The use of luminescence techniques with ceramic materials for retrospective dosimetry

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Abstract. Luminescence techniques are being used with ceramic materials to provide evaluations of integrated external gamma dose for dose reconstruction in populated areas contaminated by Chernobyl fallout. A range of suitable ceramics can be found associated with buildings: on the exterior surfaces (tiles), within walls (bricks) and within the interiors (porcelain fittings and tiles). Dose evaluations obtained using such samples provide information concerning the time-averaged incident gamma radiation field, average shielding factors and, with the aid of computational modelling techniques, dose estimates at external reference positions.

1. Introduction

Previous dose reconstruction studies in Japan [1] and the USA [2] have demonstrated the value of using thermoluminescence (TL) techniques with ceramic materials for the retrospective measurement of gamma dose many years after the event. Similar promise has been demonstrated in a preliminary study of the use of ceramic materials from the Chernobyl region [3,4].

Since the Chernobyl accident the need to apply a range of techniques for dose reconstruction has become increasingly evident because of the complexity and scale of the fallout and the sporadic nature of monitoring procedures, particularly in populated rural regions. One of the elements within the project ECP10, *Retrospective Dosimetry and Dose Reconstruction*, has been to further develop the application of luminescence techniques. The experimental studies have been combined with the use of computational modelling to aid the deconvolution of dose evaluations to ceramics and to test predicted variations of accrued dose with location. This paper discusses current progress in development of the experimental approach including, the assessment of the types of sample available in contaminated regions of Belarus, Russia and Ukraine, the levels of transient fall-out dose which can be measured and the relationship between dose in ceramic to dose at an external reference point.

2. Scope for sampling

A range of contaminated regions has been examined in programme of field work, including: Pripyat and settlements within the Exclusion Zone, Southern Belarus and the Kaluga and Briansk regions of Russia. While Pripyat has served as a study site for methodological development because of its proximity to the power plant and the extensive scientific work which has been undertaken in the vicinity of the town, the examination of more distant populated (and evacuated) settlements has provided important information concerning the nature of the fall-out, the type of buildings and potential ceramic samples.

2.1 The availability of ceramic samples

For retrospective dosimetry measurements, ceramic samples from fixed locations are sought from the exterior surfaces of buildings, from interior shielded locations and ideally throughout the walls (Fig. 1).

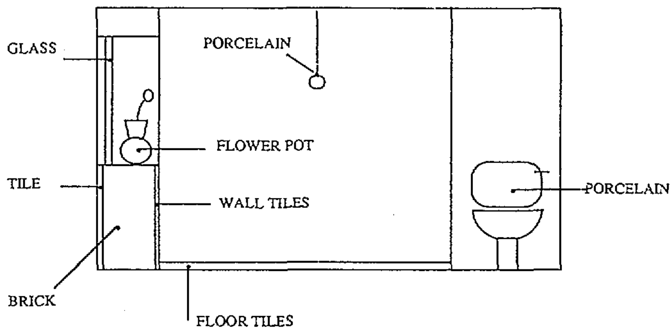

Figure 1
Location of potential ceramic samples

The construction of dwellings and buildings differ widely; in urban areas they may be of concrete with additional ceramic fittings/decorations. Most urban buildings of the 1970's, particularly multiple storey apartment blocks of the type found in Pripyat, however, are constructed of concrete. Although concrete is not, so far, suitable for luminescence dosimetry, glazed terracotta or earthenware decorative tiles fixed to the exterior walls are suitable and have been used in this study. In Zone 1 of Pripyat (the most heavily contaminated region; see [3]) there is a small number of substantial brick buildings with outer tile cladding in Pripyat which provide the opportunity to extract samples at a number of depths, although it is to be noted that mortar is unavoidably present in a region of substantial absorption for external gamma rays (Fig. 2).

