AN EVALUATION OF THERMALLY STIMULATED SOLID STATE RADIATION DOSIMETERS FOR MEDICAL APPLICATIONS

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Prepared in partial fulfilment of the requirements for the degree of Doctor of Philosophy
Solid state radiation dosimetry systems depending upon the thermal release of stored radiation energy may be based on observation of thermoluminescence (TL), thermally stimulated exo-electron emission (TSEE) or thermally stimulated currents (TSC). The thesis begins with a brief review of the physics of solid state integrating dosimeters, and each of these three systems is then described and assessed. Following this an account is given of problems associated with the introduction of a routine TLD personnel monitoring service in a group of Scottish hospitals. Research into the design of suitable readout systems for TSEE and TSC dosimetry, and into the extent to which such systems are likely to be of practical value in the near future, is then described. The next section shows how observations of all three phenomena can contribute to an understanding of the physical processes which lead to the supralinear response of some dosimetry phosphors. Finally, investigations into a series of small problems arising in routine work in the National Health Service which have been solved by use of TL or TSEE dosimeters are reported.

Throughout the thesis particular attention is paid to the associated instrumental problems, and detailed accounts are given of the TL, TSEE and TSC dosimeter readers designed for the various studies reported.
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Interest in the field started with the very promising reports of early work in TSEE dosimetry originating from Becker's group at Oak Ridge (e.g. Becker and Robinson, Health Physics 15 463 1968). At this time Mr F.H. Attix of the U.S. Naval Research Laboratories was spending a sabattical year working with the Health Physics & Medical Division at AERE Harwell which he used for developing an exo-electron reader based on a pure methane gas flow proportional counter. By courtesy of Mr D.H. Peirson who was responsible for this work at Harwell, and of Mr J. Bor who was Head of the Physics Department at the North East London Polytechnic where I was then a member of the lecturing staff, arrangements were made for me to start my research in the field by spending two days each week at Harwell evaluating the potentialities of this equipment after Attix's return to the U.S.A. My decision to try and use this work to form the basis of a part time PhD and my registration at Surrey University, arranged through the good offices of Miss B. Stern, followed soon after. This quickly led both to the development of a modified form of TSEE reader in my own department at the Polytechnic and to early experiments on the use of thin film evaporated layer TSEE dosimeter elements.

In October 1971 I took up the post of Radiation Protection Officer in the Department of Medical Physics at the University of Aberdeen. Professor Mallard showed interest in the research project and kindly arranged for the transfer of my reader so that work could be continued in his department. Unfortunately although the reader was quickly set up at Aberdeen there were no facilities for the production of vacuum deposited dosimeters. I decided that successful prosecution of the work made the construction of an evaporation unit essential and started purchasing the necessary components as and when the budget allocated to my section permitted. As a result little work was done with the TSEE reader during my...
first two years at Aberdeen. Instead during my first eighteen months there I had to put a great deal of personal effort and energy into establishing a reliable TLD dosimetry service which was needed for the routine work of my section. This inevitably led to a broader study of energy storage processes in solid state dosimeters and to a decision, taken in conjunction with my supervisor, to broaden the basis of my proposed thesis by attempting to make a comparative assessment of the relative advantages of TL, TSEE and TSC as processes of dosimetric interest.

The TSEE reader was therefore modified so that it could be adapted for studying the TSC process and it was used for this purpose until the construction of the vacuum deposition unit had been completed. At this stage TSC dosimetry was felt to be of less interest than the thin film TSEE process and was therefore discontinued. The TSC results reported here have therefore already been overtaken by the introduction of more promising procedures in other centres. For completeness these have been very briefly reviewed.

The bulk of the experimental work reported was completed by the end of 1974 but the penultimate chapter incorporates reports on some practical applications of the fundamental work which have been carried out more recently and which serve to show the variety of ways in which solid state dosimetry now contributes to the field of medical physics.
CHAPTER 1 : A REVIEW OF RELEVANT ASPECTS OF SOLID STATE DOSIMETRY SYSTEMS

1.1 • Introduction

From the time of the discovery of ionizing radiations at the end of the last century both gas ionization detectors and solid state detectors (in the form of photographic films) have been in routine use in almost every hospital and radiation physics laboratory. During this period gas detectors have been continuously developed and improved, particularly for applications involving quantitative radiation dosimetry. In contrast real progress in solid state dosimetry remained almost non-existent until the detailed studies of thermoluminescence which were initiated by Daniels and pioneered by Cameron's group at Wisconsin in the early 1960's. Despite a great deal of work carried out by this and other groups the technique has only gained very gradual acceptance and it is still used on a far more limited scale than photographic film. The work described in this thesis was carried out by the author during attempts to introduce thermoluminescence and other allied dosimetry techniques into routine use in a U.K. National Health Service hospital.

Solid state detectors have three basic advantages for practical radiation dosimetry with X-rays or gamma-rays. 
(a) Relative to gas detectors their high density allows efficient detection of radiation with very compact elements which are of the order of 20,000 times smaller than an air ionization chamber of comparable efficiency.
(b) Solid detectors do not in general require encapsulation and consequently suffer less from boundary effects inseparable from gaseous detectors.
(c) Many solids have a characteristic crystal lattice structure which may allow the possibility of some of the radiation energy being semi-permanently stored in the crystal lattice: this enables the detector concerned to be used as an integrating dosimeter. The traditional film badge where some of the radiation energy absorbed produces a latent
photographic image, is probably the most widely used example of this class of dosimeter but many others are theoretically possible and a number of them have been intensively studied during the past decade.

Table 1, based on a classification originally introduced by Schulman (1) shows some of the possible interactions which could be used for dosimetric purposes together with the readout mechanism which would need to be employed in each case. The studies reported in this thesis are restricted to those for which evaluation of the integrated dose is carried out by heating the detector. Since heating is essentially an annealing process which tends to restore the crystal to its pre-irradiation condition, dosimeters using thermally stimulated readout have the inherent limitation that in general only one evaluation of the radiation energy absorbed can be made – subsequent checks to detect and correct a faulty readout are not usually possible.

Another fundamental limitation is that only a very small proportion of the energy incident on the crystal (of the order of 0.1% for thermoluminescence dosimeters) is stored for release in the subsequent readout process. It follows that if, for any reason, this overall efficiency is not always the same dosimeters may be produced with large variations in their measured sensitivity. Similar effects may also be observed if readout, cleaning, annealing or any other process to which the detector is submitted modifies its behaviour.

Despite these limitations one method of dosimetry in this category, thermoluminescence dosimetry or TLD, is now very widely used and has been chosen for dosimetric measurements in numerous projects carried out in recent years. Two related techniques, thermally stimulated exo-electron emission or TSEE and thermally stimulated conductivity or TSC, have been the subject of much recent research and TSEE in particular possesses some novel and potentially useful features. The work reported in this thesis includes measurements of all these three phenomena. Some of the
physical processes which take place in the crystal may be elucidated by intercomparison of the results so obtained. This is considered further in chapter 5.

1.2 Essential Concepts of Solid State Physics

Solid state dosimeters of the type considered in this thesis are insulators which possess an energy band structure similar to that of semiconductors. Many of them are alkali halides with a cubic lattice structure formed by two interpenetrating simple cubic lattices associated respectively with positively charged alkali ions and negatively charged halogen ions. Major breaks in the regular crystal lattice are referred to as dislocations, local ones are termed defects. These are classified either as intrinsic defects if associated with empty lattice sites or with ions displaced from normal lattice sites, or as impurity (activator) defects if associated with the presence of an ion alien to the normal crystal structure.

In the perfect crystal all valency electrons are common to the entire lattice so that the energy levels of the individual ions merge to produce a range of permissible but normally completely filled energy levels known as the 'valence band'. Similarly unfilled energy levels merge to provide an alternative range of permissible energies known as the 'conduction band', this in insulators remains empty except at greatly elevated temperatures. No electrons can have energies coming within the forbidden range between the conduction and valence bands and the electrical conductivity is consequently zero. Semiconductors have a similar energy level structure but the forbidden band is small so that even at room temperature a few electrons gain enough thermal energy to 'jump the gap' from the valence band to the conduction band leading to measurable intrinsic conductivity which rises rapidly with increasing temperature. A more important contribution to conductivity normally results from controlled doping of the crystal with a few parts per million of donor or acceptor impurity atoms which occupy normal lattice sites. The former have an excess
electron that is readily transferred to the conduction band without any associated vacancy in the valence band so leading to electron conduction, whilst the latter have one less electron than atoms normally occupying the corresponding lattice site with the result that the valence band is not completely full and hole conduction becomes possible. (This contrasts with intrinsic conductivity which is inherently due to contributions from equal numbers of electrons and holes).

The solid state dosimeters of interest in this thesis do not normally show either intrinsic or impurity conductivity. Instead localised energy levels are formed in the 'forbidden' range usually close to either the valence or the conduction band. These respectively constitute hole or electron traps in which a trapped charge carrier is metastable. On heating the crystal strongly trapped electrons and holes can be freed from the traps so that thermally stimulated conductivity takes place within the crystal. There is also a possibility of the generation of luminescence when electron-hole recombination takes place at suitable luminescence centres, or of the complete escape of some of the electrons which were trapped very close to the surface of the crystal.

In the alkali halides typical lattice defects leading to trap formation are ion vacancies and interstitial ions. Halogen ion vacancies and interstitial alkali ions form regions of localised positive charge whilst alkali ion vacancies and interstitial halogen ions form regions of localised negative charge. Their presence is usually indicated by changes in the optical emission or absorption spectrum for the crystal. The first of these to be extensively studied were the Farbzentren or F-centres (colour centres) associated with localised positive charges resulting from a halogen ion vacancy in the lattice. Free electrons can become attached to the local positive charges which show a system of energy levels similar to those of the hydrogen atom. The excited states of these trapped electrons may then be populated by the selective absorption of
electromagnetic radiation in or close to the optical region, (Figure 1.1). This model of the F-centre was first proposed by De Boer (2) who showed that such centres were readily produced by heating the crystal in an alkali vapour to upset the stoichiometric balance between alkali and halogen ions.

Other types of electron traps are associated with more complex lattice defects and different trap depths. Examples frequently referred to in the literature are F', M, R₁, R₂, N and 0 centres, (Figure 1.2). Much research effort has been expended in studying the crystal lattice structure associated with each of these. Similarly a range of hole traps have also been classified and studied. In general these lead to shorter wavelength absorption bands than the electron traps - usually in the UV spectrum. Structurally the analogy with electron traps is not complete. A stoichiometric excess of halogen is typically associated with interstitial halogen ions and results in what are known as V centres. V₁, V₂, V₃, V₄ and V₅ centres have been identified and studied in alkali halides. Irradiation does not only populate existing traps but may also produce pairs of new electron and hole traps, these are usually F and H centres which may later recombine to restore a perfect lattice. The H centre results from a biatomic halogen molecule replacing a monatomic ion in the lattice (a 'crowdion' type defect) and is therefore not an antimorph of the F centre associated with the resulting halogen ion vacancy. The H centre energy levels come a little above the top of the valence band whilst the F centre levels are similarly a little below the bottom of the conduction band. If the energy gap between these levels and their associated band is large enough the colour centres form metastable traps which lead to the crystal exhibiting the property known as thermoluminescence; the greater the trap depth the higher the temperature which must be attained during readout before the electron or hole is released from the trap concerned.

Both with alkali halides and other practical forms of
solid state dosimeters experiments quickly show that those dosimeters produced from ultrapure crystals are usually less sensitive than ones containing appropriate trace impurities. In particular in the case of thermoluminescent dosimeters not only are the luminescence centres on which the process depends usually associated with impurity atoms in the lattice but also impurities may make major contributions to the lattice defects associated with the electron and hole traps on which the process depends. Consequently groups of traps with a number of different trap depths can be found in the same sample if several different types of intrinsic and impurity lattice defect coexist. On heating the sample charge carriers will then be released from the different types of trap sequentially, shallow traps being depopulated at a lower temperature than deep ones. The resulting graph of luminescent output against sample temperature (referred to as a 'glow curve') exhibits a series of different peaks which can be correlated with the various traps, (Figure 1.3).

For thermoluminescence dosimetry there must be a known, preferably linear, relationship between the radiation dose absorbed and the light emitted by the sample on heating. The latter in turn is proportional to the area under the glow curve so long as the sample has been heated at a uniform rate. In the case of non-linear heating a curve of light output against time and not temperature must be used. For a given absorbed radiation dose the glow curve will be characteristic of the sample material and the heating rate but experience shows that, at least to a first approximation, the area under it (or total light output emitted) is independent of the rate of heating.

The first attempt to develop an equation for the emitted light intensity from a phosphor was made by Randall & Wilkins (3). These workers assumed that:

(a) There is only one kind of electron trap corresponding to an energy level at a depth E below the bottom of the conduction band.

(b) The release of trapped electrons at any given temperature
follows first order kinetics; generally (by analogy with chemical kinetics) termed monomolecular. This means that the trapped electron density \( n \) decreases exponentially with time i.e.:

\[
\frac{dn}{n} = -p \, dt
\]

where \( p \) is a constant representing the probability of escape of an individual electron in unit time.

It implies physically either that the trap density is very much lower than the density of available recombination centres or that an electron escaping from a trap does not enter the conduction band and can only undergo luminescent recombination by returning to its ground state in the lattice centre where it was trapped. (The alternative assumption of second order or bimolecular kinetics corresponds with the physical case where the total number of available recombination centres matches the total number of trapped electrons, and is usually more valid as far as the glow curves associated with most dosimetric materials are concerned).

(c) The phosphor is heated at a constant rate and the light output depends directly upon the rate of emission of electrons from traps.

With these assumptions the theory is developed as follows:

The probability constant \( p \) above is proportional to \( \exp(-E/kT) \) where \( E \) is the trap depth, \( k \) Boltzmann's constant and \( T \) the absolute temperature. The constant of proportionality is known as the frequency factor and is theoretically given by the product of the actual frequency of oscillation and the reflection coefficient, but in practice is better regarded as a measurable parameter lower than the true lattice vibrational frequency and not readily identified with an actual physical oscillation. Since \( p \) is a function of temperature its value at a specific temperature \( T \) will be denoted by \( p_T \).

The instantaneous light intensity emitted at time \( t \) will be proportional to the rate of release of electrons from traps and will therefore be given by an expression of
the form;

\[ I_t (\propto -\frac{dn}{dt}) = \text{const.} \times n_p T \quad \text{for first order kinetics.} \]

\[ = \text{const.} \times n_s \exp(-E/kT) \]

If the phosphor is heated at a constant rate \( r = \frac{dT}{dt} \), \( n_t \), the number of traps still filled at time \( t \) and temperature \( T \), will be related to \( n_0 \), the initial number by an expression of the form:

\[ n_t = n_0 \exp \int_{T_0}^{T} \left( -\frac{p_T}{r} \right) dT \]

Hence \( I_t \propto p_T n_0 \exp \int_{T_0}^{T} \left( -\frac{p_T}{r} \right) dT \)

\[ I_t \propto n_0 s \exp(-E/kT) \exp \int_{T_0}^{T} \left( -\frac{s}{r} \right) \exp(-E/kT) dT \]

Figure 1.4 shows a set of curves plotted from this equation for various values of \( E \) and \( s \). These curves form glow curves with a single peak at a temperature characteristic of the trap depth \( E \) and the area below the curve is proportional to \( n_0 \) (which is related in turn to the total radiation dose absorbed by the phosphor). In general only very low temperature peaks (such as the 147°K peak in lithium fluoride) and crystals with relatively simple structure show this type of behaviour which is characterised by a very rapid fall off in the light intensity at temperatures above the peak of the glow curve.

For the second order model the rate equation for the luminescent recombination of electrons released from traps becomes;

\[ \frac{dn}{n^2} = -p_T n dt \]

This means that the density of trapped electrons no longer decreases exponentially as we now have;

\[ \frac{dn}{n^2} = -p_T dt \]

or \( n_c = n_0 / (1 + n_0 p_T t) \) (A hyperbolic relationship)

and \( I_t \propto -\frac{dn}{dt} \propto p_T n_c^2 \propto p_T n_0^2 / (1 + n_0 p_T t)^2 \)
Garlick and Gibson (4) derived an alternative glow curve equation in a similar way on the basis of these second order kinetics. This also for the first time allowed for the possibility of retrapping of electrons following their thermal release. A retrapping factor $R$ can be defined as equal to the ratio of the coefficients of retrapping and recombination, and in Garlick & Gibson's analysis this was assumed to be unity. The resulting glow curve equation corresponds to a situation where retrapping delays the thermal release of electrons so that a larger proportion of the light is emitted at temperatures above the peak temperature and in contradistinction to the Randall & Wilkins model, leads to an almost symmetrical glow curve which is much more characteristic of most practical thermoluminescent phosphors. A third model based on the assumption that $R$ is large (i.e. that retrapping is very much more likely than luminescent recombination) was developed mathematically by Haering and Adam (5) who showed that it would lead to a very skew shaped glow curve with a long 'tail' on the high temperature side of the peak. However this is a shape not commonly encountered in practice.

Figure 1.5 shows the calculated shapes of glow curves to be expected from each of the three preceding models. Intermediate cases were studied by Kemney et al (6) who developed a computer programme for the general solution of glow curve equations by numerical integration. This can be applied equally readily to all values of $R$. Other analyses, particularly by Schön (7) and Eränkö (8)(9), have developed general treatments of the glow curve equation which incorporate the particular solutions discussed above as special cases.

The theoretical models of the glow curve discussed above have obvious limitations. Instead of the ideal situation where only one type of trap of precisely defined depth exists, the typical thermoluminescent phosphor will possess a number of different types of centre corresponding to varying trap depths and leading to a multi-peak glow curve. Furthermore the traps associated with each type of
centre will show a spread of depths - in particular centres close to the free surface of the crystal or to major dislocations of the lattice will have their trap depth modified by the lattice configuration. More sophisticated theoretical treatments can to some extent allow for these effects. For example Broser (10) has treated systems with a quasi-continuous band of trap depths and has been very successful in explaining the properties of some photoconductors such as cadmium sulphide.

It is important to note that theoretical studies of glow curves are not also directly applicable to the corresponding curves representing thermally stimulated currents in the sample because the lifetime of the electrons in the conduction band (which governs their total contribution to the observed TSC) is a function of the free electron concentration.

One of the most important applications of a glow curve equation is to enable the trap depth corresponding to a given thermoluminescent peak to be evaluated by substituting measured experimental parameters into an appropriate relationship derived from the basic equation. Many techniques for doing this have been developed. Mention will be made of four which have proved particularly useful.

(a) The initial rise method
At temperatures well below the glow peak maximum, when the traps first begin to empty, $I_t$ is proportional to $\exp(-E/kT)$ thus a plot of log $I$ against $1/T$ will be a straight line of slope $E/k$.

(b) The half width method
The temperature span between points on the glow curve corresponding to intensities $I_{\text{max}}/2$ can be related to the trap depth $E$. The exact relationship depends upon the glow curve equation which is assumed and this method can only be used for isolated peaks.

(c) The Fermi level method
During readout of a glow curve the quasi-Fermi level at the temperature of the glow peak maximum will
correspond with the trap level.

(d) **The change of glow curve with heating rate method**

The glow peak maximum shifts to a higher temperature as the heating rate is increased and \( E \) can be evaluated from the shift. For example, in the case of the Randall-Wilkins equation, the relationship between the glow curve maximum and the heating rate is given by:

\[
\frac{rE}{k(T_{\text{max}})^2} = s \exp(-E/kT_{\text{max}})
\]

1.3 Thermoluminescence Dosimetry

*Ideally*, practical integrating dosimeter systems show a linear relationship between absorbed dose and readout signal up to a certain maximum value beyond which the signal flattens out as the response of the dosimeter saturates. Whilst this type of response is characteristic of some thermoluminescent phosphors in many others the production of additional traps by irradiation already referred to, may lead to the sensitivity of a dosimeter becoming enhanced as the absorbed radiation dose increases. A similar result is also theoretically possible if radiation damage results in the creation of additional luminescence centres leading to a greater probability of luminescent recombination of released holes and electrons. The proportion of the radiation energy converted into luminescence is so small that up to tenfold increases in sensitivity may be produced in this way by prior irradiation of some samples. This effect leads to the readout signal from the dosimeters increasing more than proportionately with the absorbed radiation dose and is consequently known as supralinearity.

The onset of supralinearity may be associated with total absorbed doses anywhere in the range between 10 rad and 100 kilorad, whilst saturation itself may correspond to total absorbed doses as low as 100 rad or as high as several megarad. Enhanced sensitivity resulting from the previous irradiation of a supralinear phosphor is not
stable - it will normally be annealed out by prolonged slow heating. These effects mean that detailed prior studies must be made of the behaviour of any phosphor intended for use in a practical dosimetry system and that phosphors must be carefully chosen to ensure their suitability for each potential application.

The requirements for a good dosimetry phosphor can be summarised as follows:

(a) A high concentration of electron and hole traps.

(b) A high efficiency for luminescent recombination when the trapped electrons or holes are thermally released.

(c) A trap depth large enough (and frequency factor low enough) to give long storage of trapped electrons or holes, with negligible fading; but not so deep as to require heating of the sample to the point where infra-red radiation leads to a high noise background during readout.

(d) A simple trap structure giving rise to a simple glow curve with, for choice, only one peak.

(e) A spectral distribution of luminescence which matches that of the detector system used, (Figure 1.6).

(f) Traps, activators and host lattice stable under the action of radiation so as to avoid the conditions under which supralinearity arises.

(g) Suitability for incorporation into small solid dosimeters by encapsulation, extrusion moulding or some equivalent process.

(h) Cheap to purchase, ideally cheap enough to use on a disposable basis.

(i) Low Z composition giving near tissue equivalence over a wide range of photon energies.

