et al. tested such uses, particularly in OSL of teeth. [1, 2, 3] While the sensitivity reported was acceptable for triage, it depended on making measurements immediately after exposure and eliminating any exposure of the sample to light. In addition, corrections for fading would be required. For example, after a 24-hour delay between irradiation and OSL measurement, the minimum detectable dose (MDD) was found to increase from approximately 0.2 Gy to approximately 1 Gy [3].

Possible triage application?

For a possible triage application, both stimulation delivery and emission collection would need to be done in situ, with the additional assumption that at least some teeth in the mouth had been protected from environmental light. This latter assumption would require verification in order to ensure the OSL measurement was valid, but there is the additional technical challenge of how to deliver the stimulation light to the teeth in the mouth and how to collect the emitted light back. While optic fiber may be used for this, its use would entail significant loss of signals and an increase of MDD to a level greater than the recommended threshold for triage, i. e., 2 Gy.

Similarly, when OSL with nails was tested, the OSL emitted was observed to occur mainly from impurities on the nails' surface such as small particles of sand [2]. This, together with a requirement that any nail considered for OSL dose reconstruction should be protected from environmental light for the entire interval between irradiation and OSL readout, complicates the possibility of using OSL for biodosimetry with nails.

Objects found near the exposed subject

Consequently, the current use of OSL for dosimetry, reviewed briefly below, focuses on fortuitous objects found near the exposed subject.

How OSL Detects Radiation-Induced Physical Changes in Tissue or Other Compounds

OSL dosimetry is based on measuring the byproducts that occur when electrons and holes, generated in a sample by ionizing radiation, are trapped on some energetic levels (so-called electron and hole traps). When the irradiated sample is stimulated by light at some wavelength (usually in blue wavelength range), trapped electrons may be excited to the conductive band and then recombine with holes emitting the light at another wavelength (usually in UV range, but other combinations of stimulation and emission wavelengths are also possible). The intensity of the emitted light is usually proportional to the absorbed dose and may easily be detected by a photomultiplier tube (PMT), making OSL one of the most attractive and sensitive dosimetric techniques.

OSL: sensitive dosimetric technique

Advantages

OSL has several characteristics making this technique favorable for dosimetry. First, OSL is a very common phenomenon. Most solid state materials will exhibit a radiation-induced OSL response in certain circumstances [4, 5].

Second, the equipment for stimulation and registration of the OSL is very simple. It consists of a light source for sample stimulation (this could be a light emitting diode or a laser), a package of optical filters to separate the weak emission light from the much more intensive stimulation beam, and an emission light detector (usually a PMT). The corresponding OSL reader may be manufactured in a portable deployable version at a cost of a few hundred US dollars [6].

Challenges

The OSL dosimetry technique has several challenges that restrict its possible use in many dosimetry-related applications. The most critical limitation, as already mentioned, is the sensitivity of samples to environmental light. This restriction means that all samples which are considered as potential retrospective/emergency dosimeters should be protected from environmental or other light for the entire interval between exposure and the OSL readout.

Sensitivity of samples to environmental light

Another possible issue is the instability (fading) of the OSL signal with time. ***Elisabeth L. Inrig*** and her colleagues first observed fading in samples of surface mount resistors [7], but this phenomenon seems to occur in most fortuitous OSL materials. Fortunately, the fading characteristics of any potential dosimetric material may be determined in advance, and corresponding fading correction coefficients may be calculated and applied to the OSL-related doses.

The next potentially important challenge is the background OSL signal, which was observed in some materials [8]. Special approaches were proposed to overcome this issue, e. g., using the difference in shape between background and radiation-induced OSL signals. The final challenge important to mention for uses of fortuitous objects as dosimeters is the possible difference in doses delivered to the OSL dosimeter, e. g., a mobile phone or a credit card, and to the individual-owner of this fortuitous dosimeter. ***Joshua R. Chandler*** and colleagues have proposed using special conversion coefficients calculated using a Monte Carlo method to determine conversion factors for mobile phones carried at typical locations on or near to the body [9].

Radiation-induced OSL signals

Types of Parameters for OSL Biodosimetry

OSL using parts and components of mobile phones

Mobile phones as possible emergency/retrospective OSL dosimeters have been popular since 2008, when Inrig and her colleagues observed the prominent OSL signal in surface mount resistors (SMRs) extracted from a phone and exposed to some ionizing radiation dose [7]. The signal was unstable and decayed with time logarithmically, which was attributed to the tunneling mechanism.

Afterwards, SMRs were the subject of numerous studies focused on improving sensitivity and understanding and characterizing the fading effect better. Several international comparisons were devoted to dose reconstruction using SMRs, which resulted in developing robust dosimetric protocols using OSL with SMRs [10]. The values of MDD for SMRs are in the range of tens of mGy for OSL readouts immediately after irradiation and hundreds of mGy if measured several days after exposure. The main drawback of OSL dosimetry with SMRs is the destructive character of this technique, i. e., to get the SMR for dose assessment, the phone must be totally disassembled, which is inconvenient for the phone owner in a stressful post-exposure situation.