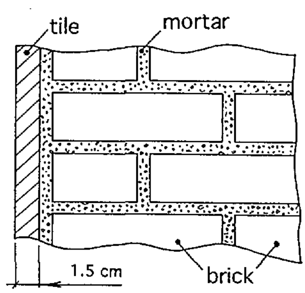

Figure 2. Location of mortar in brick wall cross-section

The evacuated and populated settlements surveyed for potential samples present a more diverse range of dwellings in age (up to ~50 years) and style. Older traditional houses are constructed of timber frame and board with plastered interiors and have a substantial brick oven with chimney stack. One or two courses of external bricks are often used as part of the foundations between the timber building and an underlying a concrete plinth; however where they are close to soil, adsorption of contamination is likely to have occurred. For more recent houses concrete bricks have been frequently used. As in Pripyat, the use of porcelain fittings and components is commonplace.

Within the interiors of buildings, high fired terracotta floor, wall and skirting tiles and porcelain sanitary and electrical fittings provide a ubiquitous source of samples, present in most rooms and available in abundance within bathrooms. Porcelain which is also available in exterior locations (usually light fittings and as electrical insulators) has an important advantage over exposed brick because the glazed surface of porcelain inhibits the ingress of fall-out products.

The measurement of accrued dose for interior, shielded and exterior samples provides information on the time-averaged shielding factors. The more difficult experimental tasks lie with samples where the transient dose is comparable with the accrued natural background dose, such as interior samples and in areas of low contamination. Given the wide availability of porcelain in rural areas in external positions and within the interior, work so far has concentrated on developing the use of porcelain for dose evaluations.

Where buildings are of brick construction, the opportunity to obtain samples of ceramic throughout the wall thickness allows the determination of dose as a function of depth (referred to here as a dose-depth characteristic). Additionally it provides dose evaluations for exterior and interior positions of the wall, indicating the effective shielding factor. Such dose profiles can provide information on the nature of the (time-averaged) incident gamma radiation field which is related to the form of the profile. In a series of experiments with laboratory photon Cs and Co sources the feasibility of obtaining dose-depth curve experimentally using ceramic material has been demonstrated (Section 3.1.1).

The investigation within ECP10 has focused on aspects which are concerned with matching capability of the method to dosimetric requirement, including:

i) The dose response characteristics of ceramic samples, in particular the minimum dose which can be resolved. There is little previous experience in the use of ceramics of the type found in the contaminated areas. Thus basic characterisation needs to be performed (e.g. linearity of response with dose, changes of sensitivity with secondary laboratory heating etc.).
ii) The location of ceramics and their suitability for providing appropriate information for use of computational modelling has been used to identify the position and orientation which could be used to differentiate different source configurations.
iii) The level of accumulated natural background dose within samples at the time of measurement.
iv) The accuracy with which the transient dose can be determined.

Table 1 Sample types

Fired clay or composites:	Brick; terracotta tiles; glazed wall and floor tiles; cladding tile; skirting tile; flower pots.
Porcelain:	Electrical insulators including, switches, lamp holders, domestic and industrial fuses, power insulators.

3. Experimental methods

The dose evaluated by luminescence techniques, D_L, corresponds to the sum of dose arising from: a) naturally occurring radionuclides within ceramic materials and the surrounding medium and b) transient fall-out contamination. In the same way that TL dosimetry phosphors can be used to register the administration of absorbed dose and, at some later time, to yield a quantitative measure of that dose, minerals present in ceramics such as quartz and feldspar can perform the same task, albeit at generally higher dose levels. The transient gamma dose arising from exposure due to an accident, D_X, determined by luminescence techniques, is given by

$$D_X = D_L - A(\dot{D}_\alpha + \dot{D}_\beta + \dot{D}_\gamma + \dot{D}_c) \qquad (1)$$

where, D_L = accrued dose determined by luminescence measurements (TL or OSL); A = sample age in years; $\dot{D}_\alpha$, $\dot{D}_\beta$, $\dot{D}_\gamma$, $\dot{D}_c$ = effective annual alpha, beta, gamma and cosmic dose, respectively, due to natural sources of radioactivity.

