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Luminescence Techniques for Dose Reconstruction in Accident Situations: Possibilities, Limitations and Uncertainties

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In a nuclear accident of even moderate size, locations will inevitably be exposed which do not have adequate monitoring. In these situations nontraditional dosimeters such as bricks, tiles or other environmental materials have historically provided measurements against which models of transport and exposure could be tested. Given sufficient speed and accuracy, the utility of TL techniques applied to natural materials can extend well beyond model verification to a variety of dosimetric applications.

Possibilities

• Short lived fission products can deliver the largest component of dose to a population, particularly if exposure begins shortly following a reactor accident. Environmental materials which record absorbed dose from the onset of exposure can provide data which field monitoring equipment placed into operation at a later time may miss. • In the case of airborne contamination under windy conditions, the major component of dose may be delivered as the contamination passes overhead leaving little residue for later detection. Bricks and tiles on the exteriors of buildings could reveal dose as a function of height from the ground given sufficient sample sensitivity. • Doses indoors are recorded in materials such as floor or decorative tiles, plumbing porcelain or tableware. Models incorporating building dependent structural shielding would be required to produce similar information. • The temporal stability of environmental dosimeters allows reassessment of doses decades after an event. If questions arise concerning the original dosimetry, as was the case in Hiroshima and Nagasaki (Ichikawa et al., 1966; Hashizume et al., 1967; Maruyama et al., 1987), areas down wind of the Nevada Test Site (Haskell et al., 1988; Haskell et al., 1994) and regions exposed to fallout from the Chernobyl accident (Hütt et al., 1993; Stoneham et al., 1993; Bailiff, 1995), then a reassessment can be undertaken using newly collected environmental materials.

Unfortunately, luminescence techniques are complex and time consuming and many factors enter into a final dose estimate. Extreme care must be taken to insure that the material being examined is suitable for analysis, that all of the factors which are required to reconstruct the accident dose

may be determined, and that all appropriate tests which may reveal problems with sample properties are performed. In situations where measurements may be used for legal purposes, the slightest omission may invalidate the findings.

Limitations

Bricks, tiles, porcelain and many other ceramic materials have the ability to store and retain information of radiation dose which they have received over long periods of time. When they are initially fired during manufacture, the past history of radiation dose is effectively zeroed and the material starts accumulating dose information once again. Archaeological materials can be dated (Aitken, 1985) using the accumulated signals which build up over time because of the relatively constant background dose from natural radiation to which they are exposed. For a sample to be dated two things must be known. 1) the total dose absorbed by the sample, and 2) the rate at which natural dose accumulates in the sample. Once known, the age is determined by dividing the total dose by the annual dose rate.

$$\text{Age (yrs)} = \text{Total Dose (Gy)} / \text{Dose Rate (Gy/yr)}$$

The same types of materials can be used for accident dosimetry since an accident dose will merely be added to the dose accumulated from natural background. In this case the age of the sample must be known in addition to the information required above.

$$\text{Accident Dose (Gy)} = \text{Total Dose (Gy)} - (\text{Age (yrs)} * \text{Dose Rate (Gy/yr)})$$

There are, of course, uncertainties associated with each of the measurements above, and the uncertainty in estimate of accident dose will be a function of those uncertainties. Errors are associated with 1) the measurement of cumulative dose itself (using TL techniques), with 2) the measurement of natural dose rate and with 3) the accurate determination of the age of the sample. As the age of a sample increases, the size of the natural dose increases and the errors associated with determination of cumulative natural dose will also increase. At some point, the accident dose will no longer be able to be distinguished from the natural background due to the size of the background dose and the increasing cumulative uncertainties.

APPENDIX 22.

Luminescence Techniques for Dose Reconstruction in Accident Situations Possibilities, Limitations and Uncertainties. (Presented at the International Workshop on Radiation Exposures by Nuclear Facilities-Evidence of the Impact on Health. Portsmouth, England July 9-12, 1996)

Minimum detectable dose.

It is important to get an indication of the minimum detectable accident dose which can be statistically distinguished above the cumulative natural background dose as a function of sample age and uncertainties associated with the measurement process (Haskell et al., 1984). Assume average conditions of: dose rate from beta + gamma radiation of 3.5 mGy per year and uncertainties in measurement of cumulative dose (using TL) and beta and gamma dose rates of ± 10% (1 sigma). And given the ages and uncertainties of ages as shown in Table 1, below, the accident dose in each case would have to exceed the values shown in italics to be statistically distinguishable from naturally accumulated background at the 2 sigma level of confidence.

Age(yrs)	±1 yr	±15 yr	±30 yr
1	*10*	*105*	*210*
5	*10*	*105*	*210*
10	*11*	*105*	*210*
20	*19*	*106*	*211*
40	*36*	*111*	*213*
80	*71*	*127*	*222*

Table 1. Minimum accident dose levels (mGy) statistically distinguishable (2 sigma) above natural background levels given dose rates and uncertainties indicated in text.

This exercise indicates that with proper analysis of materials which were new at the time of an accident, and with precise measurement of natural beta and gamma dose rate, together with an accurate estimate of the age of the samples, accident doses of less than 20 mGy could accurately be measured. Note, however, that the minimum detectable dose increases greatly with both age and uncertainty in age.