Destructive character of this technique

Another possible issue for OSL dosimetry with SMRs is the size of the resistors. The trend in improving the technology of mobile phones is to reduce their size, which makes it difficult to locate the SMRs within the phone as well as to extract them. (Note too that all these procedures should be performed under laboratory red light to protect samples from bleaching.)

Other easily available electronic components of mobile phones that may be used for OSL dose reconstruction are surface mount inductors. They have a chemical composition quite similar to SMRs (both materials are types of alumina) but usually demonstrate higher sensitivity and much larger variability of dosimetric properties [11]. OSL dosimetry with inductors has the same drawbacks as mentioned above for SMRs.

Recently researchers have paid increasing attention to a so-called 'back-glass' in mobile phones for use as a possible emergency OSL dosimeter. Most modern smartphones are equipped with back-glass, which is required if a phone is charged wirelessly. The back-glass is usually made from some special chemically strengthened glass like Corning® Gorilla® Glass, which is a very bright OSL material. If the back-glass is protected from environmental light (which happens when the phone is used with a case), then the back-glass may be used as an OSL dosimeter.

Back-glass used as OSL dosimeter

Dosimetric properties of the back-glass have been recently tested in several studies [6, 12]. The dose response appears to be linear, at least in the range below 8 Gy; the fading follows a power function; the MDD values are within tens of mGy dose range. However, the most beneficial feature of using OSL dosimetry with back-glass is the possibility to readout the OSL signal in situ, i. e., without destroying the phone. ***Sergey Sholom*** and ***Stephan W. S. McKeever*** tested this option using a custom-build OSL reader shown in Figure 1 [6]. This reader was recently tested within the framework of a large inter-laboratory comparison

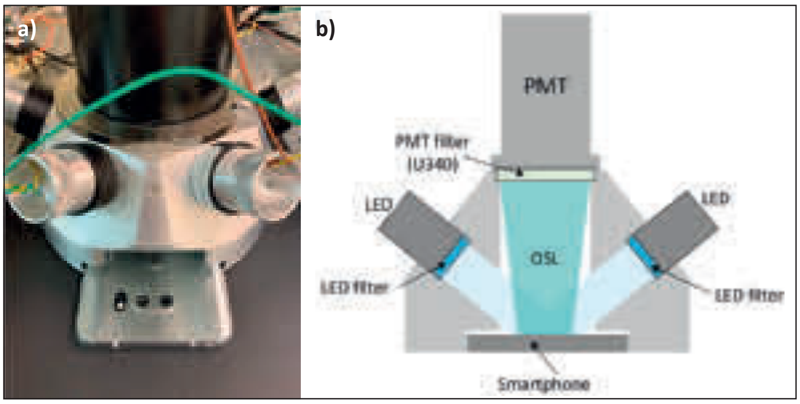

Figure 1: Custom-made OSL reader designed to measure with OSL from whole smartphones: (a) Photograph, with phone in place; (b) Schematic showing light-emitting diodes (LED)s and photomultiplier tube (PMT) filters. Reproduced with permission from Sholom & McKeever [6].

performed by RENEB[1]), which involved 46 scientific institutions and 8 different biological and physical dosimetric techniques. (Results of this inter-comparison are in process of being prepared for publication.)

After exposure, the phones, in cases, were shipped to the OSL laboratory at Oklahoma State University and measured immediately after arrival (which was 3 days after irradiation). The deviation of the reconstructed, fading-corrected doses from corresponding nominal values in the dose range 1.2 to 3.5 Gy was within 20 %, which is acceptable for potential triage applications.

In summary,

OSL dosimetry with SMRs is still considered as the most reliable and robust OSL dosimetric technique with mobile phones. This technique was validated in several international intercomparisons. The main drawback of this technique is its necessity to totally disassemble and destroy the phone, which in many case is inconvenient and even unacceptable for the phone owner in a stressful after-accident situation. An alternative component of mobile phones used for OSL dose reconstruction is back-glass. OSL measurement of back-glass is totally non-destructive; only 10 to 15 min. are required to assess the dose of one phone; the phone remains fully functional throughout all procedures.

OSL with other fortuitous materials

The current list of the fortuitous materials that can be used as emergency OSL dosimeters includes, among others:

- different plastic cards like credit/debit cards,
- parts and details of clothes and shoes,
- paper bills and business cards, and
- silicate dust.

