Luminescence Analysis for Radiological and Nuclear Forensic Application

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ABSTRACT

This paper briefly discusses recombination luminescence and its use in forensic radiation dosimetry. Recombination luminescence techniques offer a new capability for radiological forensic analysis of sites and vehicles previously cleared of isotopic contamination, enabling the determination of the *prior* presence of radioactive materials. This key ability, to provide radiation exposure data after the ionising radiation sources and radioisotopes have been removed, gives luminescence a unique applicability in scenarios where it is not possible to obtain radiation dose data by any other means. Three principal areas of application to National Security and Defence have been identified:

1. *Forensic Analysis* of sites and vehicles to confirm prior containment of radiological materials, i.e. "dirty bomb" transportation, storage and assembly sites,
2. *Support to International Nuclear Weapons Inspection* initiatives, for identification of clandestine nuclear programs in now isotopically-cleansed facilities such as buildings, bunkers and other localities, and
3. *Retrospective Population Dosimetry* - quantification of absorbed dose to the population in the recovery phase after a dirty bomb or nuclear accident.

General Terms

Experimentation, Security, Legal Aspects.

Keywords

Thermoluminescence, TL, Optically Stimulated Luminescence, OSL, Environmental Dosimetry. Retrospective Population Dosimetry, Forensic, Radiological

1. INTRODUCTION

1.1 Recombination Luminescence

Ionising radiation in the environment produces electron-hole pairs; in certain crystalline materials (such as quartz grains extracted from soil or common building materials) these may become trapped in long-lived defects in the energy band gap. At a later date, application of a suitable stimulus such as heat or light can release the trapped electrons, which then diffuse in the crystal until they encounter a trapped hole, with which they then recombine accompanied by the emission of a UV/visible photon. In principle each grain of quartz (or other suitable material) constitutes a natural radiation dosimeter accruing radiation dose from both natural and artificial radiation sources.

1.2 Environmental Dosimetry and Geochronology

A family of dating methods utilising recombination luminescence has been developed since the 1960's. These are based on measurements of the natural background radiation dose-rate and the accrued dose in quartz grain, which enable calculation of the time for a suitable trap-emptying "resetting" event, such as exposure to heat. The original application of the technique was to date fired materials such as pottery and rapidly branched into art authentication and areas as diverse as food irradiation monitoring (measuring 'thermoluminescence', or TL). New variations were subsequently invented for application to unfired sedimentary materials where the resetting event was exposure to sunlight, and are rapidly becoming cornerstone techniques in various disciplines of Earth Sciences and Archaeology (often measuring 'Optically Stimulated Luminescence' or OSL). An allied technique, Thermoluminescence Dosimetry (TLD), uses highly-sensitive artificial phosphor crystals in "film badges" and is the leading technique for radiation dose measurement for nuclear industry workers and hospitals.

Radio-epidemiological studies have made use of modern fired materials such as bricks, porcelain and roof tiles to map fallout dose magnitudes and spatial distribution from events such as the Hiroshima bombing (1945) and nuclear tests in the USA and the former USSR. The European Commission (EC) has worked towards standardising the use of TL from environmentally widespread materials such as brick, tile and porcelain for retrospective nuclear accident dosimetry. This permits the mapping of dose magnitudes and spatial distributions in the event of a leakage of radiation from a nuclear facility. Samples were collected from Prypiat (Ukraine) and other locations within the Exclusion Zone surrounding the Chernobyl power plant.

It is from these established techniques that the technology and know-how for forensic applications are drawn. Luminescence

E-FORENSICS 2008, January 21-23, Adelaide, Australia

DOI 10.4108/e-forensics.2008.2853

forensic techniques are complementary to "conventional" radiation monitoring methods: the radiation damage detected by luminescence in the aftermath of exposure to ionising radiation is closely analogous to a persistent fingerprint, as it reveals the prior presence of radiation after the source has been removed. Sampling can be done at any time following removal of the source - even years later if required. For example, retrospective population dosimetry can still be performed for the Chernobyl event 20 years later to produce new exposure dose information for the affected population, and for dispersion modelling. Hence, in the aftermath of a radiological event, luminescence analysis of crystalline materials enables accurate mapping of the accrued dose in affected areas. Importantly this retrospective capability of luminescence means that there are no requirements to have trained and equipped staff or monitoring arrays deployed in the radiation field as an event unfolds. Detailed reconstruction of the radiation field is feasible even without knowledge at the time that there is an irradiation event, as will typically be the case in the forensic context.

The detection limits vary widely depending on the dose sensitivity of the material sampled, with some natural crystals such as calcium fluoride having sensitivities of the order of 10 μGy and others virtually insensitive; however typical limits are of the order of 10 mGy. This is approximately an order of magnitude more sensitive than biological methods such as measuring chromosome damage in blood lymphocytes[1].