In practice only a few materials show sufficient sensitivity for dose measurements at low levels. The most sensitive are some forms of natural calcium fluoride which can register up to 60,000 single photon counts per mg per rad on a 5.0 cm diameter EMI 6255 S quartz window photomultiplier 50 cm from the sample. The corresponding figure for artificially produced calcium fluoride phosphor
activated with manganese is 10,060 whilst for the widely used lithium fluoride marketed by the Harshaw Chemical Company as TLD 100 it is 905.(11)

Table 2 gives some of the properties of phosphors for which the thermoluminescence has been extensively studied. Of these lithium fluoride is by far the most widely used phosphor. This is partly because, as shown on Figure 1.6, its light output is well matched to the spectral sensitivity of standard photomultiplier tube cathodes. This combined with near tissue equivalence, relatively high sensitivity, chemical stability, ready availability, and well established annealing procedures mean that it is very suitable for many of the measurements commonly associated with medical and radiological protection requirements - particularly in the case of X-rays and gamma rays. Alpha and beta dosimetry can also be readily carried out using lithium fluoride whilst for neutrons it has the advantage that there are two isotopes of lithium, $^6\text{Li}$ and $^7\text{Li}$ with about 7.5% of natural lithium normally being in the form of the mass 6 isotope. This has a large thermal neutron cross-section (750 barns) for the exothermic $(n,-\alpha)$ reaction - the emitted $\alpha$ particle producing a high thermoluminescent output. The phosphor is available in three forms, a normal form (e.g. TLD 100) containing the natural proportions of each isotope and two isotopically separated forms (e.g. TLD 600 and TLD 700) containing isotopically pure lithium. $^7\text{LiF}$ has almost no response to neutrons and can be used to measure gamma ray doses in the presence of a large neutron flux. $^6\text{LiF}$ has a very large thermal neutron response but is not widely used. Even TLD 100, the standard form of the phosphor, is about twenty times as sensitive to a thermal neutron dose as to a corresponding gamma ray dose. (Rem doses). Consequently by comparing readings from a $^7\text{LiF}$ TLD 700 dosimeter with those from a standard TLD 100 dosimeter gamma and thermal neutron doses in a mixed radiation field can be evaluated separately.

Various studies have also been made of the possibility
of designing albedo type fast neutron dosimeters and these show considerable promise. They consist of a standard phosphor dosimeter worn on the chest and shielded against thermal neutrons from the front by a cadmium foil. They then respond to thermal neutrons entering them from the wearers trunk and produced primarily by body moderation of incident fast neutrons. Typical designs have been described by Harvey in the U.K., Piesch in Germany, Mejdahl in Denmark, and Hankins and Korba both of the U.S.A., (12, 13, 14, 15, 16). In the use of such composite dosimeters an important correction arises from the different response of the two forms of LiF to low energy photons as a result of the difference in their densities and mass stopping powers. Direct measurements of fast neutron doses through recoil proton irradiation of the phosphor when surrounded by a hydrogenous layer have also been made. This method is not satisfactory when there is a mixed radiation field such that the gamma rays contribute a significant proportion of the dose. Wingate et al (17) have shown that the sensitivity to a given neutron dose is at best only a few percent of that for an equivalent gamma dose.

Apart from the neutron discriminating properties obtainable with lithium fluoride this phosphor has a number of other advantages which make it particularly suitable for general purpose dosimetry. Among these are:

(a) Non-hygroscopic and convenient to handle.
(b) Response relatively independent of photon energy (18) due to the effective atomic number being very close to that of tissue (Figure 1.7).
(c) Good resistance to fading.
(d) Light output fully drained before heating produces excessive infra-red radiation.
(e) Readily formed into solid dosimeters by extrusion or moulding.

Most of the thermoluminescence dosimetry research
reported in chapters 2 and 6 of this thesis was carried out using lithium fluoride and its special properties are further considered in more detail in section 1.6. In addition chapter 2 is entirely devoted to a study of the particular problems which arise in the routine use of LiF loaded teflon dosimeters for measurements at protection levels.

The remainder of this section consists of a brief survey both of some of the alternative phosphors listed in table 2 but currently used to a lesser extent than lithium fluoride, and of the instrumentation associated with a TLD reader intended purely for dosimetry.

Lithium tetraborate or Li₂B₄O₇ has a slightly better energy response than LiF at low quantum energies (Figure 1.7). It has therefore been recommended for routine measurements in hospitals. Most forms show appreciably lower sensitivity than LiF and this problem is accentuated by the longer wavelength of the emitted light which is harder to distinguish from incandescence radiation emitted by the heating tray. Compared with LiF it has a relatively simple glow curve and shows comparable supralinearity. Like LiF it has a very high sensitivity to thermal neutrons — in this case due to the presence of the ¹⁰B isotope in the borate (Natural abundance of ¹⁰B = 19.6%) as well as of the ⁶Li isotope. The presence of two neutron sensitive isotopes makes the production of isotopically separated Li₂B₄O₇ uneconomic. Because of its relatively simple glow curve structure lithium tetraborate is more simplyannealed than lithium fluoride but in the powder form the phosphor is rather hygroscopic and can therefore be very troublesome to handle.

Calcium fluoride is available in two forms — a natural product which is very sensitive but has a most complex glow curve, and a laboratory product containing manganese activator which has a simple single peak glow curve. Its main drawback is a high effective atomic number of 16.3 (compared with 7.6 for air and 7.2 for tissue). This
leads to a very exaggerated response to low energy photons (Figure 1.7). This limitation has been turned to practical advantage in a 'tandem dosimeter' containing both LiF and CaF$_2$ phosphors which was developed to determine the effective energy of a mixed X-ray beam from the ratio of the two responses (by Gorbics and Attix) (19). This technique for X-ray energy determination has been used in some of the work reported later in this thesis.

Calcium sulphate has been activated by the addition of samarium, manganese, dysprosium and thulium. All these four forms of the phosphor show exceptionally high sensitivity but the latter two which are the more recently developed activators are probably to be preferred. Whereas CaSO$_4$ : Mn gives a glow curve with a single peak at 100°C that of CaSO$_4$ : Dy contains many peaks but the principal one has its maximum at 140°C which gives greater resistance to fading. This is a very useful phosphor for environmental measurements at low dose levels but if the dose is to be integrated over long periods of time the phosphor must be protected from light and the ambient temperature monitored carefully - cooling being desirable.

The properties of beryllium oxide have only been studied to a limited extent probably due to the extremely high toxicity of the powdered material (D.W.L.$\text{air} = 0.01 \mu \text{g/m}^3$). However it shows a clean glow curve with a maximum at about 220°C and also has very good tissue equivalence. The light emitted is slightly too short in wavelength to match the standard photomultiplier tube cathode but using quartz window tubes with an extended UV response this phosphor is found to be very sensitive. Supralinearity sets in at the relatively low level of about 100 rad but saturation level is in excess of $10^5$ rad. In the powdered form this phosphor is unsuitable for use in personnel dosimetry but in the form of sintered ceramic type material BeO provides good tissue equivalent dosimeter elements. The glow curve maximum is at about the optimum temperature and the material shows exceptionally high stability and resistance to discolouration, changes in
transparency or damage. BeO discs have also been used very successfully for TSEE dosimetry and are the preferred sensitive elements for many workers in that field.

Many other materials have been shown to exhibit some degree of thermoluminescence and with further study of suitable activators may prove to be sufficiently sensitive to be of dosimetric interest. Examples are strontium fluoride, strontium sulphate and aluminium oxide. The latter has the advantage that like BeO it can be prepared in ceramic form but all these materials have a high effective atomic number and so their response is strongly energy dependant. There are also two natural materials, quartz and feldspar, for which the thermoluminescence is particularly important. Grains of one or other of these materials are almost invariably present in pottery and the accumulated thermoluminescence stored in these grains has been used both to determine the age of pottery sherds found by archaeological expeditions and to estimate the radiation dose received by survivors of the atom bomb dropped on Hiroshima using readings obtained from roof tiles. Such samples generally show fairly complex glow curves which depend upon their exact origin and thermal history. Quartz in particular has a most unusual TL response, in its natural form it shows a low temperature glow curve peak at 100°C for which the sensitivity is rather low. By heating the quartz this sensitivity can be enhanced by up to three orders of magnitude. This behaviour makes accurate dosimetry with quartz a complex procedure.

Instruments for readout of thermoluminescence dosimeters have been available commercially for about a decade now. The basic requirements are adequate sensitivity with acceptably low background signal and good reproducibility. It is also advantageous when the readout procedure followed eliminates or simplifies the post readout annealing requirements for the dosimeters. This latter point is considered in more detail in chapter 2 where it is also shown that in many respects the design requirements
for a dosimetry reader conflict with those for a reader intended for phosphor research.

Traditionally dosimetry readers fall into two categories, those integrating the total light output from the sample and those recording the peak signal intensity only. In practice the former procedure has proved more reliable and is almost invariably used today. Since the counting of individual light photons is not practicable at high dose levels the light output from the phosphor is either integrated as an analogue signal (e.g. in the form of a charge on a capacitor) or is digitised and fed into a counter. The instantaneous signal output is also often available to operate a recorder for the plotting of glow curves if required.

Readers differ most in the design of the heater. A lengthy readout cycle must lead to more background signal arising from the photomultiplier tube dark current so rapid heating is usually featured. If an accurate glow curve is required a linear temperature rise must be the objective, in other cases a viable alternative is the so-called isothermal readout where the sample is heated as rapidly as possible to its equilibrium temperature and then maintained at this value until the signal has been completely drained. A popular reader design of this type uses a heater with a Curie point temperature controller. The simplest heating system employs a constant heat input to the readout chamber giving an approximately exponential temperature rise. More sophisticated readers incorporate temperature controllers giving a temperature time curve which shows three or more controlled zones that may be plateaux or linear temperature ramps; the theoretical justification for this is discussed later. In many commercial instruments designed for use with phosphors exhibiting both low and high temperature peaks the signal from the former (which are more susceptible to fading than the latter) is rejected. Most heaters use a resistive element as, or in thermal contact with, a suitably shaped sample tray. Difficulties arise in ensuring uniform
heating of thick solid dosimeter elements by thermal conduction from the sample tray as the sample material (dosimeter matrix) is usually of poor thermal conductivity. Some heating systems therefore use hot nitrogen gas or some other method to heat both sides of the dosimeter simultaneously in order to minimise this problem.

A feature of almost all readers is that an inert gas can be circulated through the heating chamber during the readout process as this has been shown to appreciably reduce the background signal from the dosimeters. Another universal facility is provision to check the sensitivity of the light monitoring system by putting a standard light source into the readout chamber with the heater circuit disabled. Electronic design is usually fairly straightforward - the principal unconventional feature is that the photomultiplier tube must be operated from a negative E.H.T. supply with the anode at earth potential in order to facilitate the measurement of very small anode currents.

1.4 Thermally Stimulated Exo-electron Emission Dosimetry

TSEE dosimetry is closely analogous to thermoluminescence dosimetry but involves only the properties of a thin surface layer of the dosimeter element.

Electron band theory as discussed in section 1.2 provides a simple picture by which both thermoluminescence and thermally stimulated exo-electron emission can be explained. Absorbed radiation energy leads to electrons being elevated from the valence band to the conduction band where they migrate until trapped at impurity centres which have produced localised electron traps. In thermoluminescent materials some of these are too far below the bottom of the conduction band for the electrons to escape again unless appreciable heat energy is supplied. When this is done the trapped electrons can return to the conduction band at a characteristic 'glow peak' temperature which depends upon the energy gap between the trap and the bottom of the conduction band.

Electrons which are released into the conduction band
in this way may:

(1) Return to the valence band by a radiationless transition, the released energy appearing as phonons of lattice vibration.

(2) Recombine with trapped holes in a luminescent transition. This constitutes thermoluminescence.

(3) Escape through the surface of the crystal—probably at a point where a surface defect leads to an intense localised electric field assisting their escape, (Figure 1.8). This last possibility is known as exo-electron emission and the escaping electrons typically have energies of less than 1 eV. They can, however, be detected if released into a gas flow counter or electron multiplier system.

The simple model described above is not completely adequate. It does not explain the fact that thermoluminescence can also occur when the heat energy releases holes which migrate to the trapped electrons; although in this case exo-electron emission will not be observed. In addition it implies that a peak in the TSEE emission curve will always occur at the same characteristic temperature as one in the TL glow curve, leading to a close correlation between glow and emission curves. A slightly more sophisticated model allows for the possibility of distortion of the normal energy level system in the surface layers of the emitting crystals, and hence can account for differences in the peak temperatures shown by these two curves. Furthermore when glow curve peaks are associated with the thermal release of holes which recombine with trapped electrons at luminescence centres these will be totally absent from electron emission curves.

Among the earliest observations of exo-electron emission effects were those of Klapheke in 1931 (20). He noted that some inorganic crystals emitted electrons when heated following irradiation by U.V. light. The release of exo-electrons following mechanical stresses applied to oxydised metal surfaces was reported a few years
later. The former effect has developed into a branch of radiation dosimetry whilst the latter has been extensively applied to the study of surface strains in metals. The dosimetry developments were however only undertaken after the war - largely as a result of systematic investigations started by Professor Kramer in 1949 and carried on by a group with him at Braunschweig.

Kramer was the first to show how important electro-static effects could be in inhibiting exo-electron emission from non-conducting crystals and he demonstrated that a linear dose-emission relationship could be observed with many crystals if the sample was intimately mixed with powdered graphite. Kramer's first report dealing specifically with the use of exo-electron emission for radiation dosimetry was published in 1963.(21) Amongst the advantages listed were low cost, simple readout, wide dynamic range, good energy response and re-usability of dosimeters. He pointed out however that instability of the exo-electron sensitive surface layers could lead to sensitivity changes.

Since this time exo-electron dosimetry research has been initiated in a number of centres. Of particular importance has been the work done by the group at Oak Ridge under Becker and Gammage (22) as well as that carried out at Munich under Regulla (23). Both these groups have made extensive studies of the properties of ceramic disc BeO dosimeters. These offer a number of advantages as convenient tissue equivalent dosimeters in a geometrically stable form. Many inorganic crystals which exhibit thermoluminescence are also suitable for exo-electron dosimetry, examples are LiF, Li₂B₄O₇, CaSO₄. In addition metal oxide films formed on the surface of metallic samples can be used and offer the advantage of being almost immune to electro-static effects. For dosimetry Al₂O₃ is the most important, both because the surface of aluminium oxides readily and because of its low atomic number.

The heart of an exo-electron dosimeter reader is usually a gas flow counter head incorporating a sample heating plate on which the planchet holding the dosimeter is placed.
The basic requirement is to ensure adequate sensitivity for the reliable detection of electrons with energies perhaps as low as 0.01 to 0.1 eV. Much of the early work was carried out with simple open point counters—a form of geiger counter in which the anode is a metal sphere very much less than 1 mm in diameter and which can be operated successfully in air at S.T.P. using external electronic quenching. Later the use of gas flow geiger counters became much more usual and recently several workers have attempted to construct proportional counter systems with which adequate sensitivity can be reliably attained. One such system is discussed in detail in chapter 3 of this thesis.

The remainder of the TSEE dosimeter reader is closely analogous to a TLD reader. Exactly the same considerations apply to the design of the sample heating control circuits except that it is often useful to be able to attain a higher maximum readout temperature. (With the TSEE reader the useful limit is set by thermionic emission of electrons and not by infra-red radiation as in a TLD reader).

The output pulses from the counter are amplified and fed into a scaler to integrate the total electron emission between selected temperatures. An emission curve can be obtained by connecting a recorder to a ratemeter placed in parallel with the scaler.

At the present time the majority of TSEE measurements are carried out at centres engaged in active research in the field using their own design of reader. Nevertheless a commercial design of TSEE dosimeter has been available from a company in West Germany since 1970.* This comprises a geiger counter type reader head primarily designed for powder samples, together with all the associated read-out and heater control instrumentation built into one exo-electron dosimeter unit.

There are many applications for which exo-electron dosimeters would appear to offer advantages over other

* Atomika, Technische Physik GmbH., D-9000 Munchen 13, Bauerstrasse 19
types, the following suggestions should be regarded as merely illustrative:

(a) Charged particle and/or neutron dosimetry.
The principal difficulty with these measurements usually arises from the associated gamma ray dose. Volume sensitive dosimeter elements tend to show an accentuated gamma response as the whole volume responds uniformly (assuming electronic equilibrium) whilst charged particles or neutron induced secondaries interact almost entirely within a very thin surface layer. With TSEE dosimeters this problem is side stepped as the actual sensitive volume is limited to a very thin layer comparable in thickness with the typical range of the particles.

(b) Interface dosimetry or dosimetry in volumes where there is a very steep dose gradient.
Exo-electron dosimeters are the first which enable doses to be measured at a specific depth below the surface instead of providing an observed value which is a mean measured over a layer of appreciable thickness. Thus exo-electron measurements represent a method of semi-micro dosimetry which can be applied to such important problems as, for example, evaluating the dose experienced at the periosteal surface of a bone or by the osteo-progenitor cells close to this surface in the course of X-ray examination or treatment.

(c) Some TSEE dosimeters such as those prepared from ceramic discs of BeO appear to offer certain advantages over TL dosimeters for personnel monitoring. In particular they give a higher sensitivity at low dose levels, are cheaper and have less critical annealing requirements than lithium fluoride.

(d) Disposable tear-off dosimeters prepared from rolls of treated paper are felt by many workers to be practicable and would offer the ultimate in low cost monitoring. (Optical readout of these is thought
by Professor Kramer to be a probable development in the future.)

e) Imaging systems based on the recording of a dose distribution pattern by an exo-electron sensitive surface for subsequent readout using some sort of scanning counter (or a large area counter and a scanning laser beam readout) form another possible future development. Medical research work on the use of transmission fast neutron radiography which should theoretically be a diagnostic tool of great clinical value, was abandoned largely because photographic methods of recording the variations in transmitted intensity were not successful and no satisfactory alternative could be found. Developments in scanning exo-electrons imaging systems could lead to the use of this procedure finally becoming established.

1.5 Dosimetry based on Thermally Stimulated Conductivity

The basic mechanism of the TSC process has been explained briefly in section 1.2. It forms an alternative thermal process to TLD or TSEE for the evaluation of radiation exposure of many crystalline materials. It is also important because TSC measurements can help solve the problem of deciding between alternative models for the supralinearity effect observed with many thermoluminescent phosphors. As a dosimetry technique it offers several advantages:

(1) Relatively cheap disposable sample materials appear suitable.

(2) There is no need for the samples to be transparent.

(3) The apparatus for readout of TSC signals is particularly cheap and simple.

(4) Some organic plastic materials exhibit this effect and are closely tissue equivalent for both neutrons and gamma rays.
These advantages are offset by three disadvantages which have so far effectively prevented the use of this method for practical dosimetry. They are:

1. Many potential sample materials are unsuitable due to excessive normal conductivity at the elevated temperatures required during readout.
2. There has been only one report to date of sample materials which provide sufficient sensitivity for measurements to be made at protection levels.
3. It is difficult to obtain amorphous or polycrystalline samples which offer adequate stability in their TSC response.

Chapter 4 of this thesis is a report of TSC measurements carried out during the early part of this research project. Little difficulty was found in designing suitable readout equipment and measurements of the radiation induced TSC sensitivity of various insulating polymers were readily obtained. Unfortunately these proved to be far from reproducible and it was concluded on the basis of this work that the drawbacks just discussed made it unlikely many practical applications for TSC dosimetry would be found in the near future.

Recently more encouraging results have been reported by two groups active in this field. Harper and Thomas at Cardiff have made a detailed study of how the TSC response of plastics is related to their structure. This has enabled them on the one hand to select samples of relatively high sensitivity and stability, and on the other to 'design' plastics in which the properties required for dosimetric applications have been accentuated. Their work gives good promise of forming the basis of a convenient dosimeter system for measurements in the 1 rad to 1 kilorad range in hospital radiotherapy departments. The composition of the plastic dosimeters can be readily modified to give both a density and an effective atomic number which are exactly equivalent to various biological tissues. Another potential advantage is that these dosimeters have
a relatively large hydrogenous content which makes them of considerable interest for fast neutron dosimetry.(24)

The other group referred to above is at the University of Wisconsin. It is under the control of Cameron (The 'father' of TLD) and Moran, and has been working primarily with very high purity ionic crystalline insulators. This group have introduced the alternative name RITAC (Radiation Induced Thermally Activated Currents) dosimetry. They have reported measurements on UV grade optical window sapphire (an oxide of aluminium crystallised in a trigonal lattice) for which the impurity content has been reduced below 100 ppm. With appropriate attention to the reader design to minimise background signals arising from contact and edge current noise, measurements have proved possible for doses as low as 1 mrad, (corresponding to a circulated charge of the order of $10^{-12}$ coulomb). Problems arose from internal fields generated by polarization of the samples and exact tissue equivalence cannot be achieved with this material but the high sensitivity and great physical stability of these sapphire dosimeters makes them of considerable potential interest.(25)

1.6 The present status of lithium fluoride thermoluminescence dosimetry

Apart from the traditional photographic film no solid state dosimeters have achieved an acceptability in any way comparable with lithium fluoride thermoluminescence dosimeters. This high acceptability has resulted from convincing demonstrations that, given proper care and attention, precise measurements can reliably be carried out in almost any laboratory. Nevertheless there are many pitfalls which can lead to a disastrous loss of accuracy. One of the most serious of these is dosimeter sensitivity changes resulting from variations in the annealing procedure which is necessary both to ensure that previously used dosimeters have been completely drained of stored energy and also to bring them into a condition of standard sensitivity for re-use.
The standard annealing procedure for eliminating the effect of previous irradiation in lithium fluoride is to heat it to 400°C for one hour. Following this procedure the subsequent glow curve obtained from the phosphor shows five peaks (Figure 1.9a). The final TL sensitivity depends upon how quickly the phosphor is cooled from the annealing temperature to room temperature. By following the 400°C anneal with an 80°C anneal for a further 24 hours the low temperature peaks are almost eliminated and the LiF shows a relatively simple glow curve peaking at about 210°C (Figure 1.9b). The exact peak temperature depends not only upon the heating rate as shown by the glow curve analysis in section 1.2, but also upon the spectral sensitivity of the photo-multiplier tube response. Care must therefore always be exercised when drawing conclusions from differences between glow curves obtained with different readout systems. Glow curves also depend to some extent on the type of ionizing radiation responsible. For example Figure 1.9b shows that the LiF glow curve resulting from irradiation by neutrons shows an additional high temperature peak at about 270°C so that the normal heating cycle used for the readout of LiF following gamma irradiation will not drain the phosphor adequately after exposure to neutrons.

