Luminescence Techniques for Dose Reconstruction in Accident Situations: Technical Aspects and Results of Application

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Thermoluminescence (TL) dosimetry of environmental materials was first associated with accident dose reconstruction during the early 1960's. Thermoluminescence techniques were applied to the reconstruction of doses at Hiroshima and Nagasaki [7,16,17,19,20] The Nevada test site [9,13], and most recently, the regions downwind of the Chernobyl Nuclear power plant accident [4,15,21]. Several reviews of retrospective techniques used in these studies detail procedures, associated problems and experimental validations which have been conducted [4,11,12]. Perhaps the most extensive effort at technique validation prior to current efforts associated with the Chernobyl accident was the TL study of Hiroshima and Nagasaki conducted as a part of DS-86[19]. Validations included tests of dose rate effects, neutron sensitivities, preparation effects and numerous intercomparisons and intercalibrations. A partially blind, multilaboratory study involving NBS-irradiated annealed quartz removed from a Nagasaki brick sample produced measurements of dose which agreed with applied values to within better than ± 10% (1s) at each of 3 dose levels ranging form 82 mGy to 417 mGy. Comparison of identical tile samples, when measured by both the high temperature and pre-dose TL techniques, produced agreement of ± 10% [3] in an intercomparison involving different tiles from the same sampling sites, the laboratories also produced dose estimates agreeing to within ± 10%. The TL results showed close agreement with theoretical calculations in the city of Nagasaki. Interlaboratory agreement was again very close at a site in Hiroshima examined by the laboratories, however the overall measurements in Hiroshima at a distance from the hypocenter of approximately 1.4 km was approximately 20% higher than theoretical calculations. Further measurements using both the pre-dose and high temperature techniques at even greater distances has tended to verify these results [14,17,20]. The reason for the discrepancy between the measured and calculated gamma ray dose values in Hiroshima remains unclear, as does an even greater discrepancy between theoretical and measured neutron fluences in that city [18,22,23].

Concurrent with the Hiroshima/Nagasaki effort was a TL evaluation of doses delivered to regions downwind of the Nevada Test Site [9,13]. This effort evaluated transient doses of less than 100 mGy and relied exclusively on the use of the predose TL technique. Because the doses of interest were low compared to natural dose accumulation, rigorous diagnostic tests [8,9,10,13] were developed and applied. The study allowed comparison of measured doses in building bricks versus evaluated exposure estimates based on soil sampling measurements, reevaluation of original monitors reports and a comprehensive review project.

The Chernobyl nuclear accident has lead to several recent applications [4,15,21] of TL techniques. Again collaborative efforts were employed allowing intercomparisons of separate techniques as well as similar

techniques applied by different laboratories.
The multi-national effort sponsored by the European Community (ECP-10) in collaboration with the Former Soviet Union, continues to examine the feasibility of retrospective techniques applied to the Chernobyl accident. The program has contributed to the validation of more recently developed techniques, has incorporated measurement into Monte Carlo models at actual exposed locations, has investigated correlation of measured dose depth profiles in ceramics with theoretical calculations and has mounted a series of studies designed to insure interlaboratory reliability [5].
Perhaps the biggest question currently facing luminescence dosimetry efforts involves optimization of methods for integrating results into modelling efforts. This problem is being addressed by the ECP-10 effort and requires close cooperation of modelers and measurers at the earliest stages of reconstruction.

Thermoluminescence

If a material which has received a prior dose of radiation gives off light as it is heated it is said to be thermoluminescent. In many crystals such as quartz or feldspars, the light which is given off during heating is proportional to the amount of radiation the sample received before heating.
Figure 1 is a glow curve displaying light output as a function of temperature for a sample of quartz given a dose of radiation shortly before heating. There are several temperature regions in the glow curve of quartz where the light output is particularly high. In general, the higher the temperature at which these peaks occur, the greater the stability of the signal. High temperature TL peaks in materials such aus quartz (such as the TL peak above 350°C in Figure 1) are stable for many thousands of years. Since natural sources of radiation constantly bombard all materials, these long lived TL peaks are useful for archaeological and in some cases geological dating techniques. Signals in the lower temperature peaks seen at 110°C and 160°C fade too rapidly to be of much use for dating or dosimetry under normal conditions. The 110°C TL peak, for instance, fades in only hours.

