Thermoluminescence Radiation Dosimetry in Space: A Critique of Current Practise and Future Perspectives

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1. Introduction
Radiation dosimetry in space presents one of the few remaining challenges in the field of radiation protection. This arises mainly from the highly complex nature of the radiation fields encountered in space leading to serious problems in the interpretation of data recorded by dosimetric devices, e.g., there are currently large uncertainties in the estimation of the neutron and HZE contribution to the total dose equivalent [1]. The current approach has been to use a number of passive detectors, each capable in principle of measuring a different component of the radiation field - certainly a heroic task. LiF-TLDs are the most commonly used passive detector, however, these are said to provide no LET information [1] (not true), and tend to underestimate the total dose due to a decreasing TL efficiency with increasing ionisation density (LET). Attempts have recently been made to convert the heavy charged particle and neutron induced component of LiF-TL signals to radiation dose equivalent [2] but these are almost surely/inevitably very inaccurate [3,4].

To overcome these difficulties we introduce the concept of using existing molecular nanostructures in TL solid state materials as solid-state Q/RBE equivalent nanodosimeters. The concept is based on mimicking radiobiology (specifically the ionization density dependence of double strand breaks in DNA) by using the similar ionization density dependence of simultaneous electron-hole capture in spatially correlated TC/LC pairs (of approximately the same dimensions as the DNA molecule) in the thermoluminescence of LiF:Mg,Ti. This simultaneous electron-hole capture has been shown to lead to localized/geminate recombination and to an ionization density dependence in the relative intensity of peak 5a to peak 5 of \([5a/5] H/L = 4.25 \pm 0.6 \) (1 SD) similar to the ratio of DSBs to SSBs for low energy He ions. Hence the peak 5a Q-DSB nanodosimeter. Such a nanodosimeter, when properly calibrated could yield an estimate of the total dose equivalent combining all the components of the radiation field in a single measurement without any concurrent knowledge of the space radiation field. Much work, however remains to be done and success is not guaranteed.

The effects of ionising radiation at the molecular level are fundamental importance to human survival/health/development/genetic stability. Radiation sensitive targets in the human/mammalian cell are concentrated in the cellular nucleus and, in particular, the deleterious effects of ionizing radiation are
known to arise from radiation damage to the DNA molecule [5]. Chromosome breaks arise from double-strand breaks of DNA (DSBs) which in turn are due to pairs of single-strand breaks (SSBs) - usually when these are produced in close proximity. Indeed the degree of irreversibility and/or the degree of function lethality may be correlated with the distance between the SSBs leading to the DSB. It has been "guess-estimated" that DSBs arise from SSBs in DNA whenever the SSBs are within approximately 10 base pairs (i.e., 3.4 nm) [6]. It is, therefore, now generally accepted that energy concentrations and the microscopic spatial distribution of "energy transfer points" in volumes of linear dimensions of less than ten nanometers are of critical significance in DNA damage [6]. This, and the consequent inappropriateness of concepts based on "track-averaged" quantities such as Linear Energy Transfer (LET) as a measure of DNA radiation damage, has led to an increasingly intensive search for a solid-state radiation device of nanometer dimensions [7] (the general shape of the DNA molecule being often approximated by a cylinder of approximately 2 nm diameter) [8], (Fig 1).

A solid-state, quasi-tissue-equivalent, Q-nanodosimeter should, therefore, incorporate/fulfill the following criteria: 
(i) respond to all types of radiation and be measurable with adequate precision. 
(ii) be of dimensions comparable to the dimensions of DNA molecule. 
(iii) simulate (or be correlatable with) the ionization density dependence of certain radiobiological end effects and/or the LET dependence of the quality factor (Q) used in Radiation Protection for neutrons and heavy charged particles [9]. 
(iv) simulate the ionization-density dependence due to the proximity of two "energy transfer points" arising from highly localized energy deposition, similar to DNA double strand breaks (DSBs) which are believed to be responsible for chromosome damage leading to many irreversible radiobiological effects. 
(v) be tuneable, i.e., there should be a possibility of regulating/changing the ionization density dependence in order to mimic the ionization density dependence of various radiobiological end-points. 
(vi) for practical and economic reasons - be a part of already existing (and in use) dosimetric systems.