Zimmerman measured the half lives at 20°C of the five peaks shown on the glow curve of Figure 8 and found they were 5 min., 10 h., 0.5 y., 7.0 y. and 80 y, respectively. The purpose of the 80°C anneal is to reduce the radiation energy stored in the first two peaks which exhibit rapid fading. This procedure had been first discovered accidentally by Buch at the University of Wisconsin in 1962 and for several years prior to detailed studies of the effects of different annealing procedures it was followed as an arbitrary 'recipe'. Before this discovery Cameron used a short post-irradiation anneal of about 5 min at 100°C to eliminate any readout signal from peaks 1 or 2. In 1965 Zimmerman made an extensive study of the effect of varying both the time and the temperature for pre-irradiation
annealing. His results were reported to the First International Conference on Luminescent Dosimetry and Figure 1.10a is a set of glow curves from these results which shows the effect of varying the time of the 80°C anneal. Zimmerman concluded the existing practice of annealing at 80°C for 24 hours did indeed give optimum results. He showed that pre-annealing at temperatures above 100°C led to a modification of the glow curve, a sixth peak appearing at a substantially higher temperature than peak 5. The glow curves on Figure 1.10b show this for a pre-annealing temperature of 125°C.

Other conclusions from Zimmerman's work were that the 400°C anneal could increase the sensitivity of a phosphor by a factor of up to five and that both the final sensitivity and the exact glow curve shape were critically dependent upon the rate of cooling. Figures 1.11a and 1.11b show the effect of changes in the annealing temperature and time on the glow curve and sensitivity respectively, whilst similarly Figures 1.12a and 1.12b show the effect of changes in the cooling rate. It will be seen that the more rapid the cooling the more sensitive the resulting phosphor becomes(26). Some of the results of a later study made by Wilson on the effect of the annealing temperature on the pre-sensitising of lithium fluoride (27) are also shown on Figure 1.13.

The results of low temperature post-irradiation annealing were also extensively studied by Zimmerman. By measurement of the rate of fading of stored energy in each of the principal peaks Zimmerman was able to estimate both the depth (E) and the frequency factor (s) for the traps responsible. According to the Randall & Wilkins model (first order kinetics) the number of trapped electrons after a time t (n) is related to the initial number (n₀) and to the temperature (T) by an equation of the form:

\[ n = n_0 \exp \left(-st \exp \left(-\frac{E}{kT}\right)\right) \]
Differentiating this gives:

\[ \frac{- \log \frac{n}{n_0}}{t} = s \exp \left(-\frac{E}{kT}\right) \]

If \( \log \frac{n}{n_0} \) is plotted against \( 1/T \), \( s \) and \( E \) can be obtained from the resulting intercept and slope, also departures from monomolecular kinetics will be apparent if the resulting graph is not a straight line. Zimmerman's results confirmed the application of the Randall & Wilkins model to the principal peaks of lithium fluoride. His values for the trap depths and frequency factors are shown in table 3. He interpreted the value obtained for the frequency factor for peak 6 as indicating that this arose from the retrapping of electrons released from other traps during the post-irradiation annealing. Some of the results of these experiments are shown on figures 1.14 and 1.15.

It is interesting to note that the pre-irradiation annealing process operates more rapidly than the post-irradiation one. For example at 143°C the half lives are respectively 0.75 and 0.98 hours. This suggests that if the actual annealing mechanisms are the same for both cases the presence of a trapped electron helps stabilise the crystal lattice defect responsible for the trap.

Much other research has been published on the dosimetric properties of lithium fluoride. For example Tochilin and Goldstein (28) have shown that there is no variation in response with dose rate, even for rates as high as \( 10^{10} \) rad sec\(^{-1} \) whilst Kastner et al. (29) have shown that the response to beta rays is the same on an absorbed energy basis as the response to gamma rays. Marrone and Attix (30) have shown that the phosphor sensitivity may be permanently reduced by radiation damage following doses in excess of \( 10^4 \) rad, and Pohlit (31) showed that electrons trapped in shallow traps could relax into deeper traps during storage leading to a greater part of the readout signal appearing in the higher temperature peaks (Figure 1.16). Wagner & Cameron (32) reported
that the extent of the supralinearity shown by LiF varies with the photon energy of the radiation (Figure 1.17) and Suntharalingam & Cameron (33) have carried out similar studies with mono-energetic electrons. Similarly a number of reports have been published dealing with changes in the response of LiF when it is used for less common dosimetric applications such as measurements for alpha particles, very low energy beta particles and high energy charged particles from accelerators.

Several workers have studied the importance of grain size both theoretically and experimentally. Typically lithium fluoride is used in the form of a powder with a grain size of between 75 and 175 micron. This is obtained by using only the fraction retained between 80 and 200 mesh sieves. A detailed study of the effect of grain size was made by Chan and Burlin (34) using calculations based on cavity theory. Figure 1.18 for LiF grains in a teflon matrix (as used in many practical dosimetry systems) is reproduced from their paper. It shows that for teflon loaded dosimeters the optimum grain size is smaller than that normally used for loose powder dosimeters. Chan & Burlin's calculations have substantially confirmed experimental work previously carried out by Zanelli.(35)

Very extensive studies of the optimum composition and method of preparation of lithium fluoride for use in TL dosimetry have been made in many laboratories. Rossiter at the N.P.L. has concluded that most of the 'luminescent grade' LiF is prepared to a specification based on 80 ppm of Mg and 15 ppm of Ti.

Two other important properties of dosimetric LiF are non-radiation induced thermoluminescence and non-thermal (anomalous) fading. These are sometimes abbreviated to NRI-TL and NTF respectively.

With lithium fluoride NRI-TL shows three peaks, one is induced by grinding and is almost exactly superimposed on the main dosimetry peak for gamma rays, one comes from shaking the ground powder and appears at a readout temperature of about 315°C, whilst one builds up
slowly during long term storage but has a still higher readout temperature. If present, the peak due to grinding is usually the most serious but once it has been annealed out following the original manufacture of the phosphor it does not normally reappear unless the sample is shaken sufficiently severely to cause further damage to the surface of the grains. The storage peak comes at too high a temperature to interfere with normal dosimetry and therefore the peak caused by shaking is likely to be the most troublesome. McCall and Fix (36) showed however that when readout was made in an inert gas atmosphere this peak could be quenched and practical TL readers incorporate nitrogen flow facilities which effectively eliminate troubles from this cause. Other phosphors also show similar triboluminescent effects which can be quenched in a similar way. It has been shown that NRI-TL is entirely a surface effect, probably chemical in origin, and that the quenching is not purely a matter of the exclusion of oxygen as an argon atmosphere during readout is more effective with some phosphors (e.g. CaF$_2$ : Mn) than a nitrogen one; but extensive studies on the cause of the effect have not continued - probably partly because it can be so readily controlled.

Non-thermal fading is a phenomenon the importance of which was first realised by Christodoulides and Ettinger (37). With most dosimetry phosphors its magnitude is relatively small but it can be observed more readily for some geological samples such as quartz or lava. Its main characteristic is a rapid initial fading unrelated to the glow peak temperature and a final stage where the residual signal is completely free of the effect.(38) It often shows some temperature dependence but this is very small compared with that of the normal fading process. The explanation is probably due to quantum mechanical tunneling providing a possible escape route for some of the trapped electrons, or to resonance transitions of electrons directly between donor impurity electron traps and acceptor impurity luminescent centres if there is a small overlap of the wave
functions associated with each. Some NTF involves luminescence and some appears to be non-radiative. In the latter case it may be that luminescent centres trap positive holes from the valence band, leading to hole migration and eventual recombination at the electron trap instead of electron migration followed by recombination at the hole trap, (Figure 1.19). The migration of traps is another possible contributory explanation. Whatever its cause NTF remains relatively unimportant with dosimetric LiF.

Given adequate care in avoiding the various pitfalls discussed in this section, careful measurements made with LiF dosimeters in a good design of reader give results which are reliable at least to within 2% except at very low dose levels. The National Physical Laboratory has recently been involved in an international intercomparison of radiotherapy doses and extensive trials conducted by Dr. Rossiter showed this accuracy was readily attainable using the very convenient TLD 100 extruded ribbon solid lithium fluoride dosimeters marketed by the Harshaw Chemical Company. Measurements made with conventional powder dosimeters should in principle be even more accurate, although the practical difficulties of using powder dosimeters on a large scale may make it difficult to always achieve the optimum accuracy in practice.

Lithium fluoride is also widely used in another form - embedded in a teflon matrix. Achieving a comparable degree of accuracy with this form of dosimeter imposes a number of additional problems which are considered in more detail in the next chapter. Nevertheless the thinness of the discs in which these dosimeters are readily produced is advantageous when accurate measurements of skin dose are required and dosimeters of this type have been selected by the National Radiological Protection Board for their TLD personnel monitoring service due to be inaugurated in the near future.

The conclusion must be that LiF TLD dosimetry carried out by experienced personnel using good equipment is
inherently capable of very satisfactory results, but that at the present time many centres using the method have not attained the necessary degree of experience and, because of inadequate attention to one or more of the complications which can affect the method, have found their measurements both disappointing and inaccurate.

1.7 Survey of Work reported

A very much more comprehensive review of solid state dosimetry prepared by Becker (39) forms an important guide to both the technology and the literature of the subject. These are now so extensive that no thesis can attempt to review the field adequately and the material incorporated in the preceding sections has been restricted to topics directly relevant to the experimental work reported in later chapters; even so it has necessarily been extremely selective and is strictly limited to methods of readout involving thermal stimulation. Of these thermoluminescence is by far the longest established and most widely used. TLD techniques have now been so extensively developed that they can be used with confidence for many types of routine dosimetry. Nevertheless there are still many pitfalls which, if not properly understood, can lead to erroneous or misleading results and chapter 2, which contains a state of the art study of lithium fluoride TLD measurements made at protection levels in hospitals, constitutes an attempt to identify some of these hazards.

Chapter 3 is an account of work carried out during attempts to develop a reliable TSEE dosimeter. This method has also been studied for an appreciable time and a comprehensive general review has been prepared by Becker (40). It is now capable of reliable use for appropriate practical applications and the thesis includes a number of references to some of these. Similarly chapter 4 describes studies of TSC as a dosimetric procedure. This is a newer technique which has only relatively recently showed real promise of developing into a dosimetric system of practical value. The thesis then continues with a chapter showing
how an intercomparison of TLD, TSEE and TSC observations can be used to help elucidate the fundamental physical behaviour responsible for the properties of materials of potential dosimetric interest.

Finally in chapter 6 a series of illustrative examples is given to show the practical value of the methods discussed for carrying out some of the measurements in hospitals which have traditionally been regarded as difficult to make reliably.
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<th>Volume</th>
<th>Page</th>
<th>Year</th>
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<th>Type of centre induced by ionising radiation</th>
<th>Measurement Procedure</th>
<th>Effect</th>
<th>Description</th>
</tr>
</thead>
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<tr>
<td>Radiation induced centres stable to measuring procedure</td>
<td>Illumination with light - usually UV or visible</td>
<td>Colouration</td>
<td>Centres absorb light in originally transparent spectral region</td>
</tr>
<tr>
<td></td>
<td>Illumination with UV or visible light of shorter wavelength than is emitted in luminescence</td>
<td>Radiophotoluminescence</td>
<td>Unilluminated solid is not luminescent. New centres absorb measuring light &amp; re-emit at longer wavelengths as long as measuring light is incident.</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Degradation of luminescence</td>
<td>Unilluminated solid is not luminescent. New centres do not luminesce but quench normal luminescence which is emitted as long as measuring light is incident.</td>
</tr>
<tr>
<td>Radiation-induced centres destroyed by measurement procedure</td>
<td>Heating</td>
<td>Thermo-luminescence</td>
<td>Energy stored in centres is released as luminescence.</td>
</tr>
<tr>
<td></td>
<td>Illumination with light of longer wavelength than is emitted in luminescence</td>
<td>Stimulated luminescence</td>
<td>Energy stored in centres is released as luminescence.</td>
</tr>
</tbody>
</table>

(Based on a classification by Schulman - 1967)
TABLE 1.2

PROPERTIES OF SOME TL PHOSPHORS

<table>
<thead>
<tr>
<th>Ref.</th>
<th>Phosphor</th>
<th>Density</th>
<th>λ Max (nm)</th>
<th>Peak Temp. (°C)</th>
<th>Sensitivity of Lif.</th>
<th>Effective &amp; Range</th>
<th>Fading</th>
<th>Light Sensitivity</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>LiF:Mg</td>
<td>2.64</td>
<td>400</td>
<td>195</td>
<td>1.0</td>
<td>8.2</td>
<td>10mR-10^5 R</td>
<td>&lt;5%</td>
<td>No Complex glow curve</td>
</tr>
<tr>
<td>1</td>
<td>LiF:Mg</td>
<td>2.64</td>
<td>400</td>
<td>195</td>
<td>1.0</td>
<td>8.2</td>
<td>10mR-10^5 R</td>
<td>&lt;5%</td>
<td>No Red emission</td>
</tr>
<tr>
<td>1</td>
<td>CaF₂(nat)</td>
<td>3.18</td>
<td>380</td>
<td>260</td>
<td>23</td>
<td>16.3</td>
<td>1mR-10^10 R</td>
<td>Nil</td>
<td>Yes Poor energy response</td>
</tr>
<tr>
<td>1</td>
<td>CaF₂(Ca)</td>
<td>3.18</td>
<td>500</td>
<td>260</td>
<td>3</td>
<td>16.3</td>
<td>1mR-3x10^5 R</td>
<td>10% in month</td>
<td>No &quot; &quot; &quot; &quot;</td>
</tr>
<tr>
<td>1</td>
<td>SrSO₄:Eu</td>
<td>2.61</td>
<td>500</td>
<td>110</td>
<td>70</td>
<td>15.3</td>
<td>5μR-10^8 R</td>
<td>5% in 24 hrs.</td>
<td>Yes Very rapid fading</td>
</tr>
<tr>
<td>2</td>
<td>CaF₂(Dy)</td>
<td>3.18</td>
<td>480</td>
<td>140</td>
<td>30</td>
<td>16.3</td>
<td>1mR-10^6 R</td>
<td>10% in 24 hrs.</td>
<td>Yes Rapid fading</td>
</tr>
<tr>
<td>2</td>
<td>CaSO₄:Eu</td>
<td>2.61</td>
<td>480</td>
<td>213</td>
<td>35</td>
<td>15.3</td>
<td>1mR-10^6 R</td>
<td>2% in month</td>
<td>Yes Environmental Dosimeter</td>
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<tr>
<td>3</td>
<td>Al₂O₃</td>
<td>3.9</td>
<td>450</td>
<td>164</td>
<td>&lt;0.01</td>
<td>10.9</td>
<td>1R-10^5 R</td>
<td>Depends on Wave</td>
<td>Yes Various chemical states</td>
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<tr>
<td>4</td>
<td>BaO</td>
<td>3.01</td>
<td>330</td>
<td>180</td>
<td>1.2</td>
<td>7.2</td>
<td>20μR-10^4 R</td>
<td>Negligible</td>
<td>Yes Powder form toxic</td>
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<tr>
<td>5</td>
<td>SrF₂</td>
<td>4.18</td>
<td>290</td>
<td>0.03</td>
<td>33</td>
<td>100R-4x10^5 R</td>
<td>&quot;</td>
<td>No  High Z dosimeter</td>
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<tr>
<td>6</td>
<td>NaF</td>
<td>2.56</td>
<td>450</td>
<td>0.008</td>
<td>10.3</td>
<td>10R-10^6 R</td>
<td>&quot;</td>
<td>High temp. dosimeter</td>
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</table>

*As defined by Mayneord, Acta Int. U. against Cancer 2 271 1937

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Table 1.3

<table>
<thead>
<tr>
<th>Peak</th>
<th>E (eV)</th>
<th>n (10^-11 x sec^-1)</th>
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<td>Trap depth</td>
<td>Frequency factor</td>
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<tr>
<td>4</td>
<td>1.19</td>
<td>1.0 to 15</td>
</tr>
<tr>
<td>5</td>
<td>1.25</td>
<td>0.5 to 14</td>
</tr>
<tr>
<td>6</td>
<td>2.1</td>
<td>10^11</td>
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</table>
Figure 1.1a.
Lattice defects and colour centres.
(Schematic only)

Figure 1.1b.
Colour centre absorption and emission
(After Schäfer 1967)

- Int.
- Host crystal absorption.
- F centre absorption.
- F centre luminescent emission.

Figure 1.2
Configuration of electron trapping centres.

- (a) The model of the P centre: A halogen ion vacancy with an electron octahedrally surrounded by cations. The transitions of the transition moments of the F centre are also shown. Symmetry D₆h.
- (b) The model of the M centre: Two neighboring halogen ion vacancies with two electrons. The transition moments for one of the two electrons are also shown. Symmetry D₆h.
- (c) The model of the R center: Three neighboring halogen ion vacancies with three electrons arranged in a plane. The transition moments for one of the electrons are also shown. Symmetry C₃v.
- (d) The model of the N₂ center: Four halogen ion vacancies with four electrons arranged in a plane. The transition moments for two electrons of a different kind are shown. Symmetry C₄v.
- (e) The model of the N₄ center: Four halogen ion vacancies with four electrons arranged in a tetrahedron. The transition moments for one of the electrons are shown. Symmetry T₄h.
Figure 1.3a
The Mechanism of Electron Trapping

(a) Action of ionizing radiation.
(b) Heating. Electron trap less stable.
(c) Heating. Hole trap less stable.

Figure 1.3b
Glow curves for phosphors with single and multiple traps

\[ \text{Li}_2\text{B}_4\text{O}_7, \text{Mn} \]

\[ \text{CaF}_2: \text{Dy, (TLD 200)} \]

Figure 1.4
First Order Glow Curves. Calculated from Randall & Wilkins equation for different \( s \) and \( s \) values.

<table>
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<tr>
<th>Curve</th>
<th>( E ) (eV)</th>
<th>( s ) (sec(^{-1}))</th>
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</thead>
<tbody>
<tr>
<td>A</td>
<td>0.3</td>
<td>( 10^9 )</td>
</tr>
<tr>
<td>B</td>
<td>0.4</td>
<td>( 10^9 )</td>
</tr>
<tr>
<td>C</td>
<td>0.4</td>
<td>( 10^6 )</td>
</tr>
<tr>
<td>D</td>
<td>0.4</td>
<td>( 10^7 )</td>
</tr>
<tr>
<td>E</td>
<td>0.6</td>
<td>( 10^6 )</td>
</tr>
<tr>
<td>F</td>
<td>0.8</td>
<td>( 10^9 )</td>
</tr>
<tr>
<td>G</td>
<td>1.0</td>
<td>( 10^9 )</td>
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</table>


Figure 1.5
Theoretical glow curves for phosphor with single electron trap showing the effect of different amounts of retrapping. (R)

- --- Plotted from Randall & Wilkins equation.
- ---- Plotted from Garlick & Gibbons Equation.
- -------- Plotted from Haring & Adams equation.
**Figure 1.6**

Solid line: Emission spectrum of LiF (TLD 100)
Broken line: Response curve for type S II photocathode
Dotted line: Emission spectrum of LiF$_2$O$_2$:Mn.

**Figure 1.7**

Tissue equivalence of TL phosphors.

**Figure 1.8**

Band Theory Explanation of TL & TSEE
Figure 1.9(a)
Glow curve of unannealed LiF (TLD 100)
(Following X-ray exposure)

Figure 1.9(b)
Glow curves after optimum annealing.
LiF (TLD 100)

Figure 1.10
Pre-annealing of lithium fluoride

10a 80°C 10b 125°C

From Zimmerman et al (1965)
Figure 1.11a.
Effect of changes in annealing time and temperature for LiF (TLD 100)

- 1 hour at 400°C.
- 120 hour at 400°C.
- 1 hour at 600°C.

Temperature

Figure 1.11b.
Effect of changes in annealing time and temperature on sensitivity of LiF (TLD 100)

Relative Sensitivity

- 400°C.
- 80°C.
- 175°C.
- 125°C.

Annealing time (Hours)

Both figures from Zimmerman et al (1965)

Figure 1.12a.
Effect of changes in rate of cooling between 400°C and 80°C on glow curve and sensitivity of LiF (TLD 100)

Temperature

Figure 1.12b.
Effect of changes in rate of cooling on sensitivity of LiF (TLD 100)

Relative Sensitivity

- 100
- 50
- 0

Cooling Rate

Both figures from Zimmerman et al (1965)
Sensitivity for LiF (TLD-100) as a function of time for various annealing temperatures. All samples had previously been irradiated to $3.0 \times 10^4$ R of $^{137}$Cs gamma rays.


Resolution of the TL decay at 132°C into the exponential decays of peaks 4 and 5.

From Zimmerman et al 1966.

Change of sensitivity as a function of time.