Requirements for Natural TL Dosimeters

For an environmental material to be useful as an accident dosimeter it must possess certain characteristics: 1) It must be sensitive to ionizing radiation at the dose levels of interest, 2) it must have been „zeroed" at a known time by thermal annealing, 3) it must have the ability to retain a record of the accident exposure with known or negligible fading over the time interval between exposure and measurement, 4) it must be capable of being prepared for measurement without the introduction of confounding signals and 5) the dose of radiation accrued by the material from „natural" sources must be measurable or accurately estimated.
The importance of natural dose is illustrated in the following formula where Dx, the transient or accident dose, is determined from measurement of the total accrued dose to the sample minus independent evaluations of sample age times ambient dose rate from α, β, γ and cosmic ray sources.

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

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

TL determination of Total Accrued Dose

In an ideal sample the lifetime of electrons in traps is governed by the following formula.

$$\tau = s^{-1} \exp(E/kT)$$

Where τ is the mean lifetime of the electron in a trap for which E is the energy required to release the electron, 's' is a constant termed the frequency factor, k is Boltzmann's constant and temperature (T) is expressed in degrees Kelvin. Depending on the energy level of the traps responsible for TL emission and the trapping parameter 's', the electrons responsible for the TL signal will exhibit greater or lesser stability over time. To be effective for dosimetry a given TL peak must be stable over the time period from exposure to analysis, or be quantifiable in terms of degree of fading. To determine the region of the glow curve free from fading with the high temperature technique, the 'plateau test' is routinely performed. This test compares glow curves of sample containing the accident dose with glow curves of the sample given an additional dose of radiation in the laboratory. By plotting the ratio of intensity of the glow curves as a function of temperature it is possible to identify the stable regions which have not undergone appreciable thermal fading. A second type of instability unrelated to thermal fading, termed anomalous fading (tunneling), often occures in samples of feldspar and zircon, but has not been identified in quartz. Assuming that anomalous fading does not affect the entire glow curve equally, the plateau test will identify regions free of this effect. This type of fading often occurs over periods of days or weeks and can be monitored in the laboratory. The safest method for insuring against anomalous fading involves laboratory irradiations of a sample and then examination of the signal over a period of weeks or months. Such tests often are adequate to detect and even correct for the effect, particularly if the accident dose in question was delivered fairly recently. In an examination of roof tiles from Hiroshima and Nagasaki in the 1960's Hashizume et al. [7] examined fading of newly irradiated samples over a seven month period confirming stability of TL regions above 200°C and verifying results of plateau tests on the same samples.

Two other factors may also influence the accuracy of high temperature measurements if not adequately considered. These include supralinear growth of TL as a function of dose and change in sensitivity of the sample as a result of laboratory heating. Supralinearity may be identified and corrected with the use of the additive dose procedure on multiple samples. The tests involve irradiation of several portions of sample with different doeses in addition to the dose being measured. The light output from the sample is plotted as a function of the added laboratory dose. Extrapolation back to the dose axis would, if supralinearity had not occurred, provide an estimate of the dose being measured. Since this test alone cannot determine whether supralinearity has occurred, the once heated samples are reirradiated with similar doeses as in the first step and the growth characteristic again plotted. Since the origin is known for this growth curve, supralinearity in the low dose regions which may not have been detectable in the field sample, are determined. The value of dose obtained from the supralinearity correction will normally circumvent problems of change in sensitization as well as supralinearity since a constant degree of sensitization will not alter the intercept value. In some conditions, particularly with samples containing feldspars, the change in sensitization is not constant, but varies with dose. The test for determining whether or not the sensitization is dose-dependent is identical to the test just described for supralinearity, with the exception that the sample used to generate the second curve is given a dose in addition to the ac-

crued dose prior to annealing and retesting. A difference of 20% between the I_N (Fig. 2) obtained with these two procedures is usually taken as indication that the supralinearity is dose-dependent. Samples showing changes in sensitization as a function of dose are best discarded. Bailiff [3] performed these measurements on quartz from tile samples from Hiroshima University, and found them to be free of dose-dependent sensitivity changes to within 6%.