All these may be used, in given circumstances and limitations, for dose reconstruction at the level 2 Gy or below, which is acceptable for possible triage application. Credit/debit cards with chips are most interesting and promising among the above items. As noted by Sholom and McKeever [13], more than 470 million such cards were in circulation in 2019 in the US alone. Each such card has a chip module covered by polymer material, which exhibits a strong OSL signal after exposure to ionizing radiation. Therefore, these chips may be useful as OSL dosimeters, especially taking into account that they are totally protected from environmental light by a metal strip or an opaque plastic.

Credit cards with chips are most interesting.

Other advantages include:

- Chips can easily be extracted using scissors.
- All procedures took about 1 min per chip.
- No chemical treatment is needed, i. e., they can be tested "as found".

Tested "as found"

Additionally, the dose response of chips is linear, at least within a range 0 to 7 Gy. Signal fading changed logarithmically (OSL was found to be reduced about 64 % and 86 % following fading times of 1 and 10 days, respectively. MDDs ranged between 7.9 mGy and 26.3 mGy for different samples, which is quite acceptable values for emergency dosimetry.

In summary,

several different fortuitous materials have been tested as possible emergency OSL dosimeters. Most promising are credit/debit cards with chips. They are widely available, do not require any special sample preparation, demonstrate high radiation sensitivity and therefore could be used as emergency dosimeters for triage application.

Retrospective Dosimetry Using EPR

Since both OSL and EPR techniques assess dose at the specific location where the object was found on the body, they have the potential to assess whether the exposure was non-uniform, i. e., implying that the person received partial body exposure, which has important implications for medical response. This use assumes that the object being measured, e. g., cotton clothing, was identified with different places on the body.

Important implications for medical response

[1] See www.reneb.net/about/ for more detail about this organization.

In the following we briefly review the research using EPR on these 'fortuitous objects'. While there are potentially many candidates, the suitability of materials such as

- glass,
- clothing fibers (cotton or wool),
- sugars, and
- plastic

for dosimetry in largescale incidents depends on their likely near universal use and comparability across manufacturers. For example, the common use of mobile phones makes this personal item especially suited for estimating individual dosimetry based on radiation induced chemical changes in cell phone components, especially in its glass. In addition, since the amount and stability of free radicals produced by ionizing radiation varies significantly across materials, their suitability for EPR critically depends on their radiation sensitivity and signal stability over time.

Glass

Glass is found in innumerable objects that could be used for EPR dosimetry, e. g.,

- windows,
- windscreens,
- watches,
- display windows of electronic devices such as mobile phone screens.

In addition, glass is an easy-handling, chemically inert, inexpensive material, and it can be reduced into small-size fragments or even particles. One disadvantage is that preparation of the samples for EPR measurement usually requires disassembly or destruction of the object.

Glass an easy-handling material

Glass has been studied as a potential EPR dosimeter for both low and high doses and has been used for radiation accidents [14]. All investigated glasses have a background signal (generally caused by impurities or metals introduced during manufacturing), which partially overlaps the RIS. However, mechanical stress such as breaking glass does not generally induce signals, except when crushing glass into a fine powder (< 315 µm) [14].

The post-irradiation signal in glass is not stable. The signal fades with time, especially over the first 24 to 48 h, and depends on the storage temperature (very cold temperatures reduce fading) [14]. Decay kinetics of the RIS also vary in different glasses. Due to the complex nature of the superimposing EPR lines, in some glasses the fading results not only in a drop of the signal amplitude, but also in changes in the shape of the spectra measured at different times after irradiation. Nevertheless, if feasible to optimize the temperature and the duration of heating, dose can be estimated with relatively good accuracy, and the RIS appears to be linerally related to dose up to 500 Gy.

Dosimetric signal in glass samples

Generally, the determination of the dosimetric signal in glass samples is more accurate and reliable when using methods based on signal decomposition and fitting rather than on measuring the amplitude of the spectral lines [15].

Sensitivity of detection

In particular, studies have shown that dosimetry based on glasses from ubiquitous utility items kept close to the body, like mobile phones and wrist watches, allows achieving a sensitivity of detection on the level of 1 to 2 Gy, which is sufficient for triaging exposed individuals before receiving medical attention for acute radiation syndrome (ARS).

However, 2 crucial problems need to be overcome:

Implementating a proper correction in analytical procedures to account for the rapid fading of the RIS during the first 6 to 10 days after irradiation and for potential exposure of the irradiated glass to light, particularly to UV which can introduce significant over or under estimation of the dose.

For materials for which a linear dose curve is a reasonable assumption, one promising technique to overcome the variability in the manufacturing process is to use a dose-added approach for each sample, i. e., creating the dose response curve to be used by adding known doses to the sample after its initial measurement and observing the relationship between the incremental signal and the known dose.

New resonator geometry

One promising future development is using a new resonator geometry, the surface array resonator (SRA), that will allow in situ measurement of glass, i. e., without disassembling or destroying the glass such as in a mobile phone. The SRA limits the depth penetration of the B1 field, thereby eliminating interference from the underlying circuit components, instead focusing the analysis on the glass screen.