Uncertainties

The determination of background dose is considerably more complex than indicated above. All components of natural background must be independently determined so that the accident dose, D_X is now expressed as follows.

$$D_X = D_{TL} - (R_\alpha + R_\beta + R_\gamma + R_\chi)A$$

Where

D_{TL}	=	TL measurement of total accrued dose
A	=	Sample Age
R_α	=	alpha dose rate
R_β	=	beta dose rate
R_γ	=	gamma ray dose rate

R_χ = cosmic ray dose rate

In cases of high accident exposures or low natural doses due to the selection of young samples, the natural dose can often be estimated or neglected. However, for retrospective measurements involving samples manufactured decades previously, the natural component of dose may represent the largest source of error in the evaluation process. For very young samples natural dose errors are negligible and TL measurement uncertainties (5 to 10%) dominate. Older samples exhibit errors in natural dose determination dependent largely on the homogeneity of the ceramic matrix itself. Small uncertainties in natural background determination are seen for tiles and clays, whereas certain bricks containing large agglomerations of quartz extend uncertainties in beta contribution to more than 30%.

Background determination:

Dosimetry of the component of dose due to natural background sources over the lifetime of a sample can be an involved and time consuming process made more difficult in contaminated areas with residual exposure from a nuclear accident. With each method of background determination there are inherent uncertainties and uncertainties which may be introduced due to sample heterogeneity, alterations in the environment due to cleanup activities, changes in location of non stationary samples, variations in climate etc.

Gamma + Cosmic ray dosimetry

For dosimetry purposes the gamma and cosmic ray components are often grouped and determined (in uncontaminated locations) using *in situ* TL dosimeters or portable gamma-ray spectrometers. TL capsules must be of sufficient wall thickness to exclude beta rays and the effect of the thickness on low energy gammas must be considered. If the atomic number of the TL dosimeter differs from that of the natural material being examined (usually quartz) then uncertainties due to the difference in TL response to low energy gammas must determined. Seasonal variations in water content of bricks and surrounding soil may be significant in some locations requiring long exposure periods or rotation of TL capsules throughout the seasons. Cosmic ray changes due to variations in the solar cycle can also introduce error, however only in unusual situations of very low natural terrestrial radiation including gamma and internal beta contribution.

Internal beta contribution

Common techniques for determining the dose from internal beta emitters include alpha counting, flame photometry, atomic absorption spectrophotometry, beta TLD, neutron activation analysis, gamma ray spectrometry and fission track counting. These methods compared well (±7%, 1σ) in an interlaboratory comparison (Haskell, 1983) using a variety of brick samples.

For coarse ceramics such as building bricks, the single largest uncertainty can involve determination of beta dose to the grains being analyzed. This is due to the heterogeneity of the brick matrix which is often composed of other crushed brick as well as crushed gravel added to retard shrinkage during firing. This added material is sometimes composed primarily of large agglomerations of quartz which can fragment into particles in the size range used for TL measurements. Since energy absorption due to beta particle penetration within the quartz is a function of size of the grain as well as average beta energy, attenuation coefficients (Mejdahl, 1979) for betas from uranium, thorium and potassium-40 within the brick as well as the average effective grain size (Kaipa et al. 1988) should be determined for each brick.

The internal beta field is also greatly affected by proximity to the surface of a sample due to the adjoining material, be it air, mortar, soil, glaze etc. It is usual practice to remove the outer 2 to 3 mm near a sample's surface to insure uniformity of beta field as well as electron buildup from external gamma rays. The ability to obtain dose versus depth profiles near the surface of bricks and porcelain is greatly complicated by these field discontinuities. If such measurements are to be made and interpreted, then accurate measurements must be made of the nuclide content of the adjoining material and calculations of the change in the natural beta and gamma fields as a function of depth from the surface should be undertaken. Certain artificial TL dosimeters show large variations in TL sensitivity as function of cooling rate following anneal at high temperatures. A possible source of error which has not been investigated involves changes in TL properties near the surface of ceramics which may be induced by differential cooling following firing during manufacture.

The alpha component of natural dose

The alpha component of dose has been considered negligible in both the pre-dose technique (Fleming, 1973) and the high temperature technique when quartz grains are etched in HF acid prior to analysis. The effectiveness of alpha particles in inducing predose sensitivity in the fine grain matrix of porcelain has not yet been reported, and represents another potential uncertainty in accident dosimetry.

TL measurements

The TL measurement process itself introduces numerous uncertainties into the estimate of accident dose. The procedures and associated uncertainties are discussed elsewhere in these proceedings.

Conclusions

Luminescence techniques offer unique advantages in retrospective dosimetry of nuclear accidents, however a great deal of effort and training is required for proper technique application and interpretation of results. Few laboratories have the demonstrated capabilities of performing accurate dose evaluation and results from novice laboratories are rightly regarded with suspicion. The field of retrospective luminescence dosimetry has benefited from blind intercomparisons and international cooperation. A laboratory entering the field would likewise benefit from collaborative associations with established laboratories and in participating in interlaboratory comparisons which are now routinely conducted.

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