2. DSTO PROGRAM

2.1 Program Overview

DSTO has previously set up a basic field sampling capability, with sample preparation and analysis conducted by agreement in the University of Adelaide School of Chemistry and Physics Physical Archaeometry Laboratory. It is now proposed to expand this arrangement to become a "Centre of Expertise" (CoE) under the DSTO-University of Adelaide Alliance Program, developing a world-class luminescence analysis capability. This capability will consist of a research and measurement laboratory based at the University of Adelaide, and a deployable Mobile Field Sampling Laboratory (MFSL) to be based at DSTO, enabling back-feeding of validated samples from field locations.

Research on modern man-made materials will be required as luminescence dosimetry properties have been studied for only a few such materials. Research on natural materials has been motivated by geochronological applications which typically require luminescence signal stabilities > 100,000 years, hence only the few mineral species suitable for this purpose have been comprehensively studied. In contrast, forensic applications can exploit signals with stabilities as low as hours in some circumstances, hence characterisation of the shorter-lived signals in natural materials not valid for geochronology will also be required.

Spectral analysis and signal stability measurements are key characterisation properties of new materials, and this work will be greatly assisted by the University of Adelaide luminescence spectrometer, currently the world's most sensitive apparatus of this type. Another instrument to be moved to the CoE is the developmental Australian National University Photon-Counting Imaging System[2]. This will enable investigation of dose-depth effects. Beta particles, low energy gamma rays and X-rays are stopped or strongly attenuated on the scale of 1 cm penetration depth into absorbers such as brick or concrete: this dose-depth relation is evidence of external irradiation but has been little studied due to the limitations of existing apparatus.

Thermoluminescence is preferred to OSL for accurate laboratory dose determinations, as more information is available for validation of the results, but OSL has considerable potential for rapidly screening large areas in the field. However, a constraint is limitation to the relatively small set of materials currently known to be optically-stimulable. The discussion below relates to detailed, laboratory-based measurements.

2.2 Samples and Sampling Requirements

Samples are most favourably collected from materials subjected to heating during manufacture, preferably brick, tile or porcelain, although work on unfired materials such as concrete and gyprock (drywall) is showing promise. The range of acceptable materials can be extended beyond materials proven for dating.

2.3 Sampling and Handling Protocol

Samples are collected as blocks, disaggregated material within coring cylinders, or as cores extracted by water-cooled coring drill. Exposure to light following collection must be kept to a minimum, hence each sample must be immediately wrapped in black plastic. A calibrated NaI gamma ray spectrometer can be used to determine the background gamma ray dose-rate at the position from which the sample is collected. Additional sample material can be collected to permit more detailed dose-rate analysis if required, given the potential lack of opportunity for follow-up sampling. It is essential that samples are not exposed to X-rays at any time, as X-rays will induce a potentially-misleading TL signal.

2.4 Preparation and Quantity

A series of physical and chemical steps to extract mineral grains are performed under darkroom conditions, including hydrochloric acid (HCl) treatment, sieving, density separation and hydrofluoric acid (HF) etching, while slices and powders are obtained by water-cooled diamond wafering saw or crushing and sieving as appropriate. Measurements typically require several milligrams of mineral grains, with greater quantities providing better results. Quartz is the most ubiquitous and best-established environmental dosimeter, and as the proportion of quartz will vary between materials. Thus the quantity of raw material collected in order to permit reliable analysis is sample-dependent. However, as a guide, a few hundred grams of material such as house brick, roof tile or concrete should provide ample quartz in the matrix. For porcelain, the matrix itself is measured using slices sawn in the laboratory, and quantities as low as 10 gm collected as blocks or by coring drill are acceptable.

3. ANALYSIS

Analysis involves three principal steps:

1. Measurement of the sample raw luminescence, either by TL or OSL,
2. Administering of calibrated laboratory irradiation doses and re-measurement of luminescence to enable

conversion of the raw luminescence to accrued dose, and

3. The component of natural dose accrued is calculated from approximate knowledge of the age of the item (time since the last resetting event) and measurement of the environmental radiation dose-rate pertaining at the time of sampling. This component is subtracted, and the remainder is the dose acquired from removed radiation sources previously in the vicinity of the tested item.

4. REFERENCES

[1] Schulte, E-H., Kelly, G.N. and Coiffard, K. 2002. Radio-epidemiology. Eds., European Commission, Community Research Publication: ISBN 92-894-3839-9

[2] Spooner, N.A. 2000 A photon-counting imaging system (PCIS) for luminescence applications. Radiation Measurements, 32, 513-521