Graph showing dependence of the half-life of peak 4 on temperature from which the trap depth and frequency factor is obtained.
Transfer of electrons from shallow to deep traps in LiF as a function of storage time after irradiation.

A 30 sec. at 25°C
B 30 min. at 80°C
C 60 min. at 80°C
D 16 hours at 80°C

(from Conrad Dosimeter Manual.)

Energy response of LiF and CaF₂ grains in teflon matrix.
From Chen & Burlin 1970.

Curves drawn for three different diameter grains.

Mechanisms responsible for non-thermal fading.

(a) Conduction
(b) Band gap
(c) El. Trap

Hole migration from luminescent centre (solid line) as an alternative to luminescent electron transition. (broken line)

Non-thermal release of electron from trap, by means of multi-phonon transitions.
(Ref: Hailey Phys. Rev. 840 1967)

Direct resonance transition between donor & acceptor centres, probability proportional to square of overlap of wave functions.
CHAPTER 2: ROUTINE PERSONNEL MONITORING WITH THERMOLUMINESCENCE DOSIMETERS IN AN N.H.S. HOSPITAL

2.1 Requirements for a hospital TLD service and related problems

During 1975-76 the Hospital Physicists' Association carried out a survey of radiation protection arrangements in United Kingdom N.H.S. hospitals. The replies gave detailed information about arrangements in 44 out of 59 centres responsible for this work. From this survey it appeared that the total number of individuals subject to personnel monitoring for occupational exposure in the N.H.S. was about 34,000. Of these approximately a quarter were issued with National Radiological Protection Board (N.R.P.B.) monitoring devices and the remainder were monitored both administratively and technically entirely by the N.H.S. protection centre responsible. The size of these centres varied considerably - the largest was responsible for over 3,000 workers and the smallest for only 68 (although the smallest centre operating their own dosimetry service catered for 153 workers.) From the replies to this questionnaire and in the light of what is known about the impending Health and Safety Executive requirements for the approval of dosimetry laboratories, the H.P.A. concluded that small scale services should either be operated technically by the N.R.P.B. or should be merged to reach an efficient minimum size corresponding to about 500 radiation workers monitored.

Administrative responsibility for all aspects of radiation protection in the N.H.S. is carried by graduate physicists who are termed the Radiation Protection Advisers (R.P.A.). At present the guidelines to which the R.P.A. works are contained in the 'Code of Practice for the Protection of Persons against Ionising Radiations arising from Medical and Dental Use'. Most of the physicists concerned are only a part-time R.P.A. and have various other duties but from the returns to the H.P.A. survey an attempt was made to estimate the necessary staffing for
radiation protection duties in terms of the number of 'full-time equivalent' professional physicists who would be needed to cover all the administrative and technical requirements of the Code of Practice. In compiling these returns technicians engaged in assisting the R.P.A. or in technical work associated with the protection service were rated on the basis of relative salaries as equivalent to a half-time graduate physicist. The analysis of the returns indicated that each full time equivalent R.P.A. (as defined above) was usually responsible for between 80 and 400 radiation workers, and from the associated comments it appeared that no R.P.A. should be expected to effectively exercise proper responsibility for controlling the exposures of more than 200 radiation workers. It also appeared that relatively few N.H.S. personnel (< 1%) are subject to neutron doses and that all whole body personnel monitoring is still carried out using film badges. Although a number of centres now have a TLD dosimeter reader which is used regularly, the most common application is for radiotherapy measurements and for radiation protection purposes its use is largely limited to finger dose measurements.

The author of this thesis is Radiation Protection Adviser to the Grampian Area Health Board in Scotland. Together with a colleague, he is responsible for the operation of a personnel monitoring service covering all N.H.S. radiation workers in north Scotland, (i.e. personnel employed by the Shetland, Highland and Grampian Area Health Boards.) This service is typical of the national arrangements discussed above, it currently monitors about 600 radiation workers which is very close to the average load per capita for such services as shown by the H.P.A. survey. Film badges are issued and returned at intervals of four weeks; these are supplemented by TLD finger dosimeters where appropriate (for about 5% of the staff monitored) and by fast neutron badges issued by the N.R.P.B. in the case of a small group of personnel working with fast neutron sources.

A survey of film badge doses recorded by N.H.S.
personnel monitored at Aberdeen in the past decade shows clearly that there are very few of the routine procedures in which appreciable doses are likely to be experienced. The largest group of monitored personnel comprises the diagnostic radiographers; in many small hospitals these are the only staff members subject to personnel monitoring. Although the shielding requirements for hospital X-ray rooms are much less stringent than those imposed for industrial radiography, and furthermore radiographers may frequently be using mobile X-ray units in open wards or theatres, their personal exposures normally remain extremely small. Significant radiation exposures only arise in diagnostic X-ray departments when radiologists and radiographers are engaged in carrying out fluoroscopic screening procedures. Similarly radiotherapy machines are so well shielded that they normally involve the operators in negligible radiation risks; doses to radiotherapy personnel can almost invariably be traced either to open isotope procedures or to the use on or in the patient of relatively small sealed sources of radioactivity. (A teletherapy treatment unit may contain eight kilocuries of cobalt 60, implanted sources or surface applicators do not often exceed 250 millicuries).

In the isotope department due to their specialised work load certain individuals regularly experience appreciable doses but for others this is extremely rare. Procedures which tend to be associated with unavoidable exposure include changing isotope generator columns, preparing radiopharmaceuticals, distributing prepared doses, and nursing duties involving attention to patients undergoing examination on a scanning machine or gamma camera. A review of the causes of irreducible dose commitments associated with the work of a hospital isotope department was compiled by the author in 1975 (1).

Another area where appreciable doses may be incurred is in the operating theatre, particularly where procedures requiring extensive use of X-ray fluoroscopic examinations are carried out. Cardiac catheterisation and angiography
may be mentioned as examples.

This leaves until last the group who consistently have highest exposures – nurses in the radium wards and radium theatres. Radium needles are now being gradually replaced by caesium equivalents (a change which usually helps reduce the doses received by hospital personnel) but are still very widely used. Gynaecological applicators containing sources of up to 200 mCi radium (or equivalent) are used regularly for cervical cancers and it has proved almost impossible to prevent nurses regularly engaged in caring for these patients from receiving doses typically of between 100 mrem and 400 mrem in a four week period.

The standard film badge has a response which varies little with photon energy for quanta above about 100 keV, but below this there is a rapid increase of sensitivity with decrease of photon energy due to the high atomic number silver bromide content. Radiation doses assessed from film badges are corrected for this by shielding parts of the film with metal filters of varying thickness and density so that the energy of the radiation falling on the film can be assessed from its penetration. By this means gamma ray doses can be measured with high accuracy and errors in evaluating soft X-ray doses where there is a wide spread of photon energies are usually also small. Unfortunately the converse often applies to doses arising in the isotope department where scanning procedures are most effectively carried out by using low photon energy gamma emitters such as \(^{125}\text{I}\), \(^{99}\text{Tc}^{m}\) and \(^{57}\text{Co}\) with gamma energies in the range 30 – 150 keV. Radiations from these are easily capable of penetrating from the organ under test to the surface of the body but are not so penetrating that efficient localisation of the isotope within the body becomes impossible and patient doses from the test become excessive. Unfortunately they span the range of energies over which the film sensitivity is varying most rapidly and consequently unambiguous dose measurements are most difficult. The situation is analogous to that in an
industrial plutonium processing laboratory where again low photon energies are encountered and it is difficult to avoid rather ambiguous dose measurements if film badges are used (2). In both these cases working conditions are such that the use of TLD for personnel monitoring may be very advantageous due to the much closer tissue equivalence of phosphors such as lithium fluoride or lithium tetraborate.

Because TLD personnel monitoring would show this clear advantage for at least one group of N.H.S. staff it was decided at the end of 1972 that a detailed evaluation would be made of the problems involved in changing from films to TLD in the Grampian region. This would also offer the further advantage that the many workers who routinely experience monthly doses of below 10 mrad could be provided with TLD badges that would be worn for three months before readout. The apparent background level recorded on film badges occasionally increases rapidly when these are issued for periods longer than four weeks (probably this is often a result of mishandling in hot and humid conditions). TLD dosimeters are much less troublesome in these respects.

A Conrad 5100B TLD reader was available at Aberdeen but up to this time had only been used occasionally for measurements of large doses in the radiotherapy range using sachets of lithium fluoride powder. For a personnel monitoring service it was felt that the complications of dispensing and handling large numbers of dosimeters of this type on a routine basis would be excessive and individual solid dosimeter elements should be used instead. Since sample trays designed for the LiF loaded teflon disc dosimeters manufactured by the Conrad company were available to fit our reader it was decided that this type of dosimeter would be chosen as the basis of the proposed new service. Discussions with physicists responsible for radiation protection arrangements in other hospital board areas showed that at this time very few medical centres in the U.K. were equipped with TLD readers and still fewer
were using TLD on a routine basis for low dose measurements. This position had changed little by May 1973 when the Hospital Physicists' Association organised a meeting of interested members at the National Physical Laboratory (N.P.L.). At the time of this meeting the organisers could only find three active users of TLD from within the N.H.S. to report on their experiences, and there was little agreement about the most satisfactory TLD system. The majority of hospitals were awaiting the report on the studies being carried out by the Department of Health & Social Security in conjunction with Harper at the University Hospital of Wales at Cardiff and Messrs. D.A. Pitman Ltd. of Weybridge before purchasing a TLD reader, and in the few centres where TLD measurements were in routine use the departments concerned had nearly all opted to work with powder dosimeters. Comments on attainable accuracies made at this meeting were very interesting. Rossiter (N.P.L.) reported on an I.A.E.A. sponsored intercomparison of doses at therapy levels from which it was concluded (on the basis of calibrations made with both Farmer sub-standard dosimeters and Fricke chemical dosimeters) that using LiF powder the overall uncertainty in TL measurements of above 200 rad could be reduced to ± 1.3%. Similarly Smith of Velindre Hospital, Cardiff, claimed that in the same range they were carrying out measurements with powder phosphors to an accuracy of ± 1.5% but that enormous discrepancies with solid dosimeters had led to these being abandoned as completely impracticable (3).

By the time of the N.P.L. meeting a very intensive study had been initiated at Aberdeen by the author in an attempt to determine the cause of the discrepancies arising in the use of teflon disc dosimeters and to investigate means by which these could be overcome. This chapter describes the investigations carried out and the results which were obtained.
2.2 Characteristics of LiF loaded teflon disc dosimeters

The discs used in this work were type D-LiF-N-0.4 supplied by Teledyne (Isotopes) Inc. of the U.S.A. through Messrs Pitman Ltd of Weybridge. The teflon matrix does not absorb an excessive amount of the light emitted by the LiF but due to its poor thermal conductivity produces a glow curve for which the resolution of separate peaks is rather poor (Figure 2.1). Teflon undergoes a structural change at about 327°C which results in the discs becoming more brittle and more transparent. This treatment also leads to them becoming more sensitive. Most users of these discs limit the maximum annealing temperature to 300°C to prevent this sensitivity change and discard any dosimeters accidentally heated beyond the critical point. Sometimes however all dosimeters are deliberately heated to 400°C both before use and in annealing between uses (Wallace at A.W.R.E. pioneered this technique). Annealing must then be carried out avoiding contact between dosimeters as the teflon will soften and they may fuse together. There is also an increased hazard of surface contamination or discolouration leading to arbitrary reductions in the sensitivity of individual dosimeters.

These teflon disc dosimeters are produced by moulding from a mixture of fine teflon powder and LiF powder with a mean grain size of about 18μ. They have a LiF content of about 30%, which is the maximum that is compatible with a uniform distribution and good mechanical properties. They are produced in the form of a cylinder which is sliced by a microtome into separate dosimeters 0.4 mm thick and weighing about 112 mg. In this work they were used with heating trays consisting of a solid metal base against which the dosimeter is pressed by a metal gauze. These give good thermal contact but absorb about 40% of the light. The discs therefore give about the same TL signal as 20 mg of loose powder. Since teflon is phosphorescent it is necessary that discs which have been exposed to light are left in the dark in the readout chamber for at
least 10 sec before readout commences.

Four factors may lead to sensitivity variations between individual discs:

(i) Variations in disc weight
(ii) Variations in phosphor loading
(iii) Sensitivity variation due to differences in the annealing
(iv) Variations in the optical density of the teflon.

Table 2.1 records the weights of 100 discs as supplied. It will be seen that their mass shows a standard deviation of better than 2%. The manufacturers claim that the LiF loading of the discs, particularly those supplied in the same batch, is extremely constant. This is not easy to check but tests on the sensitivities of a batch of new dosimeters did not agree with the assumption of uniform loading. The procedure adopted was to reject dosimeters whose response (defined as the mean of the readouts obtained for three standard doses of about a rad) varied by more than 10% from the average for the batch. About a quarter of the discs were rejected by this process - a very much higher percentage than would be predicted on the basis of the weight variations.

Variations in the optical density of the teflon were investigated by means of a film badge densitometer. Initially these were quite appreciable and they became still greater as repeated use of the dosimeters took place, in particular if a maximum annealing temperature of 300°C was exceeded. Even below this temperature the surface of the teflon was found to soften rendering the discs liable to discolouration from absorbed impurities. Table 2.2 shows measured sensitivities and densities for the batch of 100 new discs for which weights were tabulated in table 2.1. It can be seen that there is a considerable degree of correlation between the optical density and the sensitivity.

A further test was made on a batch of old discs which had not been subjected to optimum treatment throughout their
lives. It was found that their measured sensitivities varied over a much wider range than those of a batch of new discs. The optical densities of these discs were then measured and used as correcting factors for the observed sensitivities. In the majority of cases these were brought back nearly into agreement showing that the differences observed were due primarily to changes in transparency. These results are recorded in table 2.3.

A related problem is loss of sensitivity from discoloration due to dust on the surface being baked in when the teflon softens during high temperature annealing. This effect can lead to a 20% or more sensitivity loss after a disc has been recycled about 30 times. The problem is made more acute by the propensity of teflon to develop a static electric charge which attracts dust to its surface and makes proper cleaning very difficult.

The remaining factor in producing substantial sensitivity changes is the effect of annealing. The work described in the following sections shows that this is unquestionably the principal factor in causing large and unpredictable changes in the sensitivity of individual discs. With loose powder samples individual dosimeters are mixed together after readout and annealed as a single batch. Variations in sensitivity are thus averaged out in the bulk supply which is always thoroughly mixed before aliquots for the next batch of dosimeters are dispensed.

Consideration of the annealing effects discussed in section 1.4 shows that slight differences in the annealing temperature or time or, even more critically, in the rate of cooling will lead to marked sensitivity changes in the individual dosimeters.

Experience showed that in general users of solid dosimeters have not made sufficient allowance for these effects and when standard commercial annealing ovens are used there can be arbitrary sensitivity changes among a batch of dosimeters which may be so great that it becomes impossible to obtain reliable readings from them. Attempts to overcome this problem are described in section 2.5.
2.3 The Conrad 5100B TLD Reader *

This reader has been designed to operate with a 15 second readout cycle, controlled by a synchronous motor, during which the sample is heated for 10.5 seconds by a constant heater current which gives an exponential rise in temperature. The final temperature reached is governed by the setting of the heater current control. For reproducible results it is clearly necessary that the initial starting temperature of each readout cycle should be the same, i.e. that a second readout should not be initiated before the heating chamber has completely cooled following the previous heating cycle. The reader did not incorporate any means of monitoring the heating tray temperature but a special replacement sample slide incorporating a channel for thermocouple leads was designed and a chromel-alumel thermocouple was fed through this channel so that the heating tray temperature could be monitored during development work on the reader. Initially the thermocouple junction was welded to the underside of a spare heater tray but this proved unsatisfactory due to the A.C. heater current flowing through the tray interfering with the thermocouple signal. This arrangement was therefore replaced by one in which thermal contact between the junction and the tray was maintained through a silicone heat transfer compound designed for mounting semiconductors onto heat sinks. With this arrangement tests showed that if the interval between readings was longer than 40 sec the final temperature reached by the sample tray showed a standard deviation of only 0.8%.

The photomultiplier tube current during the readout cycle is integrated on a 1.0 μF capacitor the voltage across which is monitored by a digital voltmeter connected through an electrometer amplifier. The charge collected is held almost indefinitely without loss and can be reduced by a factor of ten reasonably accurately, this provides range

* Controls for Radiation, Teledyne Inc., 50 Van Buren Avenue, Westwood N.J.07675 U.S.A.
switching when high doses are to be read.

A standard light source could be loaded into the readout chamber in place of the TL sample, and its output integrated over a 15 sec readout cycle as a check on the gain and stability of the P.M. tube. However, this was constructed from a beta emitter compounded with a scintillator and when withdrawn from the readout chamber it was exposed to ambient illumination. This led to it emitting an additional phosphorescent light output which only faded slowly with a half life of about 10 minutes. To ensure reproducible results from this sensitivity check a new Cherenkov light source was prepared by encapsulating a high energy beta source (0.1 $\mu$Ci of $^{90}$Sr - $^{90}$Y) in a transparent potting medium (GE silicone compound Type RTV 602). Whilst it is possible that long term radiation damage effects might be observed with this new light source it was free of phosphorescence and its light output was found to remain extremely constant over periods of up to a month - amply covering the time between the calibration of a batch of discs and their eventual readout.

Most of the original electronic circuits in the reader operated satisfactorily but some modifications were necessary before accurate dosimetry was possible. The electrometer valve, the heating current control circuit components and the somewhat primitive servo-driven potentiometer type D.V.M. were all replaced. The new heating current control unit incorporated a ten turn potentiometer for precise resetting and also included facilities for the A.C. heater current to be monitored by the D.V.M. as well as by the original moving iron type pointer ammeter. Some component replacement in the integrating circuit was also found to be necessary and a potential divider was built into the E.H.T. supply so that this voltage could be reset with the D.V.M. to a higher precision than originally possible using the pointer meter provided.

The optimum heater current for use with teflon disc dosimeters type D-LiF-N-0.4 was determined by taking a
series of these discs exposed to a radiation dose of about 200 mrad and reading each one out twice in succession for a series of gradually increasing heater currents. The results of these measurements are plotted on figure 2.2. It will be seen that for low heater current settings only part of the TL signal stored in the phosphor is released on the first readout. Consequently this reading is low and the second readout is relatively high. As the heating current increases more efficient draining of the stored signal is achieved and the first readout settles to a plateau value whilst the second concurrently falls to a minimum. For excessive heater currents both readings start to rise again due to incandescent radiation from the heater tray. The optimum heater current to use is that for which the numerical value of the second readout is a minimum.

Experience with this modified reader showed that after it was correctly adjusted there was sufficient sensitivity to make measurements with the standard disc dosimeters down to a level of about 10 mrad with a background reading due to P.M. tube dark current, infra red radiation etc of below 10 mrad. (Higher background readings were associated with dirty or contaminated dosimeter discs.) At this point it was decided that routine dosimetry with these discs could be reliably introduced and the next section discusses some problems that were encountered.

2.4 Annealing and cleaning problems with routine use of LiF loaded teflon disc dosimeters

For the purpose of routine personnel monitoring at protection levels the requirements can be summarised as follows:

(a) It must be possible to purchase dosimeters of uniform sensitivity in sufficiently large batches to eliminate any need for individual calibration.

(b) The background reading from an unirradiated dosimeter should be small - preferably less than 10 mrad dose equivalent.

(c) The dosimeter sensitivity should be high enough to enable readings down to about 20 mrad to be made and
must be adequate to give reasonable accuracy for doses above 50 mrad.

(d) After readout it must be possible to restore the dosimeters to their original condition (or at least to a condition of uniform sensitivity) by some relatively simple thermal annealing process.

(e) Dosimeters must be capable of re-use for a reasonable number of times (depending upon their initial cost) before they must be discarded.

Clearly if sufficiently cheap disposable dosimeters were available requirements (d) and (e) above would no longer apply.

The earlier sections of this chapter have shown that dosimeter discs D-LiF-N-0.4 used with the Conrad 5100B TL reader modified as described readily meet requirements (a), (b) and (c) above. To provide an attractive alternative to the widely used film badges (which cost 4p each to purchase plus perhaps 1p for processing chemicals) a TLD personnel monitoring service must be economically competitive. It was clear therefore that at a cost of 80p each these discs were not yet cheap enough to regard as disposable and serious problems arose when they were annealed and re-used. Background readings as high as 200 mrad dose equivalent and arbitrary sensitivity changes by factors of between 0.5 and 2.0 were frequently encountered. The most serious factor influencing the sensitivity and background readings of these re-used discs was found to be surface contamination. The oven used for the high temperature annealing process had been previously used for other purposes. Traces of organic vapours such as those released during the heat drying of paints and enamels had contaminated the surfaces of the discs leading to very high background readings. To eliminate this the oven was run continuously for several days at 400°C with regular purging of its air content and was then fitted with a continuous stainless steel lining over the contaminated interior firebrick surfaces. These modifications considerably reduced the maximum background readings but variations in both
background and sensitivity were still encountered. These residual sensitivity changes were eventually found to arise from variations in the high temperature annealing process. The purpose of this procedure is twofold, firstly to ensure complete draining of all radiation energy stored in the crystal lattice and secondly to remove any enhanced sensitivity produced by total previous doses which extended into the supralinear region (about 800 rad upwards for LiF). The annealing procedure initially used had been 300°C for five hours as recommended by the disc manufacturers (who also state that after large doses annealing for 20 hours at 330°C may be necessary). Careful tests showed that with the equipment available it was impossible to ensure either that all the discs reached exactly the same temperature or that they could be cooled at the same rate under different ambient conditions. Martensson (4) reported that elimination of the high temperature annealing procedure for discs exposed only to protection level doses was a successful way of avoiding these problems. He claimed that the combination of a minimum time at the high temperature associated with the readout cycle followed by rapid cooling under completely consistent forced convection conditions in the readout chamber with nitrogen gas flowing through it, provided ideal conditions for maintaining peak sensitivity with the dosimeters. His observations showed that using this procedure the variation in sensitivity of individual discs during successive readouts could be kept within 3%. It was therefore decided to adopt this procedure and to discard any discs which accumulated total doses in excess of about 50 rad. This would not only avoid any problems from radiation sensitisation but should minimise the risk of appreciable errors arising from incomplete draining of the TL signal. As a further precaution discs recording doses in excess of 100 mrad were immediately put through a second readout cycle to ensure there was no residual signal. To stabilise the cooling rate of the readout chamber a flowmeter was fitted in the nitrogen supply line and all readings were taken with the same flow rate, care being
exercised to ensure that discs were not unloaded from the heating chamber before it had cooled for at least 60 sec.