3. The Unique Ionisation Density Dependence of Glow Peak 5a.
Glow peaks 4 and 5 are the main dosimetry peaks used in LiF thermoluminescence dosimetry. Although these peaks have been intensively studied for several decades, it has only been recently illustrated [3] that peak 5 is actually a composite structure consisting of at least three glow peaks labelled 5a, 5 and 5b (Fig 2). The presence of these additional satellite peaks was established by detailed $T_m - T_{stop}$ studies using 3MeV He ions (high ionisation density radiation) following 400°C pre-irradiation annealing in both
air and dry N₂ gas. The Tm - Tstop analysis is a powerful but very labor intensive method of glow peak resolution used in the deconvolution of complex glow curves. Every plateau in the staircase-like pattern indicates an independent glow peak (Fig 3). The electron trapping centers (TCs) and the hole trapping recombination luminescence centers (LCs) responsible for the dosimetric glow peaks in LiF:Mg,Ti are spatially correlated, i.e., loosely coupled together by a long-range interaction into a TC/LC molecular complex. As the distance between the trapped electron in the TC and trapped hole in the LC decreases, a localized, recombination mechanism, termed "geminate recombination" enters into play. This type of luminescent recombination occurs directly via a quantum-mechanical tunneling process or via short-range, quasi-localized migration in the conduction band between nearest neighbor TCs and LCs. This process gives rise to peak 5a, which is the conceptual equivalent of the DNA DSB, since it arises from two localized energy transfer events. It is expected that geminate recombination would be of much higher probability in the He ion track due to the high dose levels in the track leading to a higher probability of simultaneous electron-hole population of the nearest-neighbor (TC/LC) correlated entity. The TCs and LCs are fully populated out to a radial distance of approximately 20 nm from He ion track axis - thus ensuring maximum efficiency in creation of peak 5a. In the 3 MeV He ion induced glow curves the relative intensity of peak 5a to peak 5 is [5a/5]ₜ = 0.34±0.04 (1 SD). However, its relative intensity in gamma induced glow curves (low ionisation density irradiation) [5a/5]₁ = 0.08±0.008 (1 SD). The double peak intensity ratio [5a/5]ₜ/₁ = 4.25±06 (1 SD) for 1MeV/amu He ions is thus a measure of the effect of ionisation density on the relative number of "double-energy-transfer-events" to "single-energy-transfer-events" in a nanometric sized volume and is therefore both conceptually and physically similar to the effect of ionisation density of the ratio of DSBs to SSBs, i.e., is therefore conceptually equivalent to RBE or alternately Q when and if biological end-point is known to arise from DNA DSB radiation damage.

The viability of the concept of using existing molecular nanostructures in TL solid state materials as solid-state Q/RBE equivalent nanodosimeters has been demonstrated. The concept is based on mimicking radiobiology (specifically the ionization density dependence of double strand breaks in DNA) by using the similar ionization density dependence of simultaneous electron-hole capture in spatially correlated TC/LC pairs (of approximately the same dimensions as the DNA molecule) in the thermoluminescence of LiF:Mg,Ti. This simultaneous electron-hole capture has been conclusively shown to lead to localized/geminate recombination and to an ionization density dependence in the relative intensity of peak 5a to peak 5 of [5a/5]ₜ/₁ = 4.25±06 (1 SD) similar to the ratio of DSBs to SSBs for low energy He ions. Hence the peak 5a Q-DSB nanodosimeter. What remains to be carried out is the establishment of an empirical correlation between the ratio of peak 5a to peak 5 in a variety of mixed neutron/gamma radiation fields and/or heavy charged particle radiation fields of known RBE. This will then
lead to the measurement of Q and/or RBE in any radiation field without any need for characterization of the radiation interaction parameters of the radiation field.

![Image](image_url)

**Fig 2 (Left)**- Deconvoluted glow curve TLD-100 following irradiation by 6.8 MeV He ions.

**Fig 3 (right)**- The results of $T_m-T_{stop}$ measurements following He ion irradiation. Of special interest is peak 5a, preferentially populated following heavy charged particle, high ionisation density, irradiation.

**References**