Several luminescence (using either thermoluminescence, TL, or optically stimulated luminescence, OSL) techniques are available to evaluate D_L. Apart from the means of stimulating luminescence, they are differentiated by the mineral composition of the sample extracted for measurement, the grain size, the procedures employed to determine the accrued dose and, most pertinent to dosimetry, their sensitivity. These aspects and the various components of the natural background dose-rate are determined using direct and indirect analytical techniques. The details of the experimental work are given in a companion paper [5]. Three of the techniques used are referred to as the *fine-grain*, *quartz inclusion* and *pre-dose* techniques which were originally developed for dating archaeological ceramics [6]. The pre-dose technique [7] generally offers the highest sensitivity and has also been applied to porcelain from interior locations. Porcelain is usually prepared as cut slices rather than crushed and sized material in the case of bricks and tiles.

During the measurement of TL, the release of the trapped charge carriers and consequent emission of luminescence is achieved using thermal stimulation; for OSL measurements the release of trapped charge is achieved by stimulation using light within particular wavelength regions. Under constant illumination of a sample and using suitable optical filters in the detection system to reject the stimulating light, an OSL decay curve is obtained. The wavelengths used for stimulation differ according to the mineral(s) within the sample; in the case of quartz green light (e.g. 514 nm, Ar ion laser) is used for stimulation and for feldspars it has been found that infra-red wavelengths are also suitable, enabling simpler light sources to be used. Under infra-red stimulation the emission is referred to as infra-red stimulated luminescence (IRSL). Procedures employing optical stimulation are a recent introduction to the field and as such are under development [8-11].

For samples receiving little or no transient fall-out dose, D_L is substantially due to natural radiation dose. The annual dose in equation 1 arises from the decay of the ^{238}U, ^{232}Th series and ^{40}K within the sample and the surrounding medium. For typical brick structures the accrued dose due to natural sources of radiation is expected to be in the region of 2.5 - 3.5 mGy/a with contributions of approximately 60%, 25% and 5% for beta, gamma and cosmic components respectively (using inclusions, where the internal radioactivity and external-grain alpha dose contribution is assumed to be negligible). Thus the accrued dose due to natural sources of radiation is expected to be roughly 30 mGy per decade - this forms the baseline above which further transient dose contributions are made. In areas of low fallout or in heavily shielded locations the uncertainty associated with D_X will be strongly influenced by the uncertainty associated with the determination of the age and annual dose-rate. Consequently for heavily shielded interior locations, both high luminescence sensitivity and accurate annual dose evaluations are sought. The precision (and accuracy) of D_X achieved will depend on an assessment of the particular material and location tested. A full appraisal of the levels of minimum resolvable transit dose in areas of low fall-out/high shielding awaits more extensive testing.

3.1 Sampling and testing

One of the potential rôles for luminescence is the provision of benchmark dose evaluations which can be used in conjunction with computational modelling techniques for dose reconstruction. In populated areas the nature and pattern of the fall-out is generally complex due to the variability of the weather, building structures, clean-up operations, resuspension etc. Multiple sampling is consequently desirable to test for variability and also multiple testing by means of inter-laboratory comparisons to evaluate consistency. As a first step towards this objective sites have been sought where samples are readily available and where extensive monitoring has been performed since the accident to allow comparisons of integrated dose obtained by luminescence and by computational modelling.

Field samples from Pripyat and Southern Belarus have been examined to: i) test samples from a common location in Pripyat by different laboratories using both TL and OSL techniques; ii) measure dose as a function of depth for a roof-top location and at ground level for a house in a rural district.

Evaluations of dose were obtained at the site of three apartment blocks constructed of concrete in Pripyat which have been the subject of modelling calculations [12] . Samples of external glazed tile (at ca 1m above ground level) and porcelain from interior locations were analysed [5] from the apartment blocks 8, 18 and 28 in Lenin Prospect. The mean values of the accrued dose obtained by four laboratories using TL and OSL techniques for the exterior samples (as part of the first stage of the intercomparison completed during 1995) ranged from 1.5±0.2 Gy (SD 4) to 2.0±0.3 Gy (SD 4) for four locations. Porcelain samples from a selection of locations within rooms on each floor of one of the apartment blocks were also tested, from which a mean transient dose of 100±10 mGy (SD 9) was evaluated for all locations. The ratio of external to interior transient dose (0.06±0.01) is consistent with that expected on the basis of shielding calculations [13]. A detailed calculation taking into account each sampled location is now needed to test whether the experimental results are consistent with expectation on the basis of modelling. Nonetheless, the broad agreement in the overall shielding factors is very encouraging, given that the level of integrated dose for the interior is within the lower dose range of the method.