Using this procedure the variations in sensitivity between successive readings taken with the same disc were greatly reduced - but not to the 3% level reported by Martensson. Discs treated in this way had a sensitivity about twice as high as the new discs supplied by the manufacturer (which had been annealed for 5 hours at 300°C followed by 24 hours at 80°C) whereas those cooled very slowly (at rates of the order of 1°C per minute) had their sensitivity reduced to about 65% of its original value.

Attention was next given to the abnormally large background readings (up to ~100 mrad) obtained from some discs. This was found to be a triboluminescent signal resulting from ultrasonic cleaning of the discs between readout and re-exposure. It was strongly time dependant and faded almost completely within a few days. It was made more stable and appreciably larger if active cleaning agents such as 'Decon' or 'Labrite' had been added to the water in the ultrasonic bath, but could always be eliminated by putting the discs through one (or occasionally two) readout cycles between readout and exposure. At a later stage it was observed that a solvent such as propyl alcohol gave more satisfactory results in the ultrasonic cleaning bath than distilled water.

The residual sensitivity changes proved very difficult to overcome. After many tests it was shown that these could be attributed to variations in the low temperature annealing process and they were eliminated if low temperature annealing was abandoned. However this change had a very adverse effect on the associated glow curve. Lin and Cameron (5) had shown that substantial changes in the glow curve resulted from changes in the low temperature annealing procedure, (Figure 2.3a). Without an 80°C anneal there is a large low temperature peak which is very subject to fading. The annealing serves an essential function in restructuring the electron trap distribution to give a glow curve suitable for practical dosimetry. The restructuring
is associated with regrouping of the impurity Mg activator atoms. Following the readout process these exist as dipoles produced by a substitutional Mg atom adjacent to a Li ion vacancy. During the low temperature annealing these dipoles diffuse to form trimers consisting of rings of three dipoles in the 111 planes (Figure 2.3b). The dipoles are associated with the emission of the light forming the low temperature peaks (peaks 1 and 2) of the LiF glow curve, the trimers are related similarly to peaks 4 and 5 - the required dosimetry peaks.

The need for precise control of this low temperature annealing process if the sensitivity of each individual dosimeter is to be the same has been shown by the work of Webb (6) who reported that a 1°C variation in the temperature of the disc during annealing led to a sensitivity change of 5%. Consequently to improve the precision of this process a special tubular stainless steel disc holder similar to that used successfully by Docherty & Marshall (7) was constructed, (Figure 2.4). Inside this the discs were packed between two lengths of teflon rod held together by pressure from weak stainless steel springs. It was assumed that the centre of the teflon column so formed would be held at a uniform temperature throughout both the annealing and the associated cooling.

Experience showed that two problems still arose with this new arrangement:

(i) Used discs are in general slightly concave. This leads to variations in the heat conductivity between adjacent discs in the tube spoiling the uniformity of temperature required.

(ii) The tube wall had absorbed oil during the reaming carried out when it was constructed. Despite repeated baking and ultrasonic cleaning cycles traces of this oil continued to diffuse out and contaminate the edges of the discs which were annealed in it.

Two conclusions were finally drawn from the above work:

(a) Almost all the discrepancies reported by workers using LiF in teflon dosimeters were probably due to slight
variations in thermal treatment between individual discs. This problem does not arise with the use of powdered phosphors because individual aliquots are mixed back into the bulk supply after use and a mean sensitivity measurement is made for this supply at the time the next set of individual dosimeters is being dispensed.

(b) Elimination of this effect using recommended annealing procedures would involve the use of exceptionally accurate stabilised ovens operated under closely controlled ambient conditions. This would probably cost as much or more than the actual TLD reader. Such a solution would not be economic as far as the small scale user (who would gain most from the use of disc dosimeters in preference to a powder phosphor) is concerned. It appears that these users would benefit more by utilizing the very precise temperature control built into their reader sample chamber to give them consistent annealing conditions. Although this would reduce the maximum throughput of dosimeters attainable, few small scale users would be operating their TLD reader at saturation level so this would remain the most economical solution to the problems of annealing. This possibility is examined in the next section.

2.5 Modification of Conrad 5100B TLD reader to incorporate auto-annealing facilities

As designed the Conrad 5100B reader integrates the entire TL output from the sample up to the maximum temperature reached. It is assumed that where phosphors such as LiF exhibit low temperature peaks susceptible to fading, annealing procedures which restructure the glow curve and eliminate these low temperature peaks will be followed. An alternative method of eliminating the effect of the fading of low temperature peaks is to reject the readout signal they generate. Many commercial readers use this method - they incorporate two adjustable temperature limits between which the light signal from the phosphor is
recorded but outside which it is rejected. In this way
the signal from a well defined peak of a composite glow
curve can be recorded independently of all other peaks.
Unfortunately the low thermal conductivity of teflon makes
this method unsuitable for use with LiF-teflon dosimeters
as, except for very slow heating rates, different parts of
the disc will normally be at different temperatures and the
separate peaks in the glow curve will not normally be
well resolved during readout. The alternative method which
is more suitable for the LiF-teflon disc dosimeters, is to
use a low temperature post-irradiation anneal (a pre-heat)
to eliminate the energy stored in the low temperature peaks
before readout of the main peak commences. With this
method the linear sample heating rate desirable for studies
of glow peak structure must be replaced by a three or
preferably four step heating process:

(i) Samples should be raised as quickly as possible
to a suitable pre-heat temperature for draining
the low temperature peaks and held at this tem-
perature for whatever time is necessary to com-
plete this process (primarily a function of the
thickness of the dosimeters).

(ii) Sample temperature should then be raised as
quickly as possible to the readout value and held
at this value for a sufficient time for complete
draining of the main peak to take place.

(iii) Facilities should then be incorporated for
further heating of the sample to the maximum
safe temperature; between 300°C and 320°C for
teflon discs. This temperature must be main-
tained for long enough to ensure complete draining
of any residual signal.

(iv) The sample should then be left to cool under con-
trolled conditions in the readout chamber, at
least until its temperature has fallen to below
100°C. For this purpose one modern reader
(the Pitman 'Toledo') incorporates electronically
controlled linear cooling of the readout chamber
but forced convection cooling with a controlled
constant gas flow through the chamber provides a
reasonable alternative to this.
The advantages of this type of heating cycle have been discussed by Webb and Phykit (8), and the required temperature time relationship is shown on figure 2.5a.

For dosimeters not normally exposed to doses in excess of 200 mR stage (iii) of the heating cycle may be unnecessary and may merely lead to an undesirable increase in the readout time required for each dosimeter. It will therefore be advantageous if this part of the cycle can be disabled at the discretion of the operator. In this case care must be taken not to mix dosimeter discs for which different readout cycles were followed as these will emerge with different sensitivity and will have to be calibrated separately.

Readers designed to follow this type of heating cycle invariably use a temperature signal to control the heater circuit, whereas the Conrad 5100B reader does not incorporate any temperature monitoring facility. Instead the readout cycle is controlled by a 4 r.p.m. synchronous motor fitted with sets of cam operated contacts to initiate and terminate the various operations. The heater is supplied from a constant current generator connected through a set of contacts operated by one of the cams. Three other cams are necessary to fully programme the complete readout cycle. One switches off the motor at the end of the readout cycle, one controls the period over which the TL signal is integrated and one discharges and resets the integrating capacitor before signal collection commences. A consequence of this method of reader operation is that modifications to the heating cycle are not readily made. Nevertheless in view of the problems encountered with external annealing techniques it was decided to attempt to redesign the reader to give a suitable four part cycle similar to that discussed above. A ten cam Honeywell programme unit was built into the reader in the space formerly occupied by the servo type D.V.M.: this was driven from a 1 r.p.m. synchronous motor allowing up to 60 sec for the complete new readout cycle. A drum on the programmer shows the
elapsed time from the start of the cycle and the unit was placed so that this was visible through the window previously used to read the old D.V.M. This gave a visual indication of the state of the readout cycle. Instead of the original single heating period the new readout cycle utilised six separate periods, each brought in by a different programmer cam.

The first heating period brings the sample tray up to preheat temperature, but after the heating current is disconnected the tray temperature begins to fall exponentially instead of being held at a plateau value as in a temperature controlled reader. A second short heating burst in the middle of this 'hold' period can be adjusted so that the temperature variation during the period is kept to a minimum and is very small. The procedure is then repeated to bring the sample tray to readout temperature and to hold this whilst the stored signal is completely drained. Finally a further repetition of this process comprises stage (iii) of the cycle - a machine anneal of the dosimeter which has been read out. A complete readout cycle thus comprises six distinct heating periods, each controlled by contacts on a separate individually adjustable cam, which give a resultant time-temperature relationship for the new heating cycle as shown on figure 2.5b. Curve A of figure 2.5b shows the original temperature time relationship with the heater current adjusted to the optimum value for readout of LiF-teflon discs. Curve B shows the new four part heating cycle as finally adjusted and curve C shows this same cycle with the final period of high temperature annealing disabled. Curves B and C were plotted for the same heater current as curve A. The maximum temperature reached by the heating tray in the original readout cycle was 240°C. With the new readout cycle the preheat temperature was 130°C, the readout temperature was 237°C and the final anneal temperature was 305°C. The period during which the TL signal is integrated can be reduced with the new heating cycle from the previous value of 14.5 sec to 13 sec thus giving a small reduction in the background
signal from P.M. dark current.

To test the reproducibility of the new heating cycle and programmer a set of temperature time curves were plotted for a series of equi-spaced heater current settings corresponding to settings of the ten turn potentiometer dial of between 780 and 880. These are shown on figure 2.6 and indicate that the precision of time control of the Honeywell programmer is adequate to give completely reproducible readout cycles. The heating periods, which are listed in table 2.4, have been adjusted to give the closest possible approach the optimum readout temperature for the use of D-LiF-N-0.4 dosimeters. At lower heater current settings the temperature rises during the readout "plateau", and at higher currents it falls slightly.

To complete the reader modifications a small timer, adjustable over the range 1 - 4 min was built into the instrument and used to operate a red light over the sample loading slide. This gave warning of the elapsed time necessary for the dosimeter discs to cool down to a temperature of below 100°C so that controlled cooling was not interrupted by premature unloading of the sample tray. In practice it was found that a complete readout cycle, including sample cooling, occupied a time of 1.6 min when the nitrogen flow rate was set to 3 l/min and the warning light was therefore set to operate for this period.

2.6 Evaluation of modified Conrad 5100B TLD reader

When the modifications and adjustments described in the preceding sections had been completed it was found that the background reading obtained from a properly prepared unirradiated disc was typically 1-2 mV and was due largely to the P.M. tube dark current contribution. At these same settings D-LiF-N-0.4 disc dosimeters gave a TL signal corresponding to about 0.25 mV per mrad. Hence the background reading was equivalent to doses of between 4 and 8 mrad whilst dose readings of 20 mrad upwards could be readily made, (Figure 2.7).

Tests were made on fading of the stored signal using
both the new and original readout cycles with no external annealing of the discs. Checks were also made on the extent to which the main peak was affected during the pre-heat period with the new heating cycle, and on the maximum dose that was fully drained during a single readout cycle. Further tests were then carried out to determine to what extent sensitivity changes in a group of initially equivalent dosimeter discs still took place, and to investigate the effect of ambient light on disc dosimeters annealed and read out on the modified reader. These various tests are described in the remainder of this section. Table 2.5 shows the results of the fading tests. It is clear from these results that using the new readout cycle no substantial problems from fading will arise during the maximum period personnel monitoring devices are likely to be worn before readout.

The checks on the effect the pre-heat has on readout of the main peak were lengthy as the effects of cam adjustments to make minor changes to the heating cycle were unpredictable and thus very time consuming. However a series of readout cycles with as nearly as possible the same readout temperature and duration, but with different pre-heat temperatures were eventually evaluated.

The results are shown on figure 2.8 from which it can be seen that the variation of light output with pre-heat temperature averages about 1% per °C over the range from 115°C to 145°C and is least for a value of 130°C. This implies that draining of the low temperature peak without serious attenuation of the main peak is most efficiently realised at this pre-heat temperature so the cams were finally set to give this value.

Figure 2.9 shows the pre-heat, read and anneal zones marked against the recorded glow curve for a disc following instrumental annealing only. Table 2.6 shows how completely doses of various magnitude are read out using the new heat cycle. It can be deduced that for doses of up to 1 rad no error is introduced by putting a disc through one readout cycle only before re-use, but for
doses in excess of this two successive readout cycles were normally followed to ensure complete thermal draining of the stored signal.

The sensitivity variation for a disc following instrumental annealing only is shown in table 2.7 and was felt to be quite acceptable. Table 2.8 records observations on ambient light sensitivity and shows that this effect can still be one of the most important contributions to background signal. It is felt that it would be helpful if the plastic sachets supplied for holding the dosimeters and attaching them to fingers etc were fitted with an opaque black window instead of the transparent one supplied at present.

In general spurious light induced signals were not observed following exposures to artificial light or interior daylight of less than five minutes and no discs were found to have light sensitivity sufficiently high to necessitate loading of the sample into the reader being carried out in subdued light. Although comparable figures had not been obtained, the sensitivity to ambient light had been found more troublesome before the reader was modified to incorporate the pre-heat facility. This may however have been due to inadequate appreciation of the importance of phosphorescence in teflon previously discussed in section 2.2.

2.7 Intercomparison of personnel monitoring with film badges and TLD discs in a hospital environment

Badges for personnel monitoring with TLD discs differ completely in principle from those used with film detectors. For the latter because of the variation in sensitivity which is shown on figure 2.10, the film holder incorporates a series of filters which attenuate different energy radiations to differing extents. The aim in evaluating the film is to estimate both the radiation energy and the skin dose. Doses to other organs can then, if necessary, be calculated from this data. TLD badges on the other hand measure the integral skin dose from all energies
directly. Ideally they should be covered by a layer of plastic 7 mg/cm² thick so the measured dose corresponds with that given to the basal layer of the skin. Additional dosimeters can then be provided covered by suitable thickness absorbers to give direct readings of the male gonad and mean bone marrow or female gonad dose (the latter require approximately equal filtration). The optimum filters for this purpose have their composition tabulated in table 2.9. (15)

A simpler form of TLD badge, originally designed by Perry at Winfrith, has been more widely adopted in this country. It uses two discs only: one to record skin dose and the other to give an upper limit to either gonad or bone marrow dose. Dr. Perry kindly sent a sample of one of his badges and the specifications for the filters over the two discs so arrangements were made for a supply of badges to his specification to be moulded from high density polythene for use in personnel monitoring in the Aberdeen Royal Infirmary. One of the badges produced for this purpose and currently in use at Aberdeen is shown on figure 2.11. The filters covering the two dosimeter discs consist of 7 mg/cm² mylar and 1 mm aluminium respectively.

Before implementing a change over from film badges to TL dosimeters it was felt necessary to conduct a field trial to ensure that no serious loss of accuracy would arise under the conditions of use concerned. A number of other workers had already carried out such intercomparisons under various conditions of use e.g. Johnson 1967(9), Hall 1967(10), Suntharalingam 1966(11), Mather 1968(12), Schayes 1968(13), and Eastes 1968(14). Although these all tended to show that both methods are capable of consistent and accurate readings under a wide variety of conditions none had been conducted in an establishment comparable with a N.H.S. hospital and several points of difference require consideration when a change-over is contemplated. Some of these were referred to briefly in section 2.1. For example film badges have the advantage that once processed a permanent record is available from which doses can be re-evaluated a number of times if necessary. So far the TL glow curves have not normally been regarded as equally satisfactory in this respect and film badge records
have been preferred by administrators such as the Factory Inspectorate. Conversely films have to be processed when reasonably fresh so integrated doses cannot be accumulated for periods greater than about four weeks. They are more likely to be affected by adverse ambient conditions (especially accidental immersion) and are very subject to erroneous dose evaluation where low energy photons are concerned. Figure 2.10 shows the extent to which Kodak personal monitoring film has an enhanced energy response for low energy photons. The importance of this effect was demonstrated during a comprehensive study of TLD personnel monitoring methods carried out by Brunskill at Windscale. During this study film, LiF and Li$_2$B$_4$O$_7$ TLD dosimeters were intercompared in two different areas - a general area where exposure was to a wide range of mixed beta and gamma radiations and a plutonium area where the principal dose was from low energy photon radiation emitted by the plutonium (39 and 52 keV). The results showed that all three methods normally gave very reasonable agreement in the general radiation area but that in the plutonium area whilst readings of the two TLD dosimeters correlated very closely, those of the film badges showed enormous discrepancies from the TLD readings and were almost certainly unreliable.

The Aberdeen Royal Infirmary is a large general hospital in which a very wide range of diagnostic tests are carried out using ionizing radiations from both X-ray sets and radioactive isotopes. The latter typically involve photon energies of 150 keV or less since, as explained in section 2.1, radio-isotopes administered to patients are normally selected from low energy gamma emitters which give a smaller radiation dose and can be localised more accurately on scanning machines (e.g. $^{125}$I emitting a 35 keV photon, $^{57}$Co, a 120 keV photon, $^{99}$Tc$^m$ a 140 keV photon etc). It appeared from Brunskill's report that the use of TLD for personnel monitoring in these diagnostic departments would be likely to prove particularly advantageous. It was therefore decided that the intercomparison of film
badges and TL disc dosimeters would be carried out with groups of workers drawn from three different departments:

(a) The radiotherapy department where the principal doses arise from radium handling and are due to mixed high and low energy gamma radiation.

(b) The diagnostic X-ray department where doses are due to low energy X-rays with a continuous spectrum of photon energies.

(c) The nuclear medicine department where doses arise from handling radio-isotopes most of which emit low energy monochromatic gamma quanta.

Such an intercomparison is inevitably a lengthy process - only a few doses of above reporting level (20 mrem) are recorded during each four week period. Consequently only a limited number of readings have been obtained so far and the intercomparison programme is still being carried on. Results to date are shown on the intercomparison graph which forms figure 2.12. In practice few large doses arise and many of the available points plotted come into the region of appreciable uncertainty at the bottom end of the dose scale. This has tended to mask the differences between the two systems.

To ensure that the two dosimeters were not separated and exposed to different doses and also to avoid asking individuals participating in the intercomparison to wear two different badges this experiment was conducted by covering the TL disc with a thin polythene sheet and securing it to the back of the film badge with a piece of PVC tape in such a way that it would lie under the open window of the conventional film badge holder when the film was inserted into the badge holder correctly (Figure 2.13).

Both with the standard TLD personnel monitoring badge previously described and with TL discs attached to the back of a film for insertion into a film badge holder, it is necessary to consider the effect of variations in the angle of incidence of the radiation falling on the disc. This has been investigated and the conclusions are reported
in chapter 6 where it is shown that there is a falling off in response with increasing angles of incidence. The same uncertainty arises in the case of film. However an AERE report (16) which studied this effect in greater detail concluded that the variations in angular response do not follow any well defined pattern and that angular corrections cannot readily be evaluated. For the purpose of this intercomparison angular effects have been ignored. Any correction required would be most unlikely to be large enough to significantly modify the experimental results shown on figure 2.12.

It is difficult to draw extensive conclusions from the number of readings which have been collected so far and in particular this is not large enough to usefully separate the results for the three groups of individuals involved. However the following comments appear justified:

(a) As expected from Brunskill's work considerable differences between TLD disc dosimeters and film badges are encountered in a hospital environment.

(b) A substantial number of readings show appreciable TL doses of up to about 40 mR although no dose was recorded by the film. It appears likely that some of these TL doses were spurious - one possible explanation would be the issue of discs which had accumulated some surface contamination.

(c) In general TL doses exceeding film doses have been observed more often than the reverse. This is probably due to the effective background for the discs being greater than the 10 mR measured for prompt readout of relatively new discs. The discrepancy can be accounted for by assuming that under the conditions of the survey the effective background of the discs had an average value closer to 20 mR.

(d) It can be concluded that within the limits of accuracy required for a personnel monitoring service both TLD and film badges give acceptable results. The greater resistance of TLD discs to accidental heating or immersion, combined with their more uniform energy response must be balanced against their higher cost.
and a tendency to show slightly higher background readings.

2.8 Conclusions

It is clear that a TLD personnel monitoring service for whole body exposures can be operated successfully on the basis of the experimental procedures discussed in this chapter. The extent to which it would be advantageous to use TLD badges rather than film badges for an N.H.S. monitoring service is less easy to decide. A cost comparison would only become possible after such a service was initiated. Factors which preclude precise advance costing include:

(a) The proportion of lost or damaged TLD discs cannot readily be estimated in advance although experience with film badge holders suggests that the cost of replacement TL dosimeters would probably be greater than the cost of films.

(b) Film processing chemicals and bottled nitrogen gas might well roughly balance in cost.

(c) The labour costs associated with either system would depend greatly upon the extent to which it could be automated, but would appear likely to be somewhat less with a TLD system.