Other Materials as Fortuitous EPR Dosimeters

Sugar

Sugar is not feasible for use in largescale incidents but has been used successfully in a few radiation accidents [15]. The EPR spectra of pure-form irradiated sugars are complex and differ for each sugar type because of the presence of several radicals and due to the hyperfine coupling. For example,

a) At least 3 radicals have been identified for sucrose with 9 hyperfine coupling tensors with protons.
b) The number of induced radicals associated with extreme powdering of sugar has been reported to be equivalent to < 10 Gy; and
c) Some non-irradiated samples show a background signal [14].

Nevertheless, the dose-response curve was found to be linear in the 0.5 to

100 Gy dose range for gamma rays irradiation [14].
Similar to glass, the EPR signal is unstable during the first hours after irradiation and depends on dose and humidity. On a longer time scale, the number of radicals scarcely changes over several years in the natural environment, even when the atmospheric temperature varies [14].

Plastics

Investigations of the types of and objects containing plastics differ somewhat by their intended use as EPR dosimeters, e. g., using

- polyvinyl chloride (PVC) floor plates,
- polyethylene (PE) plastic bags, credit cards, and plastic buttons

in the context of small radiation accidents and using plastics from

- mobile phones, eyeglasses, watches, and badge holders

in the context of larger unplanned radiation exposures [14].
Most plastics exhibit different kinds of background EPR signals prior to irradiation, and the shape of RIS also depends on the type of plastic. Thus, most of the irradiated plastic buttons investigated (mainly made of polyesters) exhibit a singlet line similar to the background signal while EPR spectra of other plastics, e. g., poly(methyl-methacrylate) (PMMA), polycarbonate, and Columbia Resin #39 (CR-39) show various, more complex patterns after irradiation. Even for the same type of plastic, e. g., CR-39 or polycarbonate (the main constituents of eyeglasses), various spectrum shapes were observed. Moreover, the dose response was usually nonlinear; the dose sensitivity was remarkably variable, i. e., up to ten-fold, depending on type; and the RIS faded with time precipitously, becoming quickly indistinguishable from background (after 5 to 7 days) [14]. While some investigators hold out promise for buttons, these general problems obviate the use of plastics as reliable physical dosimeters in large incidents.

Dose response usually nonlinear

Cotton

Use of cotton in fabric is obviously ubiquitous and its use in clothing would allow mapping the uniformity of dose by measuring samples in contact with different parts of the body. Some EPR studies showed good agreement between doses based on cotton clothing and physical samples (teeth or bones) from the body of victims of accidental radiation exposure. Background signals, specific for each manufacturing process, have been reported. After a nonlinear, preliminary stage due to the background signal, cotton shows a linear dose-response curve to gamma-rays in a range from 10 to 104 Gy, although the signal is also detectable at < 1 Gy [14]. Cotton exhibits EPR signal fading with time at room temperature, revealing the existence of several decay components.

Signal is also detectable at < 1 Gy

Different factors further complicate the analysis of irradiated cotton. The signal fades in a complex pattern, and exposure to UV, detergent residue from washing, food or dust residue on the fabric can cause variations in the signal. Water absorption in particular can change the mechanical properties of the fiber [14].

Wool

Despite its near-ubiquitous use in fabric, only one study has reported using wool for radiation accident dosimetry purposes and none at lower doses. The EPR signal of an irradiated sample of dry wool measured in air is a singlet. Its intensity is quite weak because of the high radical recombination due to the oxygen molecules diffused into the wool fiber. The EPR background spectra are multi-component, attributed to the pigments. Signal decay is a function of time and increases with sample handling, e. g., the scales, which are densely cross-linked with disulfide bonds, are the most susceptible to any outside interference [14].

Conclusions Regarding OSL and EPR Dosimetry Using Fortuitous Objects

The use of fortuitous objects, using either OSL or EPR techniques, for estimating the dose received by an individual in a largescale radiation incident is determined both by the likely ubiquity of its being found on or near the victim's body and by the likely uniformity of its manufacture for all the objects to be measured (monopoly would be advantageous here!)
Moreover, the ability to obtain a reliable, unique, sensitive signal to estimate dose during the period between exposure and likely measurement of the sample is also critically important.
These are indeed difficult challenges, especially for use in triaging a large number of victims for immediate treatment of ARS, instead of for retrospective reconstruction of accidental exposures.
Mobile phone glass is generally believed to hold the most promise for both techniques for this purpose, despite having many challenges yet to be solved, as reviewed here.

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Bemerkenswert

„Auch eine Enttäuschung,
wenn sie nur gründlich genug ist,
bedeutet einen Schritt vorwärts."
Max Planck (1858–1947)