3.1.1 Dose depth profiles

The measurement of accrued dose as a function of depth is a potentially powerful tool in employing luminescence in the field. While, as discussed below, dose determinations produced for use in dose reconstruction are likely to be based on the outer 1 cm or less of an external ceramic, the measurement of accrued dose at greater depths in the absorber medium provides verification of the presence of an external photon field in the past and is directly related to the time-averaged energy spectrum of the external radiation (assuming that any bremstrahlung generated by external beta radiation forms a minor component of the accrued dose).

The dose depth characteristic requires either cores or whole bricks to be extracted in the field. During recent fieldwork, the use of a portable percussive drill with chisel bit has been found to be convenient to remove mortar and extract the whole brick. This usually restricts sampling to one brick depth. Several experimental approaches have been used to determine the accrued dose as a function of depth in samples: i) cutting thin slices; ii) drilling with a fine masonry drill; iii) OSL scanning of the cut surface of the ceramic [5].

As part of the evaluation of the use of different experimental procedures to determine the dose depth characteristic in ceramic [5, 14, 15], measurements have been performed with tile stacks (Fig. 3a) irradiated with either Cs or Co photon beams in the laboratory. Good agreement has been obtained (Fig. 3b) between the experimental and calculated dose-depth characteristics [16].

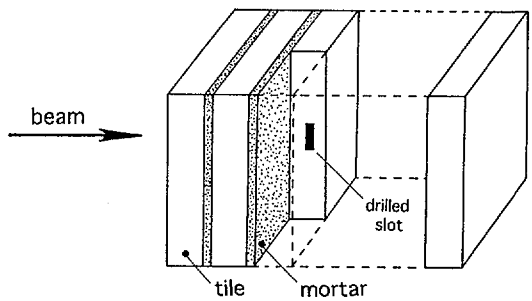

Figure 3a

Modern terracotta tiles (each of 25 mm thickness) were assembled in stacks of varying depth and irradiated (dose at surface ~2 Gy) with ^{137}Cs and ^{60}Co photon beam facilities at GSF Neuherberg. IRSL was used for measurements, giving stronger emission than TL.

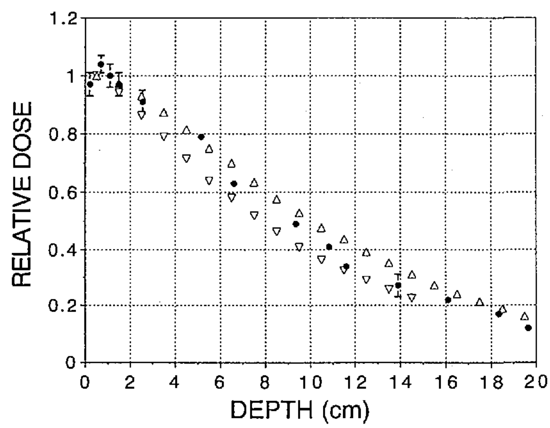

Figure 3b

Open triangles: Monte Carlo calculation of relative kerma in terracotta tile due to parallel irradiation with a Cs photon beam. The average kerma is calculated for a volume element (1cm thick x 2 cm x 2 cm) at various depths located: 1) along the central axis of the tile stack (upper symbols) and 2) along an axis parallel to the central axis located such that the volume element is located at the (same) corner of each tile (lower symbols). Filled circles: Measured absorbed dose vs depth in stack measured using IRSL. Calculation by R. Meckbach, GSF.