These various factors are too complicated to permit any realistic assessment of their relative effect and furthermore material costs at the present time are extremely unstable. Any decision to change to the TL system would probably therefore have to be based on other considerations. Some of these are tabulated overleaf.
Advantages of changing to TL monitoring system

(1) Dose assessment for low energy gamma radiation would be improved
(2) TL dosimeters could be issued for longer periods than four weeks where exposures are small
(3) TL dosimeters are less sensitive to extreme environmental radiation sources and mental conditions (including used at different times, becoming wet).

Advantages of continuing to use film badges

(1) Processed films can be stored and doses received redetermined at a later date if necessary
(2) An exposed film badge can have 'diagnostic' value in indicating possible errors in working practice made by the operator – particularly if operator – particularly if
(3) TL dosimeters are less sensitive to extreme environ-regular operation of various radiation sources are
mental conditions (including used at different times.

Any decision cannot be made purely on scientific and economic grounds - administrative requirements must also be considered. In the near future all laboratories intending to carry out personnel monitoring may have to obtain the prior approval of, or at least to meet the standards set by, the British Calibration Service (now under the control of the National Physical Laboratory). The Health and Safety Executive at present only accept film badge monitoring services for measurements of personal radiation exposures conducted in accordance with the requirements of the Factories Act, but services based on TL monitoring are now for the first time permitted to apply for recognition (summer 1976) and it is expected that approval of the first TL monitoring service will be given within the next few months. It seems likely that this approval may be conditional upon a TL glow curve, being retained as an alternative to the processed film. Any such requirement for routine glow curves must in turn affect the relative costs of the two systems.

The present intention at Aberdeen is to seek approval for a TL monitoring service rather than of the existing film badge service. Nevertheless it is expected that supplementary film badges will still be issued with TL dosimeters in cases where this is felt to be justified by their diagnostic value.

Further chapters of this thesis report other work relating to the use of solid state dosimeters for non-routine special investigations associated with the work of the N.H.S.
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2.11 SUNTHARALINGAM, N. Health Physics 12 1595 1966

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2.13 SCHAYES, R. Health Physics 14 251 1968


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### Table 2.1
Weights of 100 TLD discs

<p>| | | | | | | | | | | |</p>
<table>
<thead>
<tr>
<th></th>
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<td>109.7</td>
<td>112.0</td>
<td>108.9</td>
<td>109.9</td>
</tr>
</tbody>
</table>

Mean weight $\bar{x} = 109.9$ mg

Standard deviation

$$
\frac{\sum(x - \bar{x})^2}{N} = 1.14 \text{ mg}
$$

The percentage standard deviation in the weights of a batch of 100 new discs is therefore

$$\frac{1.14}{109.9} \times 100 \% = 1\%$$
TABLE 2.2.

**PROPERTIES OF 100 NEW LIP-TEFLON DOGMEETER DISC TYPE D-LIP-U-C-G**

<table>
<thead>
<tr>
<th></th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>D</th>
</tr>
</thead>
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<td>1</td>
<td>423</td>
<td>493</td>
<td>455</td>
<td>298</td>
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<td>2</td>
<td>643</td>
<td>540</td>
<td>647</td>
<td>851</td>
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<tr>
<td>3</td>
<td>0.32</td>
<td>0.31</td>
<td>0.36</td>
<td>0.32</td>
</tr>
<tr>
<td>4</td>
<td>172</td>
<td>170</td>
<td>170</td>
<td>170</td>
</tr>
</tbody>
</table>

All readings made for dose of 600 mR with the reader set to give an expected output signal of 1 digit per mR dose.

**For first reading:**  
Mean = 528, Standard deviation = 96  
Percentage standard deviation = 54.15%  

**For second reading:**  
Mean = 565, Standard deviation = 49.9  
Percentage standard deviation = 8.8%  

**For optical density:**  
Mean = 0.32, Standard deviation = 0.024  
Percentage standard deviation = 7.5%  

**For second reading multiplied by optical density:**  
Mean = 181, Standard deviation = 9.54  
Percentage standard deviation = 5.3%  

**NOTES**  
The wide spread in initial reading values is largely attributed to spurious U-V induced signals of up to about 200 mR dose equivalent. The rapid cooling associated with an instrument may appreciably double the TL sensitivity of the discs. Without the U-V enhancement of their response the first reading from the discs would be likely to have a mean value of below 500.

The reduction in standard deviation to only 5.3% shown in the final set of figures indicates that even with new discs variations in optical transparency make a for greater contribution to the variations of dosimeter sensitivity than non-uniformity in the weight of the discs.

**TABLE 2.2 - See also opposite.**
### Correlation of Sensitivity of Soiled Dosimeter Discs for Loss of Optical Transparence

Reader sensitivity set to nominal value 1 digit per mm.
16 disc samples chosen from dosimeters which had all been in regular use for 3 years and which showed marked discoloration.

Readout values below for beta dose (90 Sr) of 3 rad.

<table>
<thead>
<tr>
<th>Readout</th>
<th>2020</th>
<th>3000</th>
<th>2720</th>
<th>2810</th>
</tr>
</thead>
<tbody>
<tr>
<td>Measured O.D.</td>
<td>0.72</td>
<td>0.50</td>
<td>0.62</td>
<td>0.64</td>
</tr>
<tr>
<td>Product: Readout X O.D.</td>
<td>1440</td>
<td>1500</td>
<td>1744</td>
<td>1616</td>
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</tbody>
</table>

<table>
<thead>
<tr>
<th>Readout</th>
<th>2830</th>
<th>3390</th>
<th>3920</th>
<th>3180</th>
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</thead>
<tbody>
<tr>
<td>Measured O.D.</td>
<td>0.74</td>
<td>0.50</td>
<td>0.63</td>
<td>0.47</td>
</tr>
<tr>
<td>Product: Readout X O.D.</td>
<td>1700</td>
<td>1690</td>
<td>1690</td>
<td>1490</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Readout</th>
<th>3760</th>
<th>3600</th>
<th>2550</th>
<th>4150</th>
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</thead>
<tbody>
<tr>
<td>Measured O.D.</td>
<td>0.64</td>
<td>0.59</td>
<td>0.67</td>
<td>0.43</td>
</tr>
<tr>
<td>Product: Readout X O.D.</td>
<td>1500</td>
<td>1680</td>
<td>1510</td>
<td>1780</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Readout</th>
<th>3150</th>
<th>2350</th>
<th>2650</th>
<th>3340</th>
</tr>
</thead>
<tbody>
<tr>
<td>Measured O.D.</td>
<td>0.54</td>
<td>0.63</td>
<td>0.66</td>
<td>0.48</td>
</tr>
<tr>
<td>Product: Readout X O.D.</td>
<td>1700</td>
<td>1490</td>
<td>1490</td>
<td>1600</td>
</tr>
</tbody>
</table>

Mean values:
- Readout 2799 with standard deviation 560 (20%)
- Measured O.D. 0.56 with standard deviation 0.08 (14-5%)
- Corrected readout 1615 with standard deviation 166.9 (9-75%)

### Conclusions
With use the optical density of the discs almost doubles although the dosimeters still retain acceptable sensitivity. If the readout from the discs is corrected for the variations in transparency which have developed, the standard deviation in their sensitivity is reduced to less than half the uncorrected value and they then remain capable of giving readings of acceptable accuracy for most general purposes even after several years of regular use.
TABLE 2.5.

Fading of LiF-terfon disc dosimeters following reader annealing only.
Tested in batches of four. Results expressed as percentage of
prompt readout value.

<table>
<thead>
<tr>
<th>Original reader heating cycle.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Time before readout.</td>
</tr>
<tr>
<td>------------------------</td>
</tr>
<tr>
<td>1 week</td>
</tr>
<tr>
<td>1 month</td>
</tr>
<tr>
<td>3 months</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Modified reader heating cycle.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Time before readout.</td>
</tr>
<tr>
<td>------------------------</td>
</tr>
<tr>
<td>1 week</td>
</tr>
<tr>
<td>1 month</td>
</tr>
<tr>
<td>3 months</td>
</tr>
</tbody>
</table>

TABLE 2.6.

COMPLETENESS OF ERADING OF TL SIGNAL FROM DOSIMETER DISCS WHEN USING INSTRUMENT ANNEX ONLY.

<table>
<thead>
<tr>
<th>Radiation dose</th>
<th>Heater current setting</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 krad.</td>
<td>140 mR. 104 mR. 44 mR.</td>
</tr>
<tr>
<td>100 rad.</td>
<td>93 mR. 5 mR. Background</td>
</tr>
</tbody>
</table>

The three heater current settings above correspond to readout temperatures of 210°C, 235°C and 260°C respectively and to anneal temperatures of 252°C, 300°C and 322°C.

It can be concluded that for doses of less than 10 rad the normal readout cycle leaves negligible stored TL signal in the dosimeter discs.
TABLE 2.7.
Sensitivity Changes with LiF-teflon Disc Dosimeters during repeated use.

Twenty successive readouts made 24 hours after exposure to beta ray dose of 100 mR, for five different dosimeter discs.

<table>
<thead>
<tr>
<th>Reading</th>
<th>Disc 1</th>
<th>Disc 2</th>
<th>Disc 3</th>
<th>Disc 4</th>
<th>Disc 5</th>
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<tr>
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<td>89</td>
<td>106</td>
<td>112</td>
<td>103</td>
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<td>2</td>
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<td>103</td>
<td>117</td>
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</tr>
</tbody>
</table>

Mean 91.5 90.0 104.0 110.0 103.0

Standard Deviation 3.0% 3.0% 3.2% 3.2% 4.4%

The fact that no progressive sensitivity change can be detected over twenty readout cycles can be clearly seen from a graphical representation of these readings.

Graphical Representation of Data on Table 2.7.
TABLE 2.8.

Sensitivity of LIF-teflon TL dosimeter discs to ambient illumination.

A Readout of 20 undosed discs following 1 year storage under normal conditions of laboratory illumination.
B Readout of same undosed discs following 12 hours exposure to light at 40cm from fluorescent lamp only.
C Readout of same undosed discs following 12 hours exposure to normal daylight only.

All readings expressed in mSv dose equivalent.

<table>
<thead>
<tr>
<th></th>
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<td>104</td>
</tr>
<tr>
<td>2</td>
<td>115</td>
<td>30</td>
<td>22</td>
</tr>
<tr>
<td>3</td>
<td>274</td>
<td>34</td>
<td>16</td>
</tr>
</tbody>
</table>

<table>
<thead>
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<th></th>
<th>A</th>
<th>B</th>
<th>C</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>133</td>
<td>618</td>
<td>43</td>
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<tr>
<td>2</td>
<td>135</td>
<td>107</td>
<td>43</td>
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<tr>
<td>3</td>
<td>17</td>
<td>181</td>
<td>22</td>
</tr>
</tbody>
</table>

<table>
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<tr>
<th></th>
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<th>C</th>
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<tbody>
<tr>
<td>1</td>
<td>79</td>
<td>148</td>
<td>46</td>
</tr>
<tr>
<td>2</td>
<td>68</td>
<td>18</td>
<td>14</td>
</tr>
<tr>
<td>3</td>
<td>166</td>
<td>20</td>
<td>14</td>
</tr>
</tbody>
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<th></th>
<th>A</th>
<th>B</th>
<th>C</th>
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</thead>
<tbody>
<tr>
<td>1</td>
<td>117</td>
<td>121</td>
<td>116</td>
</tr>
<tr>
<td>2</td>
<td>47</td>
<td>27</td>
<td>19</td>
</tr>
<tr>
<td>3</td>
<td>48</td>
<td>24</td>
<td>13</td>
</tr>
</tbody>
</table>

NOTES:
With most discs the spurious signal developed during prolonged storage builds up to a maximum value substantially grealer than that resulting from 12 hours exposure to light although in a few instances (e.g. for disc no. 2) this does not happen. A majority of the discs showed approximately the same spurious signal after 12 hours exposure to either daylight or artificial light, but three discs (numbers 1, 6 & 11 above) which showed a very much higher level of sensitivity in both cases, were twice as sensitive to daylight as to the fluorescent light. There must be some change in the physical state of the LIF in these discs which gives them both their particularly high sensitivity and their very different response to the two types of illumination. It is possible that structural changes which could account for these observed effects could be identified by studying the spin resonance response of the discs on an ESR or NMR spectrometer.

TABLE 2.9

Optimum filters for three element TLD personnel monitoring badge utilising three lithium fluoride-teflon dosimeters.

<table>
<thead>
<tr>
<th>Element</th>
<th>Si</th>
<th>Fe</th>
<th>Cu</th>
<th>Mn</th>
<th>Mg</th>
<th>Cr</th>
<th>Zn</th>
<th>Tl</th>
</tr>
</thead>
<tbody>
<tr>
<td>Accident filter</td>
<td>0.5%</td>
<td>0.7%</td>
<td>1.0%</td>
<td>0.3%</td>
<td>2.5%</td>
<td>0.3%</td>
<td>5.0%</td>
<td>0.2%</td>
</tr>
<tr>
<td>Routine filter (2)</td>
<td>0.5%</td>
<td>0.7%</td>
<td>0.2%</td>
<td>0.1%</td>
<td>0.1%</td>
<td>0.1%</td>
<td>0.3%</td>
<td>0.16%</td>
</tr>
<tr>
<td>Balance in both cases aluminium</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Skin filter (3) 7 mg per sq cm mylar film.

Note:
(1) Accident filter gives female gonad and mean bone marrow dose.
(2) Routine filter gives male gonad dose (upper limit to population genetic dose contribution
(3) Skin filter gives dose to germinai epithelium.

Total thickness of accident filter 1.25 mm. Dosimeter reading to be multiplied by 1.1 to give true organ dose (correction factor for 1 MeV photons if dosimeter discs are calibrated to read Roentgen in air) Total thickness of routine filter 1.0 mm. Corresponding correction factor to give true organ (bone marrow) dose 0.85.

Reference Jones D., Webb G.A.W. (15)
Figure 2.2
First and second readout for LiF-Teflon discs v heater current

LiF-Teflon glow curve in Conrad 5100B read out
(a) Unannealed (b) After 80°C preanneal
The effect of the low temperature annealing of LiF:Mg is associated with the conversion of Mg dipoles into trimers in the (111) planes as shown on the diagram above.

Photograph of disc annealing tube assembly and of holder used for ultrasonic cleaning of discs.
Fig. 5a. Idealised Graph Showing Optimum Heating Cycle for Annealing LiF/Teflon TLD Dosimeters During Readout

Fig. 5b. Recorder Trace of Heating Cycle for Modified Conrad 5100B TLD Reader

A = Heating cycle of unmodified instrument
B = Alternate heating cycles for modified instrument
C = Preheat
D = Read
E = Anneal
F = Cool (without high temp. anneal)
G = Cool (with high temp. anneal)

Temperature °C

Time

0 10s 20s 30s 40s 50s 1 min

10mV Calibration Pulse
Figure 2.6

Thermocouple Voltage

Heating cycle of modified Conrad TLD reader for varying heater current settings

Figure 2.7

Calibration curve for modified Conrad TLD reader

60 kVp X-rays

Dose
mrad

+ First calibration
• Second calibration

1000

10

10

DVM (mV)

100

1000

1000.
Figure 2.8. - See graph opposite.

Variation of light output with preheat temperature for modified TLD Reader.

Heating cycle modified to give preheat plateau temperatures of 110°C, 120°C, 130°C (normal), 140°C, and 150°C.

With corresponding readout temperature of 235°C in each instance the light outputs for five TLD discs, normalised to unity at 150°C, were as follows:

<table>
<thead>
<tr>
<th>Temperature</th>
<th>Disc 1</th>
<th>Disc 2</th>
<th>Disc 3</th>
<th>Disc 4</th>
<th>Disc 5</th>
<th>Mean</th>
</tr>
</thead>
<tbody>
<tr>
<td>110°C</td>
<td>1.22</td>
<td>1.26</td>
<td>1.17</td>
<td>1.19</td>
<td>1.21</td>
<td>1.21</td>
</tr>
<tr>
<td>120°C</td>
<td>1.12</td>
<td>1.05</td>
<td>1.09</td>
<td>1.08</td>
<td>1.07</td>
<td>1.07</td>
</tr>
<tr>
<td>130°C</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>140°C</td>
<td>0.90</td>
<td>0.84</td>
<td>0.88</td>
<td>0.97</td>
<td>0.95</td>
<td>0.91</td>
</tr>
<tr>
<td>150°C</td>
<td>0.73</td>
<td>0.84</td>
<td>0.86</td>
<td>0.80</td>
<td>0.77</td>
<td>0.80</td>
</tr>
</tbody>
</table>

The optimum preheat temperature comes at the point where the total readout changes least for small variations in the preheat temperature. It can be seen from the graph opposite that the adopted value of 130°C comes reasonably close to this optimum point.
Figure 2.9

Glow curve of LiF-teflon disc from reader with modified heating cycle.

Figure 2.10

Sensitivity of Estar base Kodak RM Film

![Graph showing film sensitivity vs. energy keV]

- Preheat
- Read
- Anneal
- Cool

![Graph showing film sensitivity vs. peak voltage-filtered X-ray tube]
The Aberdeen TLD personal dosimeter badge flanked by Harshaw and Conrad badges for their automatic readers.

Incorporation of TLD Discs into Standard Film Badge Dosimeters

Harshaw - Aberdeen - Conrad

Figure 2.12

Intercomparison of TLD and Film Badge Measurements

Areas of high uncertainty (doses below 10mR) shown shaded

- Film dose uncertain
- TL dose uncertain
CHAPTER 5: EXPERIMENTAL INVESTIGATIONS INTO THE PRACTICABILITY OF A TSEE DOSIMETRY SYSTEM

3.1 Introduction

Chapter 1 incorporated a discussion of the principles on which TSEE dosimetry is based together with a brief review of some of the early work carried out to assess its potential value as a practical dosimetric procedure. Most of that work involved measurements made on relatively thick samples using a Geiger counter to detect the emitted electrons, although, where the physical mechanisms associated with the exo-electron process were of principal interest, several workers stressed that advantages could be gained by reading out the dosimeters under vacuum using an electron multiplier as detector.

In this chapter an account is given of the development of a reliable TSEE reader based on gas flow proportional counting for the detection of the emitted electrons. This method was chosen because any experimental arrangement involving readout of a dosimeter under vacuum is inherently very difficult to adapt for use in a practical dosimetry system, whilst a gas flow counter operating in the Geiger mode tends to saturate at high counting rates and therefore unduly restricts the working range of such a system.

Only electrons emitted from traps located very close to the surface of the dosimeter have a finite possibility of escape and there is therefore no loss of efficiency in the use of very thin layer dosimeters for TSEE measurements. Holzapfel (1) has suggested that the exo-electron sensitive layer can be regarded as having an effective thickness approximating closely to the mean free path of the electrons in the crystals. This means that in principle TSEE dosimeters can be prepared with a sensitive layer only a few tenths of a nanometer thick. Such extra thin dosimeters would provide an immensely powerful new tool for the investigation of certain problems in the fields of interface dosimetry, charged particle or neutron dosimetry, and microdosimetry. An account of work carried out to develop
and evaluate the performance of such ultra-thin layer
dosimeters comprises an important part of the material
included in this chapter.

3.2 Apparatus for exo-electron studies

A TSEE reader incorporating a proportional counter
was developed by F.H. Attix during a sabatical year spent
at AEERE Harwell and subsequently described by him in 1971.(2)
By the courtesy of Mr. D.H. Peirson of the Health Physics
and Medical Division at AEERE I was given the opportunity
of using this reader as a visiting scientist following
Attix's return to the U.S.A. It incorporated a complete
set of the electronic modules used with the Harwell 2000
TLD reader. These had been modified for exo-electron
work by replacing the normal TLD head and current to pulse
rate converter with a specially designed hemispherical gas
flow proportional counter and amplifier-discriminator system,
(Figure 3.1a).

The counter head is shown on Figure 3.1b. It uses a
12 volt heater (designed for an automobile cigarette
lighter) fitted with a gold plated cup which accepts 1.5 cm
diameter graphite planchetts with a 1 cm diameter recess
capable of holding samples normally of about 10 mg but if
necessary up to 30 mg. Attix worked chiefly with Harshaw
LiF type TLD-100 whilst the work described in the following
sections was carried out with Conrad LiF-7.

Work with this reader system showed it had certain
dis-advantages. In particular problems arose during
sample loading. The sample holder was designed to be
plugged into the counter and secured through a bayonet type
fixing. This led to powder or loose crystal samples being
physically disturbed during loading and unloading which
causd considerable spread in successive readings of the
same sample. It was therefore decided to develop an alter-
native reader design in which a sample can remain undis-
turbed on a baseplate during a series of dose-readout
cycles. To enable an assessment of the importance of
counter geometry to be made it was arranged that the new
reader head would have a difference height to diameter ratio (1 : 3 instead of the 1 : 5 used for Attix's reader head). In addition the loop anode used by Attix was replaced by a needle point anode, and to permit intercomparison of TSEE observations with others in which the TL or TSC properties of the same samples were studied, alternative reader heads were designed to fit the same base-plate. The proportional counter head for TSEE studies could be quickly detached and replaced by either a PM tube head for observing TL signals, or a special electrode assembly designed for TSC measurements. These various reader head configurations are shown on figures 3.2 and 3.3. Whilst the signal from the TSEE counter head consists of a series of counting pulses which are fed into a conventional pulse amplifier, that from both the alternative heads has the form of a D.C. current of magnitude between $10^{-10}$ and $10^{-4}$ amp. To enable this to be monitored on the same ratemeter and scaler as the counts from the TSEE head a D.C. current to pulse rate converter covering this range was constructed as a NIM module. This used essentially the same circuit as the one developed for Perry and used with the Harwell TLD reader(3). Figure 3.4 is a calibration graph for this new module and it can be seen that the conversion is linear over the range of input currents specified above.