The Pre-Dose Technique

The pre-dose TL technique [6] is based upon the dose-dependent increase in sensitivity in the 110°C TL peak of quartz when the sample has received a thermal treatment at approximately 500°C following application of the dose in question. This increase is proportional to the dose accumulated („pre-dose") prior to the thermal treatment. The effect is depicted in Fig. 3 where S_O is the initial sensitivity of the sample measured with a small 10 mGy test dose. S_N is the sensitivity measured (again with a 10 mGy test dose) after the sample has been heated to 500°C. A calibrating dose followed by a thermal treatment produces yet another increase in sensitivity S_{N+b}.

Accrued dose is estimated with a single portion of sample by taking the ratio of the sensitivity change due to the accrued and to the calibrating dose and multiplying by the calibrating dose b (the multiple activation procedure, MA; without quenching, Fig. 3).

A similar method (the additive dose procedure) is used for dose estimation using two aliquots of sample with the exception that the sensitivity of one of the portions is measured following thermal activation of a calibrating dose applied on top of the accrued dose prior to thermal activation.

Variations of the Pre-Dose Technique

Factors which potentially affect the degree of sensitization observed as a function of dose have led the modifications and additions to the orginal methods of analysis:

Thermal Activation Characteristic (TAC)

The TAC is a plot of the sensitivity of the 110°C TL peak of quartz as a function of temperature achieved during thermal treatment. A sample with unknown field dose is sequentially heated to higher and higher temperatures and tested for sensitivity enhancement at each step. This procedure is useful for determining temperatures to be used for later analysis. By applying an additional pre-dose to the sample following the first heating sequence and recollecting the TAC additional information may be collected concerning ambient sensitization of the sample as well as changes in degree of sensitization resulting from the thermal treatment. The two can be distinguished by yet another TAC involving a pre-dose application in addition to the original unknown dose [10].

Radiation Quenching Correction

Radiation quenching (decreased sensitivity following application of a laboratory dose prior to thermal treatment) ist attributed to L center deactivation resulting from application of the calibrating dose. The correction [1] involves measurement of the sensitivity of the sample following application of the calibrating dose but prior to thermal activation. This sensitivity level $S_{N'}$ is taken as the new base line against which the subsequent increase in sensititivy is measured. This correction is now a routine part of multiple activation analysis.

Additive Dose Pre-dose Analysis

This method involves measuring the sensitivity increase due to the accrued dose on one portion of sample, and comparing this to the increase in sensitivity due to the accrued dose plus a calibrating dose applied to a second portion of sample. Normalizing by weight, or initial sensitivity allows an

estimate of accrued dose unaffected by thermal stress to be obtained. Inconsistencies and problems with normalization unfortunately render this method the least precise of the pre-dose methods.

The Modified Additive Dose Procedure (Pre-dose)

The modified additive dose procedure for pre-dose analysis is similar to the high temperature additive dose procedure in that a pre-dose is added in addition to the unknown dose prior to analysis (Fig. 4), [2,8]. The sample is then analyzed using the multiple activation procedure (Fig. 5, with quenching correction). Additional samples are analyzed in this manner but with increasing pre-doses. A plot is made of estimated dose obtained from pre-dosed samples versus the amount of laboratory pre-dose. Extrapolating through the data points back to the applied dose axis provides an estimate of accrued dose independent of sensitivity change. The intercept on the MA evaluated dose axis should correspond to the dose estimate obtained with the multiple activation procedure alone.

Modified Additive Dose with Anneal

A recent addition to the modified additive dose procedure involves a high temperature (>700°C for 1 minute) anneal of the sample following each sensitization [10]. This returns the sensitivity to near baseline values avoiding nonlinearities which may otherwise be encountered. The procedure also has the tendency to maximize thermally induced changes in degree of sensitization per unit dose allowing the effect to be clearly observed and quantified.