A dose depth characteristic obtained for a sample of brick from house in the village of Masani (north of reactor on the Belarus border) is shown in Fig. 4 - the reduction in dose with depth is consistent with an external isotropic photon field dominated by the presence of Cs. Although there are limitations to what may be interpreted concerning the specific composition of the time-averaged field [15], it provides confirmation of the energy range of the incident field. Such confirmation is important in the subsequent use of the luminescence data, as discussed further below.

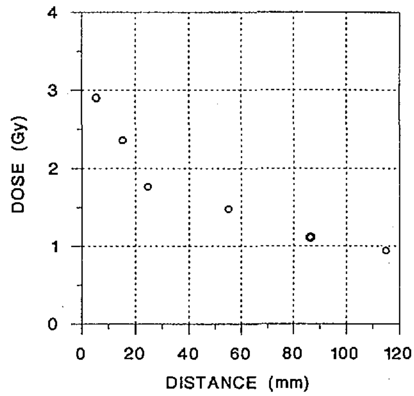

Figure 4
Dose-depth profile in brick: Masani, Belarus.

2.2.1 Roof of Polyclinic, Pripyat

The variation of dose with height was tested using samples from the upper regions of the 4-storey building in Pripyat. In the absence of full scaffolding or special working platforms, the extraction of bricks from the walls of upper levels clear of the roof was not possible. Dose evaluations for the bricks obtained from the roof (using red TL emission as discussed in [14]) indicate a substantial contribution from contamination on the roof (Fig. 5), while the dose in the outer brick is significantly lower. These results illustrate the care needed in selecting samples - for such elevations, samples from an exterior face more distant from the roofline are required to avoid the contribution of the (intermittent) roof contamination .

5. Deployment of luminescence results

Dose evaluations performed with ceramics (or other natural materials) taken from fixed structures provide determinations of the time integrated dose for the sampled location. Thus no temporal resolution is possible unless the sample had been removed from the contaminated area at a known time. A procedure of deconvolution is required to relate the dose in ceramic to dose in air at a reference point. and this can be achieved with the aid of computational modelling techniques. The luminescence determinations are consequently not entirely independent of modelling, but an analysis of uncertainty and sensitivity to variation is being developed in the computational analysis as part of ongoing work .

Calculations by Meckbach [17] have been made to relate dose in ceramic at various locations on buildings to dose at the reference position for: a) a semi-infinite cloud source and b) a semi-infinite ground source based on a source energy of 662 keV (Fig. 6). Sampling at Pripyat has provided the opportunity to investigate the predicted variations of dose with elevation (multiple storey buildings), orientation and source type. The dose to the first 2 cm of external ceramic of a wall at 1 m from the ground is calculated to be about half that at the same height at a reference position. This provides a convenient 'ready reckoner' (i.e. doubling the dose evaluation) in preparing luminescence measurements for use in dose reconstruction. At greater depths in the ceramic, a greater dependency on the energy of the incident radiation [17] and the source configuration is predicted. Thus the dose-depth characteristics perform an important role in providing confirmation of the assumptions made concerning the incident radiation when relating the dose in ceramic to the dose at the reference position.

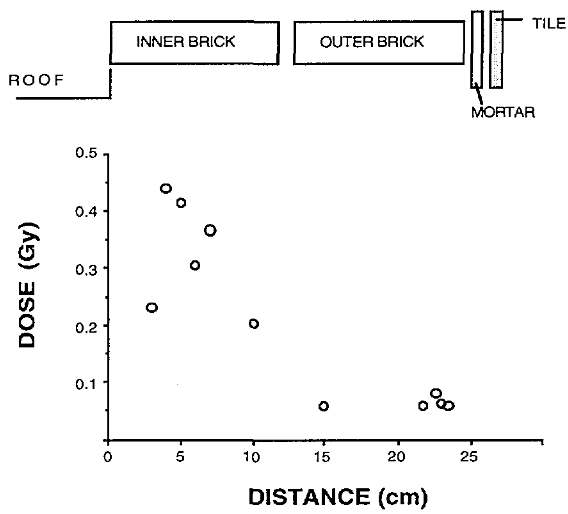

Figure 5

Accrued dose, evaluated by TL, as a function of depth in bricks located on the roof of the polyclinic, Pripyat. The upper schematic diagram indicates the location of the bricks and tiles relative to the (flat) roof.