The other electronic units employed were standard commercial NIM modules, and included an E.H.T. unit, amplifier-discriminator, ratemeter and scaler. In addition for particular readings where this was required either a single channel or multichannel (Nuclear Data 1100 series) analyser could be plugged into the vacant positions in the NIM bin. The temperature control system was based on a direct reading Ether thermocouple controller with two set points which determined the limits between which the sample counts were integrated.

This new reader incorporated a two channel Rustrak recorder for monitoring glow or emission curves (TL or TSEE 'thermograms') and heating pan temperature, and also a
linear heating rate control unit. The latter differed from the thyristor switched design employed in the Harwell 2000 TLD reader by using a D.C. 12 volt 10 amp heater supply controlled through a series transistor. This was an OC 36 germanium power transistor on which the bias was varied by the output from a differential amplifier which compared the thermocouple output with the voltage from a ramp generator. This arrangement was adopted to overcome another of the problems that had been encountered with the Attix reader at Harwell where spurious bursts of TSEE counts had been noticed as a result of switching pulses in the thyristors of the A.C. temperature controller. Although the D.C. heater supply is more complex it was found that a very smooth temperature control could be achieved by this method.

The complete reader assembly is shown on figure 3.5. For a period readings were taken concurrently with this system and with the Harwell reader, a procedure which proved invaluable when the reason for unexpected observations was being sought.

3.3 Early Observations

The initial readings were all obtained using the Harwell reader with loose crystal samples in graphite planchets. Tests were made to determine whether there was any advantage in mixing the sample material with powdered graphite to minimise electrostatic effects - a practice first adopted by Kramer(4) - but it was found that with samples of only a few milligrams spread thinly on graphite planchets this precaution was unnecessary although saturation might set in at a somewhat lower dose level if the powdered graphite was omitted. The irradiations were carried out using a $^{90}$Sr - $^{90}$Y beta ray source and covered a dose range of 1 mrad to $3 \times 10^4$ rad. A few randomly chosen readings were repeated using X-ray irradiation and showed no deviations from the results with the beta source. This beta source fitted into a specially designed perspex jig which enabled various source sample separations between
1 cm and 10 cm to be selected - the jig was precalibrated using an extrapolation ion chamber so that the absorbed dose rate for each such separation was accurately known. The X-ray doses were determined using an MIL 37C air equivalent wall ionization chamber and electrometer; they were all measured at the time of irradiation.

It was found that in general the relationship between exo-electron counts and radiation dose was of the form:

\[ \text{E.E. counts} = A \times D^n \]

where A is a constant, D is the dose in rad and the index n depends upon the composition of the sample but is in general less than unity. This form of relationship had been previously reported by Kramer but was not observed in other investigations made by Becker who reported a linear relationship, (4) (5). The table below lists the values of A and n obtained with samples used for this report.

<table>
<thead>
<tr>
<th>Sample material</th>
<th>n</th>
<th>A</th>
</tr>
</thead>
<tbody>
<tr>
<td>TLD LiF-7 (Conrad) fused</td>
<td>0.94</td>
<td>4 \times 10^3</td>
</tr>
<tr>
<td>TLD LiF-7 (Conrad) vacuum deposited</td>
<td>0.98</td>
<td>1.2 \times 10^3</td>
</tr>
<tr>
<td>Reagent grade LiF (Extrapure)</td>
<td>0.66</td>
<td>1.5 \times 10^2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.5 \times 10^3</td>
</tr>
<tr>
<td>Extruded LiF (Harshaw TLD-700)</td>
<td>0.93</td>
<td>2.7 \times 10^5</td>
</tr>
<tr>
<td>BeO - B.D.H. (powder)</td>
<td>0.89</td>
<td>4.5 \times 10^5</td>
</tr>
<tr>
<td>BeO sintered ceramic disc</td>
<td>0.82</td>
<td>8 \times 10^3</td>
</tr>
</tbody>
</table>

It was also found from these early results that in the case of Conrad LiF-7 fading was not a serious problem for dosimeters stored in the dark even if readout was deferred for periods of up to eight weeks. However, the delayed readout did lead to a change in the structure of the emission curve. Electrons lost from shallow traps appeared to be predominantly retrapped in deeper ones so that counts lost from low temperature peaks were balanced by enhanced emission from those at higher readout temperatures.
(See figure 3.7). Similar changes in the TL glow curve of lithium fluoride have been reported (6). During this work the statistical variation between repeated readings tended to be several times greater than the expected counting variations (S.D. equal to $\sqrt{N}$ where $N$ is the total count) and results were therefore based on the mean of a number of readings. This larger variation could be explained by postulating that the number of actual emission sites is smaller than the number of electrons emitted. Such postulated emission centres would be analogous to the luminescent recombination centres responsible for thermoluminescence.

If this hypothesis is correct and if we further assume that the emission centre is activated at some point during the readout process (either thermally or by changes in the electric field configuration at the surface associated with the emptying and/or filling of traps) when a population of free electrons had already been established, there will be a possibility of almost simultaneous multiple emission of electrons immediately adjacent to the newly activated centres. This would probably be followed by emission of other single electrons which diffuse to the centre within the next few microseconds. The use of a high vacuum channel multiplier detection system with a very fast response time might then enable groups of closely bunched and decreasing amplitude events to be observed in the recorded time spectrum of the emitted pulses.

Although it was not practicable to carry out such observations some experimental support for this highly speculative model is given by the work reported in section 3.5(e). Very recently Gammage and Simpkin: (7) have also reported observations on ceramic BeO TSEE dosimeters which give experimental support to the concept of such centres being associated with strong local pyroelectric fields developed along the c axes of the BeO crystals as a result of thermal stressing.

More serious discrepancies occurred with the readings from about 10% of the samples for which the observed count appeared to bear no relationship to the expected value.
Such spurious readings were usually excessively large but occasional samples were found which gave virtually no counts. Repeating sets of readings on a group of about 10 samples several times in succession it was typically found that one of these would give a quite abnormal count on each occasion, but that the sample concerned was not the same for different runs.

Careful observations showed that unexpectedly high counts were typically associated with readout cycles in which very short bursts of very intense counting occurred at rates equal to or approaching the maximum that the resolution time of the counter assembly and amplifier permitted. This pointed strongly to counter instability as the cause. Equally it was thought that very low readings were due to electrostatic effects associated with inadequate earthing of the samples. It was however difficult to exclude the possibility of variations in sample composition being responsible in all cases. Thus very high count rates could be due to either an unusually large number of emission centres, or a group of centres having an abnormally high efficiency, being exposed by the re-arrangement of sample grains which inevitably occurs when unloading the reader and re-irradiating the sample. Conversely it had already been found that very low counts could follow the exposure of the prepared samples to certain organic vapours such as those from plasticising agents. The reduced emission appeared to always be annealed out by heating so that normal sensitivity was restored by taking the sample concerned through a readout cycle and this made it difficult to be certain that apparently spurious low counts were not due to temporary poisoning of the emission centres.

Similar poor reproducibility was encountered by early workers on the photo-electric effect and was not overcome until both sample preparation and examination were carried out under vacuum to prevent surface contamination. Following the initial TSE3 measurements it was felt that similar precautions might be necessary for consistent
observations of exo-electron emission, which would make the use of this effect for practical dosimetry much more difficult to develop successfully. Although the later work described in the rest of this chapter has gone a long way towards establishing conditions under which consistent results can be obtained with a practical dosimetry system based on gas flow counting it is clear that further research using an all vacuum channel multiplier detection system (with the exo-electron emitting surface freshly prepared under vacuum) would be of great value both in assessing the importance of surface contamination effects and in improving the time resolution of the detection system.

To help differentiate between sample and counter effects the second reader described in section 3.2 was developed. This was designed to enable samples to be re-irradiated and read out repeatedly without being disturbed in any way. Counter instability was reduced by the use of a rigid needle point anode and the design gave improved earthing of the sample planchets. With these changes the spurious results were much rarer and variations between successive readings made on the same sample when left undisturbed in the sample holder were considerably reduced. Handling of prepared samples could be shown to lead to small changes in their response. A two point programme was therefore planned for the continuation of the investigation, the aims being:

(a) To develop samples which were geometrically stable and would give consistently repeatable readings.

(b) To eradicate spurious counts arising from reader instability.

These developments are described in the following sections.

3.4 Sample preparation

A critical requirement for the production of satisfactory TSEE dosimeters with a reasonably linear response over a wide range of doses is that the exo-electron sensitive material must be deposited onto its planchet or
other substrate in good thermal and electrical contact with the heating element(8). The majority of Attix's work at Harwell had been carried out with samples which were directly weighed out into graphite planchets as 80 : 20 mixtures of loose crystals and powdered graphite. Others were deposited from a suspension of finer powders in acetone as described by Becker (9) (10). Attix also experimented with the use of sodium silicate as a binding agent as previously reported by Svarcer and Fowler (11) and Kramer (4). Unfortunately this led to a very considerable loss of sensitivity with exo-electron samples even when the quantity of silicate used was the minimum which would give reasonable binding.

In the present investigation many months were spent studying alternative techniques for the production of samples in a stable form that could be relied upon to give closely corresponding counts following a series of identical doses. Samples which could be produced in the form of solid discs (e.g. ceramic BeO discs or single crystals of LiF) gave no greater consistency than loose powder samples, probably because of electrostatic effects arising from difficulty in effectively earthing the emitting surface. For this reason the use of compression to facilitate binding of powdered samples as described by Shambon and Condon (12) was not attempted. Deposition from solution was not practical for LiF as no suitable solvent could be found. (It was however possible to prepare acceptable \( \text{Li}_2\text{B}_4\text{O}_7 \) dosimeters from an aqueous solution). Several alternative binding agents were investigated and trace quantities of aluminium stearate proved the most satisfactory of these. It softens during the heating cycle thus minimising the problem of stresses which could affect the readout, (mechanical stress is an alternative technique for stimulating the release of exo-electrons). In this way some samples were prepared with only about a 10% sensitivity loss compared with loose powders, however reservations were felt about the long term stability of these as over a period of time this binding medium might 'flow'
over the surface of the crystals so reducing the effective emitting area and furthermore there were signs that after repeated heat cycling aluminium stearate became more brittle and could develop cracks.

Next, very thin layers of various cements were applied to the bottom of stainless steel sample planchets and used to secure relatively large crystals* of luminescent grade lithium fluoride. It was found that some of these cements could be used without serious loss of sensitivity. The first cement used successfully in this way was MS 840 silicone thermosetting resin. This however is only rated for heating to 250°C and the manufacturers (Midland Silicones Ltd.) advise against even short term heating to beyond 300°C. Even then there is a possibility of thermally induced stresses being set up in the cement layer following repeated heat cycling. A more satisfactory product in this respect is the polyimide thermocuring plastic varnish RK 692 Pyre ML (Du Pont de Nemours, Swedish Division). This remains stable up to at least 600°C. Samples produced with these two cements showed essentially similar response. The most satisfactory were produced with the thinnest layers of cement and had sensitivities about 10% less than those of equivalent loose crystal samples. The silicone resin was more easily used as it could be readily diluted and spread with a small brush. Nevertheless the Pyre ML varnish was not difficult to use; and although the samples produced tended to be a little less sensitive they could be heated freely to read out the 320°C peak in lithium fluoride and could be expected to retain better long term stability with repeated heat cycling. However, following these and other studies it was eventually concluded that the most satisfactory way of producing stable samples from Conrad LiF-7 was by thermal fusion. Crystals of this material soften at about 800°C and adhere together to form a continuous layer. If they are heated in a stainless steel

* Those retained between sieves with mesh sizes of 80 and 120 (i.e. between 200 μm and 75 μm)
planchet good adhesion is readily achieved but the stainless steel, which is not tissue equivalent, forms a less satisfactory substrate for dosimetric purposes than the usual graphite planchet. After a number of trials two satisfactory fused samples were also prepared on these graphite planchets by the simple although irreproducible technique of heating the loaded planchet in a bunsen flame until it reached a bright cherry red colour. Using this method, if the planchet is overheated the LiF beads melt and run together to form one large unstable drop which does not readily adhere to the graphite surface; also if there is a trace of moisture in the graphite the planchet disintegrates during heating. A controlled method of producing such dosimeters was developed later using the vacuum evaporation plant which was constructed for the preparation of vacuum deposited thin layer dosimeters. This involved placing previously carefully dried graphite planchets on a tungsten strip connected between the evaporation electrodes, evacuating the chamber and then gradually increasing the current through this heating strip until the melting point of lithium fluoride was just reached.

Fused samples on both stainless steel and graphite planchets proved extremely stable, and when problems of instability in the counter had been overcome they gave repeatable results with a variation not appreciably greater than that expected purely from counting statistics. This development opened the way for the detailed study of factors affecting the counter stability and sensitivity reported in the next section.

A considerable amount of work was carried out with beryllium oxide samples where the high toxicity of the powdered material discourages laboratory production of dosimeters by any of the above methods. Tests of this material were carried out with the apparatus under a fume hood and using samples dispensed and used under the same hood to prevent beryllium contamination of the laboratory. They showed that it is probably the most sensitive near tissue equivalent sample material for use in exo-electron
dosimetry: The toxicity problem is most readily overcome by using samples prepared in the form of sintered ceramic BeO discs. Such discs* were concurrently being evaluated for thermoluminescent dosimetry by Scarpa (13) and a small supply was obtained for evaluation in the TSEE reader. Initial results were very erratic and this was attributed to electrostatic fields at the surface of the discs affecting electron emission. To combat this a fine earthed metal gauze was pressed firmly against the emitting surface during readout and was found to greatly reduce the variations in output counts. About the same time Becker's group at Oak Ridge reported evaporating an ultra-thin metal film over BeO discs to be used as TSEE dosimeters for the same purpose (14).

One further technique of sample preparation was studied rather later - deposition on a substrate under vacuum. Access to a vacuum evaporation plant in 1971 had enabled the practicability of this method to be demonstrated but a change of post by the author prevented the work being followed up until late 1973 when the necessary parts had been collected together for the construction of a suitable evaporation plant in a new laboratory. The vacuum deposition of luminescent films has been studied by Feldman and O'Hara (15) who reported that to retain good luminous efficiency the evaporated film should be baked after deposition. The new evaporation plant was therefore designed so that samples could be evaporated onto stainless steel planchets and subsequently a thin molybdenum strip adjacent to the planchet could be raised to white heat to bake the deposited film before it was exposed to the atmosphere. Tests were carried out on lithium fluoride (both Luminescent and General Purpose Reagent grades) lithium tetraborate and magnesium fluoride. In all cases films were prepared in three different ways, without baking, baked in air and baked whilst still under vacuum. Results showed that heating either in air or under vacuum to

* Obtained from Consolidated Beryllium Ltd., Milford Haven
temperatures appreciably above the normal readout and annealing temperature merely reduced the exo-electron emission sensitivity. However the samples prepared without baking all showed sensitivities of the same order of magnitude as those of loose powder samples of the same materials.

After the production of dosimeters evaporated onto stainless steel planchets had been shown to be practical attention was turned to the problem of finding a more tissue equivalent substrate material which would not adversely affect the energy response curve for the exo-electron sensitive layer. Most plastics were excluded by the requirement for thermal stability during the readout cycle but teflon sheet proved satisfactory for readout temperatures of below 300°C. Later Kapton polyimide film* from Du Pont was found to be the ideal answer to this problem. With dosimeters evaporated onto a plastic substrate it is essential that a thin layer of aluminium is evaporated over the substrate before the sensitive material in order to avoid electrostatic fields developing at the surface and leading to erratic exo-electron emission. The use of Balzer's substrate cleaner number 2 was found to help considerably in achieving good adhesion of the deposited layers.

Samples of a non-luminescent grade of pure LiF were found to have almost as high a TSEE sensitivity as 'dosimetric grade' material such as Conrad LiF-7, but the exo-electron emission curve showed only one broad peak (Figure 3.6a) which implies that it can only have a relatively simple trap structure. Figure 3.8: shows that the exo-electron emission curve of the luminescent grade lithium fluoride after evaporation has been greatly modified and becomes not dissimilar to that of the non-luminescent grade material, although the overall TSEE sensitivity of the material has not been greatly altered. Taken in conjunction with the failure of attempts by others

* A polyimide of pyromellitic acid dianhydride and 4, 4' diaminodiphenyl ether
to use evaporation of luminescent grade lithium fluoride under vacuum for the production of thermoluminescent dosimeters (16) this suggests that impurity atoms (such as the titanium luminescence centres) either are not carried over or are inactivated during evaporation; and this also probably explains why vacuum deposition is not a practical method for the production of thin thermoluminescent dosimeters.

Evaporated films of different thickness were produced by using different weights of lithium fluoride in the evaporation boat and readings of the exo-electron sensitivity of these showed that even very thin layers are effectively infinitely thick exo-electron emitters. It is hoped that eventually these observations can be put on a quantitative basis by using a series of carefully prepared films of known thickness to determine whether the exo-electron emitting layer does approximately correspond with the mean free path of the electrons (≈ 30 - 100 Å) in the sample material as predicted by Holzapfel (1). Eschbach has reviewed the problems involved in preparing such films (17).

3.5 Counter Instability

Apart from occasional spurious results which suggested counter instability, the original Harwell form of the reader also showed a steady loss of sensitivity with use, necessitating an increase in the operating potential of about 50 volts for each month of intermittent use. In less than a year this carried the required voltage beyond the range of the available E.H.T. supply so that exo-electrons could no longer be detected. (Consistent sensitivity was maintained by adjusting the reader gain to give a standard count rate for a radioactive source containing a low energy beta emitter, which served the same function for the TSEE reader as the standard light source does for a TLD reader). At first this loss of gain was thought to have taken place in the type 0058 pre-amplifier used for the reader but a more detailed investigation showed that it arose from a reduction in the gas gain of
the counter head. This was eventually traced to spark polishing of the anode wire showing that the original sensitivity (which was restored by replacing the anode wire) must have been due to Townsend avalanches occurring at minor irregularities and imperfections on the surface of the wire. Following this it was decided to carry out a full investigation into factors which could affect the sensitivity or stability of the counter. The following five factors were studied:

(a) Counting chamber shape and nature of anode.
(b) Gas flow dynamics.
(c) The effect of the heating cycle on the gas gain.
(d) The choice of counting gas.
(e) The relationship between the pulse height spectrum from the counter and the energy spectrum of the emitted exo-electrons.

(a) Counting chamber shape and nature of anode

Campion et al have studied the conditions under which proportional counters will detect single electrons and also the various mechanisms by which spurious pulses may be produced in methane or argon-methane flow counters (18), (19), (20), (21), (22), (23). His investigations demonstrated that single electrons were only detected when the E.H.T. voltage was advanced beyond the point corresponding to the end of the plateau for a radioactive source. Under such conditions spurious afterpulses may be induced by the action of both positive ions and photons (from excited gas atoms). Campion concluded that positive ion afterpulses were less serious with methane than with an argon-methane mixture. This probably explains why the Harwell TSEE reader was particularly prone to instability when operated with argon-methane.

Initially therefore, observations were restricted to methane and involved comparisons of the stability achieved with various types of anode when used with counters having chambers of different height to diameter ratio. The importance of this ratio has been studied by Roost et al (24)
who concluded that to minimise photon induced afterpulses it should be large. The alternative reader head described in section 3.2 was therefore designed with a counting chamber of the same height but only half the diameter of the one designed by Attix. It was fitted with a needle point anode formed from an ordinary hand sewing needle (Millwards number 3 Sharp). Used with methane this reader gave no indication of instability or erratic counting and proved to have a sensitivity closely comparable with that achieved by Attix. Even when used later with argon-methane (90% A, 10% CH₄) it still proved perfectly stable and reliable.

This needle anode assembly was therefore transferred to the Harwell reader where it gave very similar results, indicating clearly that for exo-electron measurements the nature of the anode was more important than the counter geometry. Work was continued into testing a series of different anodes. The first was a stretched straight wire. After this a piece of razor blade was used and then one of the special wire anodes developed by Dr. J.A. Cairns at AERE for low energy X-ray spectrometry and shown on figure 3.9, (25), (26). Finally an Aloester surgical needle anode was fitted and tested. Plateau curves using the radioactive source were plotted with each of these anodes are shown on figures 3.10 to 3.20 inclusive. A comparison of all these curves suggested that the value of the radius of curvature of the anode is critical for efficient counter operation. Photomicrographs of the two needle point anodes were therefore taken and are shown on figure 3.21. The radius of curvature of the Millward needle was measured as 0.022 mm but the surgical needle, has a much sharper point. The radius of the wire anodes are about half that of the Millward needle whilst that of the edge of the razor blade is about one tenth as great. The data suggests that, as reported by Campion and his co-workers, single electrons are only detected efficiently for operating points beyond the end of the normal plateau. This effect is least marked with the Cairns anode (which showed the longest plateau) and most marked with a surgical
needle (which shows the shortest plateau). The razor blade anode with a nominal radius of curvature along its edge of 0.003 mm rather surprisingly required a higher E.H.T. than the 0.025 mm diameter wire anodes which probably indicated the influence of surface irregularities in controlling the exact operating voltage required with wire anodes. Due to limitations in the available E.H.T. supply (maximum output 5 kV) this razor blade anode could only be operated with the argon-methane mixture, and with this it showed serious instability. The same difficulty occurred with the argon-methane mixture when using all the anodes other than the needle points. This suggests that the problem arises from positive ion induced afterpulses possibly associated with corona discharges at different points along the perimeter of the extended anodes.