Ultra Violet Reversal:

A simple test to determine if the sensitivity of a sample has been increased due to ambient temperatures or fires involves a phenomenon initially reported by Thompson (1970). This effect involves a reversal of sensitization to near initial levels by stimulating an already sensitized sample with UV irradiation (255 nm). By testing an unanalyzed sample in this manner and comparing the initial sensitivity with the sensitivity following UV treatment, ambient activation can be detected.

Potential errors associated with predose measurements

The predose technique is very sensitive at low doses of radiation, but it has the drawback that the relationship of sensitivity increase to applied dose is nonlinear as saturation is approached. A saturation effect of some type is seen in all forms of thermoluminescence. What complicates predose analysis is that saturation effects can occur at relatively low doses and at several stages in the predose sensitization process. Saturation at each stage behaves differently, has a different effect on the dose measurement, and must be tested in different ways. Because the predose phenomenon is a multistep process, saturation effects can best be understood by following the steps in sequence. The first step in the process involves ionization during irradiation causing electrons to be trapped in electron traps and holes to be trapped in hole traps. Some of these hole traps are termed reservoir centers in the predose model. For dosimetry purposes the filling of reservoir centers should be proportional to the applied dose. As the reservoir centers approach saturation, however, there are fewer empty centers available to capture holes and the number captured slows dramatically. Nonlinear filling results and transfer of holes to reservoir centers during irradiation is no longer proportional to dose. This effect is termed „R" center nonlinearity.

The second step in the predose process at which saturation effects can occur concerns the transfer of holes from the reservoir centers to the inactive luminescence centers during thermal activation. For accurate do-

simetry this process should involve transfer of holes to luminescence centers in proportion to holes released from reservoir centers during thermal activation. However as more and more luminescence centers become activated, the number of inactive centers available to receive holes from reservoir centers is reduced and filling is no longer proportional to holes released from the reservoir centers. Dose estimation is again compromised as a result. This effect is termed „L" center nonlinearity.

Another phenomenon which effects dose estimation is a reduction in the number of active luminescence centers which occurs during laboratory irradiation. This effect, termed radiation quenching which was mentioned above does not result in a decrease of the original sensitivity of the sample (S_0) but it does reduce the sensitivity of samples which have already been thermally sensitized (such aus S_n or $S_n+\beta$). The phenomenon is observed as a decrease in sensitivity of the 110°C TL region seen immediately after application of the calibrating dose of radiation and is the reason that measurement of the sensitivity of a sample must be made following application of the calibrating dose before thermal activation. Failure to account for radiation quenching can result in an overestimate of the unknown dose.

A fourth effect which must be considered in all types of predose analysis, and which is impossible to detect without proper examination, is a change in degree of sensitization which occurs between the first thermal activation and all subsequent activations. This means that the unknown dose of radiation might produce an increase in sensitivity which is considerably more or less than the sensitivity increase which will be induced by the same dose applied after the first thermal activation. This effect is shown in Figure 5 for the case of underestimate of dose, although both cases have been overserved.

The four effects described above, R center nonlinearity, L center nonlinearity, radiation quenching and change in degree of sensitization do not act in isolation, but effect subsequent stages of the predose phenomenon. For instance a change in degree of sensitization following first thermal activation will mean that the proportionality relationship assumed in equation 2 above is inaccurate. R center nonlinearity means that filling of the L centers will likewise not be proportional to dose, even if transfer from R centers to L centers is linear. Radiation quenching can have a large effect on L center filling as well. Assume that the L centers are nearly full so that only a small increase in L filling results during thermal transfer of holes from R to L centers. If a large calibrating dose of radiation is applied, radiation quenching will result in a partial emptying of L centers and may push the linearity of subsequent R to L center transfers back into the linear region. The interaction of these effects is complex and difficult to detect, however they may be seen at any dose level. Each sample analyzed must be checked for their occurrence since factors as diverse as impurity content of the clay and heating and cooling rates during manufacture can effect them.