To seek evidence to test this, however, we have seen that measurements on rooftops need to be considered with some care, since the flat bitumen roofs of multiple storey apartment blocks in Prypiat have a complex history of retention and loss of contaminants, as discussed above.

The comparison of dose estimates based on modelling calculations and experimental dose determinations using luminescence is one of the objectives of this project. In Pripyat there are extensive monitoring data which relate to the period shortly after the accident until evacuation and following re-occupation during 1987. So far there appear to be important gaps in data during the first year following the accident which will introduce additional uncertainties in calculating a value for the integrated dose since the accident at a location/area sampled for luminescence measurements. Such estimates based on integration and including an analysis of uncertainty are not trivial and will form part of future work.

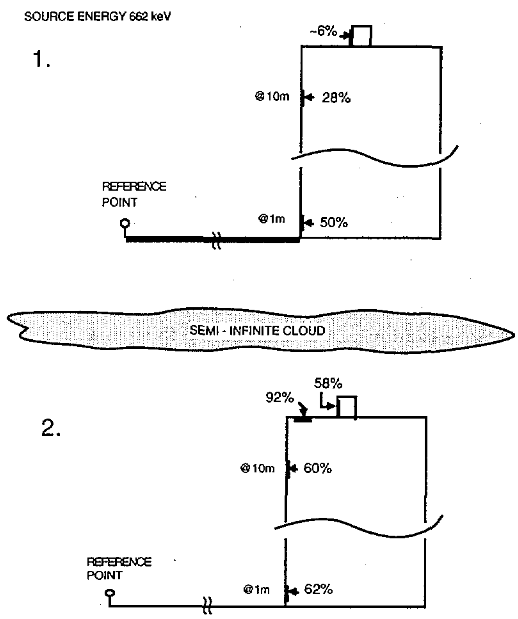

Figure 6
The calculated ratio of dose in ceramic (0-2 cm depth) at the locations indicated to dose in air at the reference point for two source configurations (1) distributed on the ground and (2) a homogeneous radioactive cloud. The source energy is 662 keV.

6. Capability and future directions

Ceramic materials, including porcelain, terracotta and bricks, have been sampled from populated areas contaminated by Chernobyl fall-out and, in general, have been found suitable for retrospective dose evaluation. The existing techniques are capable of measuring total integrated dose from ~10 mGy (at best) to well beyond 10 Gy. Substantial improvement of sensitivity has not been a major issue in this project, although it is of importance in testing heavily shielded samples. The main body of developmental work lies in the appropriate deployment of the method and the optimization of the particular techniques.

The use of porcelain inside buildings provides a widely available dosemeter material for measurements in shielded locations and its glazed surface also provides the advantage of low fall-out retention when used in exterior locations. The use of brick from walls to obtain a dose-depth characteristic provides important information concerning the nature of the time-averaged incident field. New luminescence techniques, based on optical stimulation, are promising and may offer advantages in routine dose evaluations and in the evaluation of dose-depth profiles using cut brick sections [15].

The development of computational methods has provided an important adjunct to the experimental determinations enabling i) luminescence dose evaluations in ceramic to be related to reference points; ii) dose-depth profiles to be interpreted in terms of incident radiation; iii) the variation of dose with location and orientation of external samples with source configuration.

This project has enabled some of the key factors in deploying luminescence for retrospective dosimetry to be put into place and the potential for the method is clearly established in this work. An important aspect for future use of the method is in bringing it from a research base into routine use since a large number of determinations will be required for each settlement selected for study. There is much work to be done in settlements, in examining levels of resolvable transient dose, the degree of spatial variation and in seeking correlations with dose estimates based on other data such as soil contamination and monitoring, if available. The establishment of experimental capability in 'novice' laboratories within the CIS has commenced under ECP10 and their full development will be an essential part of completing this work.

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