One of the greatest hazards of predose analysis is that the effects described above are not easy to detect and are not easy to correct for. A sample may appear to behave in a consistent, linear fashion, but may still experience nonlinearity or change in degree of sensitization which will effect dose estimates. Because of the potential for substantial, yet unobvious, errors in predose measurement, procedures have been devised to detect, and in some cases correct for the errors. Unfortunately the procedures are time consuming, and are best performed on equipment capable of performing rapid, repeated irradiations and glow curve mea-

surements. Because of this they are omitted in many laboratories, and the results obtained in their absence must be considered tentative until the effects have been demonstrated to be absent.

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Figure 1

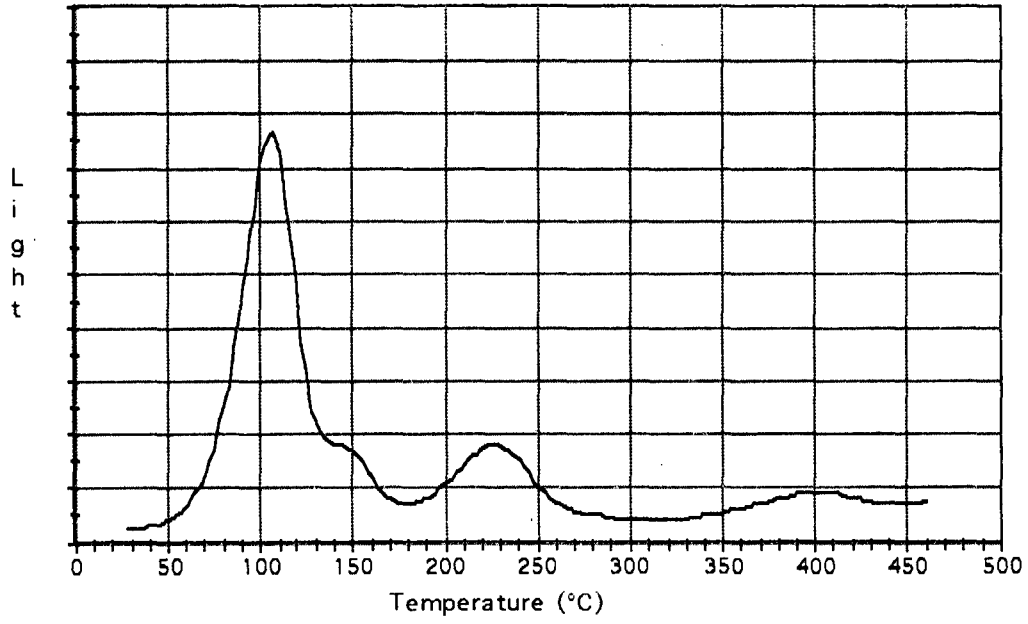

Figure 2

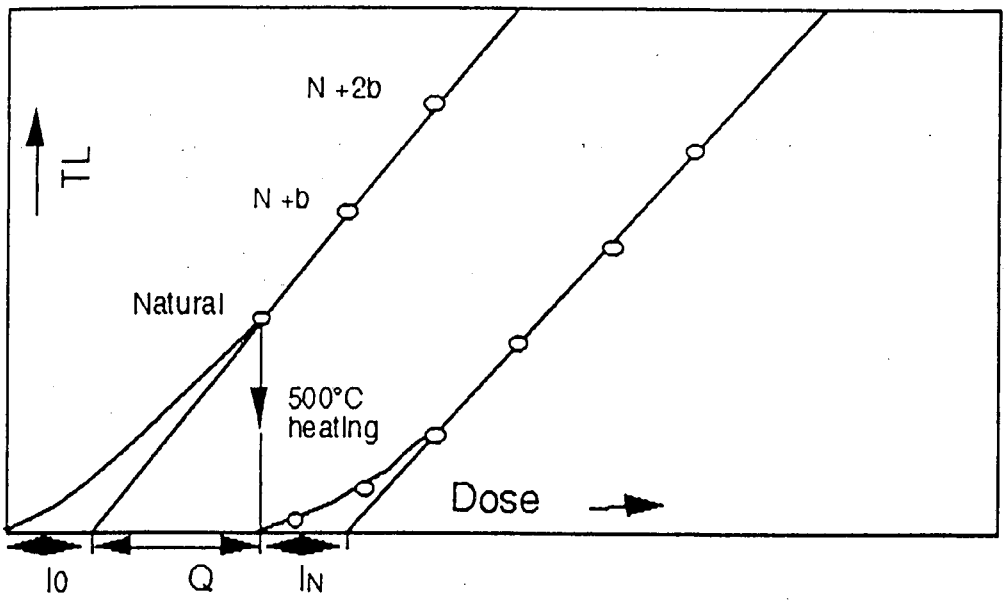

Figure 3

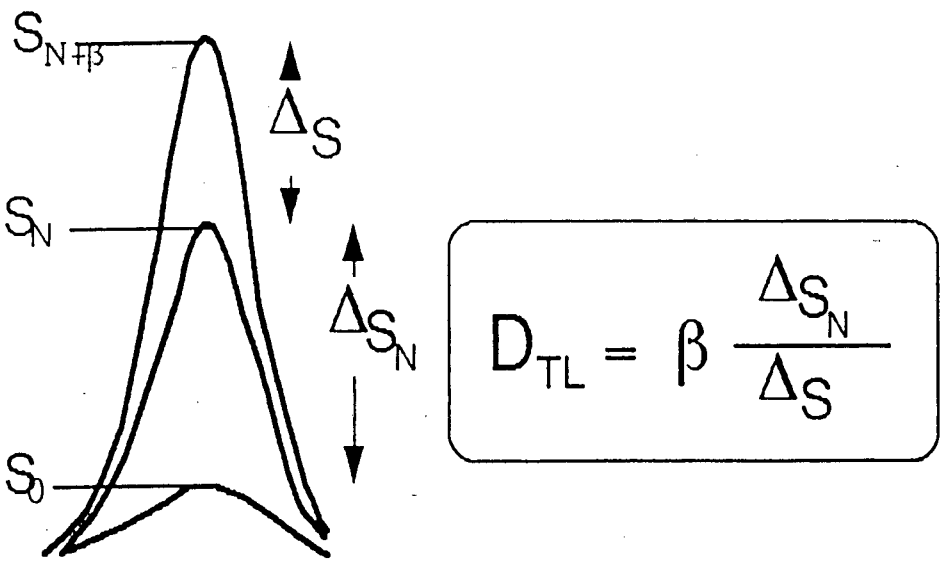

Figure 4

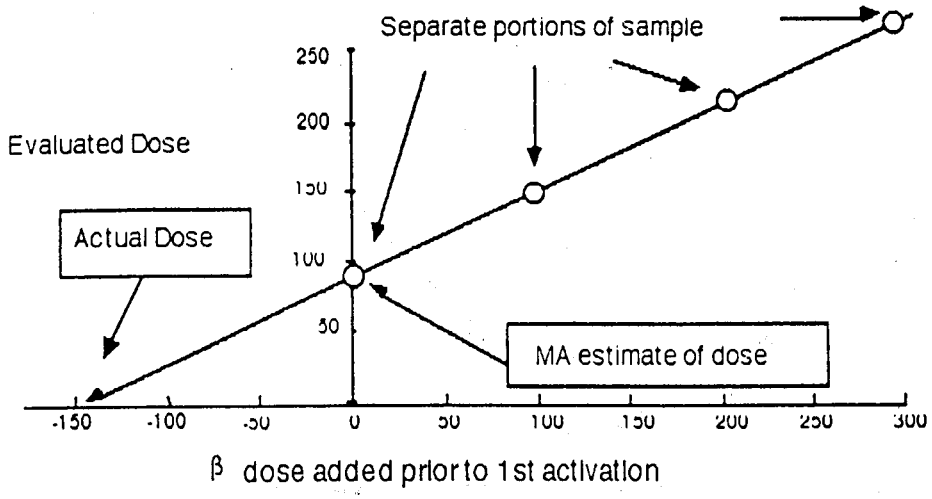

Figure 5