Instability was never detected with the needle anodes and this was put down to the presence of space charge which completely fills the sensitive region and serves to quench any afterpulses. However with the extended sensitive length of the wire anodes instability was present to a limited extent even when the counting gas was pure methane. Therefore such afterpulsing appeared to be the principal cause of the erratic results encountered with much of the early work carried out on the Harwell reader. Although with the Millward needle the plateau plotted for the radioactive source is rather short and the voltage required for good exo-electron sensitivity comes well beyond the end of this plateau, the overall performance of the counter is undoubtedly better. It shows completely stable behaviour for either chamber geometry, either counting gas and a wide range of discriminator settings. From the characteristic curves however, it does appear that a needle point with a slightly blunter end having a radius of curvature at the top of about 0.05 mm would probably give a longer plateau and optimum exo-electron detection without raising the counter working voltage unduly.
(b) Gas flow dynamics

Both the volume and direction of the gas flow through the counter were varied, the former being monitored by a flowmeter. The expected statistical variations in either the counts per minute from the radioactive calibration source, or the total exo-electron count obtained from a fused LiF-7 dosimeter exposed to a dose of 1 rad, were observed if a series of identical readings was repeated with any value of gas flow rate between 0.5 l/min and 5.0 l/min, (see figure 3.22). But if the gas flow rate was reduced to below 0.5 l/min variations exceeding what would be statistically expected were occasionally encountered. These were attributed to residual traces of air in the counter, and consequently the operating conditions were subsequently set so that there was a flow rate of about 1 l/min at all times. Tests were also carried out to check whether it was necessary to leave the counter for any specific minimum period after loading in order to ensure that any air introduced during loading had been flushed out. It was found that with the range of flow rates quoted above no signs of erratic operation were encountered even when readout was immediate.

Further tests were then carried out with the direction of the gas flow through the counter reversed. Figure 3.23 shows comparative readings for the two cases. The direction of flow does not appear to have any significant effect on the operation of the counter. In practice all other work was carried out with the gas flowing through the counter in a downward direction (the reverse of that used by Attix) largely because it was felt that this would minimise the possibility of air being forced into the counter during the loading of a dosimeter. The importance of this is shown by Figure 3.24 which compares the behaviour of the counter when filled with argon, argon-methane and air.

(c) The effect of the heating cycle on the gas gain

The sample heating process leads to a reduction of gas
density which can be expected to produce an increase in the
electron mean free path and hence a reduction in gas gain.
To check whether the change is significant the count rate
from the radioactive calibration source was recorded by the
ratemeter both during isothermal operation and during a
normal heating cycle. The result, shown on figure 3.25,
demonstrates that the effect of these changes is equivalent
to a decrease of about 8% in detection efficiency as the
temperature is increased from ambient to about 400°C. To
ensure that the effect when counting single electrons is
not appreciably different from that observed with the
radioactive source, this was replaced by a heated wire which
served as a source of single thermionic electrons. It was
found difficult to keep the temperature of this wire
sufficiently constant to maintain uniform electron emission
throughout a heating cycle but the result shown on Figure
3.25 correlates reasonably well with that obtained using the
radioactive source. The effect of the dosimeter heating
cycle on a recorded emission curve is therefore to produce
a small consistent reduction in the height of high tempera-
ture emission peaks relative to low temperature ones.
To show that this effect is small a series of readings
were taken with the reader head inverted so that the heater
was at the top of the chamber and the anode at the bottom.
Under these conditions convection currents in the chamber
should be minimised and the temperature of the gas near the
anode should therefore remain more nearly constant.
Figure 3.26 shows that the change only affects the sensitivity
of the counter appreciably when operating at very low E.H.T.
voltages where the overall counting efficiency is low.

(d) The choice of counting gas
The principal gases known to be satisfactory for pro-
portional counting are pure methane, argon-methane and
helium-isobutane. Under appropriate conditions all of
these are capable of giving consistent results. However the
helium-isobutane mixture showed no advantages over the other
two gases, and as it was both more expensive and less readily available its use was discontinued at an early stage. Of the other two gases argon-methane has the advantage of operating at a much lower working voltage which is well below the threshold voltage for counting in air and it therefore shows a high tolerance to residual traces of air in the chamber during readout. Nevertheless methane is less liable to produce spurious pulses and with other than point anodes its use appears obligatory for this reason.

With either counting gas, traces of oxygen resulting from residual air in the counting chamber can lead to quenching due to the large cross-section of electronegative gas molecules for the absorption of low velocity electrons. This effect should be less marked with argon-methane where the electron mobilities are much higher.

The measurements discussed in the next section of this report were based on the use of an argon-methane gas mixture with the anode formed from the Millward no. 3 Sharp needle. This combination is regarded as giving the best overall reader performance for sensitivity, stability and reproducibility.

(e) Counter pulse height spectrum

An experimental investigation into the statistical variation of output pulse amplitudes from a proportional counter has been carried out by Curran's group at Glasgow (27), and their results have been confirmed theoretically by Byrne (28). They are not however easily applied to the case of counters with non-cylindrical geometry or to pulses initiated by single exo-electrons emitted with an energy far less than the ionisation energy of the counter gas (14.5 eV for methane and 15.7 eV for argon). Experimental studies were therefore made with the TSEE reader to investigate the variation in the amplitudes of pulses initiated by single thermal electrons (emitted from a hot wire), exo-electrons and $^{14}$C beta particles from a radioactive source. In each case the variation of count rate with amplifier gain was determined and the results are
all shown on figure 3.27. It can be seen that with the greater input signal from the ionizing beta particles there are relatively few small pulses which fall below the counter threshold as the gain is reduced, but that with both the low energy electron sources there is a linear relationship between the logarithm of the count rate and the amplifier gain (in decibels) which extends over a wide range of gain settings. It is also clear that the exo-electron initiated pulses have an output pulse distribution with a longer tail at large amplitudes than the distribution due to the thermal electrons.

To determine the reason for this the reader was modified by the introduction of a negatively biased grid into the counter to estimate the energies of the emitted exo-electrons. The results showed that for Conrad LiF-7 the number emitted falls off exponentially with increasing energy and that virtually none of the exo-electrons were emitted with energies in excess of one volt (Figure 3.28). This proves that the variations in the pulse amplitudes of exo-electron induced pulses cannot be accounted for by anomalously high energy electrons leading to multiple initial ionization processes. It also verifies that all the counts recorded by the reader are due to the emission of true exo-electrons (or at least to electrons originating from the sample) and not to photo-electrons which could be emitted from the counter walls as a result of illumination by thermoluminescent light from the sample.

A possible alternative explanation of the results shown on figure 3.27 would be multiple emission of exo-electrons. For the thermal electrons it can be seen that halving the gain halves the recorded count rate whereas for the exo-electrons the reduction in counts is only about one third as great. It therefore appears that up to two thirds of the exo-electron counts are initiated by the simultaneous emission of more than one electron, thus producing larger amplitude pulses than the ones initiated by individual exo or thermal electrons.

The relative probability of multiple emission at
different temperatures can also be assessed from the readings. Figure 3.29 shows the variation of ¹⁴C and TSEE counts with discriminator bias and should be compared with figure 3.30 showing the recorded emission curves at each setting. Similarly figures 3.31 and 3.32 show the effect of changes of amplifier gain and of E.H.T. voltage on the recorded emission curves. All these three families of curves indicate that the high temperature peak contains the largest proportion of small pulses and is therefore the most strongly affected by either an increase of bias or a reduction of system gain.

These results of course can be partially explained by the reduction of gas gain with increasing counter temperature discussed in 3.5 (c) above. However, the effect appears more marked than would be expected on this basis alone and furthermore when working with Harshaw LiF TLD 700 Attix reported finding the greater proportion of low amplitude pulses associated with the low temperature peak which, for this material, is the one that shows the more intense exo-electron emission. He explained this by suggesting that the high temperature peaks have a larger proportion of high amplitude pulses because multiple exo-electron emission is more likely at higher temperatures.

The results of the present work confirm that a mechanism of multiple emission seems to be necessary to account for the pulse height distribution of recorded exo-electrons and also indicate that there is a negative correlation between the mean pulse height and the instantaneous intensity of exo-electron emission. The use of a sample material with a different exo-electron emission curve has however shown that Attix's hypothesis of multiple electron emission being a direct function of temperature is not correct, and further work is now clearly desirable. As discussed in section 3.3 this could be carried out using a reader based on a channel multiplier with a much faster response time which would enable any fine structure associated with the pulses to be resolved. In this way it might be possible to add appreciably to detailed
understanding of the actual emission process.

3.6 Conclusions

From the work reported in this chapter the conclusions which can be drawn about equipment suitable for use in a practical TSEE dosimetry system can be summarised as follows:

(a) When used with sufficiently stable samples TSEE dosimetry carried out with proportional counters is capable of giving reliable and consistent results.

(b) A loop or straight wire anode is not very suitable for use in exo-electron readers. A needle point is much more satisfactory and gives completely stable operation over a wide range of discriminator settings using either methane or argon-methane as the counting gas. An investigation into the optimum needle point radius shows that an unexpectedly large value of $\sim 0.05$ mm, coupled with a higher than usual E.H.T. voltage, appears to give the least critical operating conditions.

(c) The counter must be operated at a point well beyond the end of the normal counting plateau as plotted with a soft beta emitting radioactive source. Multiple counting of exo-electrons due to after pulsing does not appear to be a problem under the foregoing conditions.

(d) A radioactive source is very useful for the initial setting up of the reader and reproducible sensitivities are readily obtained for exo-electron detection under different ambient conditions by adjusting the E.H.T. to give a standard count rate with the radioactive source. The count rate against voltage curve for a $^{14}$C source is much steeper at the operating point than the corresponding curve for exo-electrons counted from dosimeters exposed to a standard calibration dose. Variations in this source count rate therefore provide an extremely sensitive indicator for any changes in the exo-electron detection sensitivity.

(e) A heating cycle which can be accurately reproduced is essential. It is desirable for comparative studies that
this should incorporate a variable linear heating rate controller. Alternating current controllers working by means of thyristors are not satisfactory for exo-electron readers as on-off control appears to be associated with erratic counter operation. A modified system using a D.C. heater supply drawn through a power transistor controlled by a feedback loop proved much more satisfactory.

(f) The mechanical agitation associated with loading powder or crystalline materials into dosimeter planchets leads to a small additional count over that produced by the radiation exposure. This fades rapidly and is avoided if a short interval, of the order of a minute, is allowed between loading and readout. (This waiting period also serves to minimise the possibility of the readout being affected by traces of residual air in the counter). This small spurious count is probably related to the triboluminescent signal in TLD measurements. It depends upon the type of sample but is typically equivalent to exposures of between 5 and 50 mR. It is particularly marked in the case of beryllium oxide powder dosimeters.

(g) Bright illumination leads to fading of the stored signal and the reader should not be loaded in bright sunlight. Exposed dosimeters should also be protected from daylight. Contrary to reports made by some other workers, no fading of the stored signal was observed to result from exposure of the dosimeters to fluorescent lights.

(h) To retain constant sample sensitivity requires very consistent annealing procedures and these are most easily realised by allowing the samples to cool to below 100°C before the reader is unloaded - a predetermined gas flow rate being maintained throughout to ensure consistent sample cooling rates.

(i) Where dosimeters have to be subjected to external annealing it is difficult to avoid arbitrary sensitivity changes unless the annealing ovens used have an unusually sophisticated temperature control system. In practice external annealing was, wherever possible, only used for large batches of powder or crystalline sample material
and was then followed by a redetermination of the sensitivity of the batch.

(j) In practical dosimetry when there is a risk of the readout signal from the low temperature peak having faded there should be some advantage to be gained from the use of a non-linear three part heating cycle analogous to that developed for thermoluminescent dosimetry and described in chapter 2.

(k) It was not the intention of this work to make definite recommendations about the most satisfactory sample material but certain restricted conclusions on this matter can also be drawn: unless measurements of integrated counter current are made (this method of working has been studied at some centres and has been shown to be quite practicable for high dose measurements) TSEE dosimetry is analogous to TLD dosimetry carried out by photon counting and has the same limitations at high doses. Saturation of the response appears to set in at much lower dose levels than would be expected from analogous conventional TLD observations. Such saturation is seldom a fundamental property of the sample material. More usually it is an artefact resulting from the effect of electrostatic charges in the sample, counter spacecharge, the deadtime of the counter, the limited count rate response of the reader etc, and the dose level at which it is observed can be varied within certain limits by altering the heating rate during readout. In practice this means that it is not easy to design a TSEE dosimeter which will operate successfully over a dose range of more than about four decades, and that the choice of optimum sample material will depend greatly upon the dose range of interest.

Of all the materials tested BeO was the most sensitive and in the form of sintered ceramic discs it constituted a very stable and reproducible form of sample. Without special preparation it was capable of measurements down to a few mrad and work elsewhere has shown that by suitable surface treatment the lower limit of sensitivity can be improved to a level of a few microrad.
(1) Apart from sintering into ceramic discs or pellets the most practical methods for the production of TSEE dosimeters appear to be thermal fusion of crystalline samples onto an electrically conducting substrate and vacuum deposition of thin layer dosimeters. Crystalline materials can also be compounded into solid dosimeter discs by the use of thermocuring resin cements although this technique appears to offer no fundamental advantages other than convenience in use. The vacuum deposited thin layer samples appear to be intrinsically the most suitable for TSEE work as they take full advantage of the surface sensitivity of TSEE materials in cases where it is necessary to make measurements for which volume sensitive detectors are unsuitable.

(m) Numerous factors can affect the sensitivity of an individual dosimeter, for example chemical impurities, state of oxygenation or hydration, completeness of contact with substrate and stresses in the exo-electron sensitive layer, electrostatic charge dispersal etc. This makes it very difficult to produce a large number of samples that closely correspond in sensitivity.

(n) It is unlikely that very thin layer surface sensitive dosimeters exposed both to mechanical damage and chemical contamination in the course of routine use will ever achieve a degree of stability comparable with that of volume sensitive solid state dosimeters. Predictions that the use of TSEE dosimetry will in time replace TLD dosimetry therefore appear unrealistic. Nevertheless when used under controlled conditions and within the limits set by the various points discussed above it has been demonstrated that TSEE dosimetry is a perfectly viable technique which offers a number of specific advantages if compared with thermoluminescence. This is confirmed by work reported in chapter 6 of this thesis which describes the use of TSEE measurements for the solution of a group of specific practical problems encountered in routine dosimetry.
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FIGURE 3.1
THE HARWELL EXO-ELECTRON DOSIMETER READER

N.B. Numbers are references to Harwell 2000 Series Modules.

Fig. 1
PROM:
J.A. Dennis. 'Progress Report for the Neutron Dosimetry Section.' Reference Report AERE/HP/GEN/71
Schematic Diagram of EOE—Electron Reader Head Assembly

Spring loaded cantilever arm for holding counter head against sample planchet

Gas in

Needle Anode

Gas out

Spring loaded skirt giving gas seal against 'O' ring on reader baseplate assembly

Counting Chamber

Water Cooling Coil

Dosimeter Element for Readout Heater cup assembly

'O' ring seal

Fixed baseplate and heater assembly

Thermocouple

Heater Connections

Block Diagram of Complete TSEE Reader

0-5 kV

PRE-AMP

AMP

DISC

SCALER

RATEMETER

RECORER

ETHER TWO POINT TEMP CONTROLLER

12 V

LINEAR HEAT CONTROLLER

Scaler inhibit except between preset temperatures

Lower Upper set temperature

Heater cut-off at upper set point
Reader heads developed for the reader used in this research
FIGURE 3.4
Calibration graph—Current to Pulse Rate converter

FIGURE 3.5
The complete TSEE reader.
FIGURE 3.6a
TSEE 'Thermograms'
for various materials
Response for one rad doses

TSEE Count Rate

50°C

FIGURE 3.6b
TSEE Dose response curves for various materials
FIGURE 3.7
Fading in lithium fluoride—Conrad TLD-7.
All curves approx mean of ten samples exposed to one rad.

TSEE
COUNTS
--- Prompt readout
- - - Readout after one week
- - - Readout after three weeks
- - - Readout after five weeks

FIGURE 3.8
Exo—electron Emission Curves for Powder and Vacuum Deposited LiF
TSEE Dosimeters
FIGURE 3.9
SPECIAL FORM OF X-RAY PROPORTIONAL COUNTER ANODE
Developed by J.A. Cairns

FIGURE 3.10
LOOP ANODE METHANE GAS DISCRIMINATOR 1V
FIGURE 3.11
NEEDLE POINT ANODE
MILLWARDS No 3 SHARP DISCRIMINATOR 0.3V
METHANE

FIGURE 3.12
NEEDLE POINT ANODE
MILLWARDS No 3 SHARP DISCRIMINATOR 1.0V
METHANE
FIGURE 3.21
PHOTOMICROGRAPHS OF NEEDLE ANODES
Millwards No 3 Sharp Hand sewing needle
Aldester Surgical Needle 4 cm x 1 mm

FIGURE 3.22
VARIATION OF COUNT RATE WITH GAS FLOW RATE THROUGH COUNTER E.H.T. 2.2kV DISCRIMINATOR 4V ARGON-METHANE
**FIGURE 3.23**

EFFECT OF REVERSING GAS FLOW IN COUNTER

NEEDLE POINT ANODE

DISCRIMINATOR IV

ARGON-METHANE

**FIGURE 3.24**

COUNT RATE CURVES FOR 14C RADIOACTIVE SOURCE

NEEDLE POINT ANODE

DISCRIMINATOR IV

VARIOUS GASES
FIGURE 3.25
Variation of gas gain of counter during heating cycle

FIGURE 3.26
EFFECT OF OPERATING COUNTER INVERTED NEEDLE POINT ANODE DISCRIMINATOR IV ARGON-METHANE
FIGURE 3.27

VARIATION OF ELECTRONIC GAIN
NEEDLE POINT ANODE. E.H.T. 2.1 KV
DISCRIMINATOR 0.5V. ARGON-METHANE

FIGURE 3.28

USE OF BIASED GRID IN TSEE COUNTER
NEEDLE POINT ANODE. E.H.T. 2.1 KV
DISCRIMINATOR 1V. ARGON-METHANE
FIGURE 3.29
DISCRIMINATOR BIAS CURVES
NEEDLE POINT ANODE
ARGON-METHANE

FIGURE 3.30
Variation of TSEE 'thermogram' for fused TLD7 dosimeter with discriminator setting.
E.H.T. 2.1kV Argon-methane
Amplifier gain 73.5 db.

- Discriminator 0.5V
- Discriminator 1.5V
- Discriminator 2.5V
- Discriminator 3.5V
- Discriminator 4.5V
Variation of TSEE 'thermogram' for fused TLD7 dosimeter with amplifier gain

Figure 3.31
E.H.T. 2.1kV Argon-methane Discriminator bias 0.5 volt

- Gain 73.5 db
- Gain 67.5 db
- Gain 61.5 db
- Gain 55.5 db
- Gain 49.5 db

EE Count Rate

20°C Temperature 400°C

Figure 3.32
Variation of TSEE 'thermogram' for fused TLD7 dosimeter with counter E.H.T. Amplifier gain 73.5 db Argon methane Discriminator bias 1.0 volt

- E.H.T. 2.3kV
- E.H.T. 2.2kV
- E.H.T. 2.1kV
- E.H.T. 2.0kV
- E.H.T. 1.9kV

EE Count Rate

20°C Temperature 400°C
CHAPTER 4 : STUDIES OF THERMALLY STIMULATED CONDUCTIVITY

4.1 The TSC Reader

Section 1.5 of this thesis reviews the basic mechanism of the TSC process and discusses the advantages and disadvantages inherent in the use of this process for dosimetric measurements. It also briefly reports work carried out by two groups active in this field subsequent to the authors measurements which were made at Aberdeen in 1972-73 and which the following sections describe.

The reader used for the Aberdeen work was a simple modification of the general research reader designed for intercomparisons of TSEE, TLD and TSC response which was described in Section 3.2. For TSC measurements the reader utilised the basic stand with heating element and gold plated sample cup designed for the TSEE observations already reported in chapter 3, but the spring loaded reader head was fitted with the simple electrode shown in figure 4.1b in place of the proportional counter or photomultiplier assembly employed for the TSEE or TLD observations.

Polarising voltages of up to 5 kV from a 1359A E.H.T. unit were applied through the top PET 200 socket which was fitted on an insulating plate. The TSC current was measured on the earth return line from the lower electrode using the PET 100 socket fitted to the side of the electrode assembly. Large flat disc samples are necessary in order to obtain TSC currents of reasonable magnitude and so the contacts were made to them through circular graphite pad electrodes pressed into contact with the sample by the spring loaded reader head. The lower graphite electrode was insulated from the heater and sample cup assembly by a piece of Kapton film which was capable of standing heating cycles of up to 600°C without deterioration. To ensure good thermal contact between the sample and the heater cup assembly the mating surfaces were smeared with a layer of electrically conducting paste. (Dow experimental water soluble electroconducting resin type XD 8579.)
The current measurements can readily be made using any sensitive D.C. electrometer but since the electronic units developed for the TLD measurements included a current to pulse rate converter covering part of the necessary current range this was, where practicable, used for recording both the integral signal and the TSC equivalent to the glow curve (the TSC 'thermogram'). The output of this converter was fed into the same ratemeter, scaler and recorder system as was used for the TLD and TSEE studies. It was hoped that the digitisation of the TSC signal obtained in this way would prove advantageous in comparison with the analogue output characteristic of most previous studies of TSC, but in practice the reproducibility of the results obtained were not such that this advantage could be fully exploited. Furthermore the sensitivity of the current to pulse rate converter (two pulses per 100 pC) was only adequate for plotting TSC 'thermograms' of heavily dosed samples and thermograms made at lower dose levels had to be recorded using a Keithley electrometer in place of the modified TLD reader system.

A photograph of the complete reader system set up for the TSC measurements is shown on figure 4.1a. Figure 4.1b is a diagram of the actual electrode configuration. The circular electrodes have a diameter of 15 mm and the most satisfactory form of sample was a thin disc of appreciably greater diameter than this so that edge current leakage effects were very small. In practice, however, some samples (particularly the ceramic beryllium oxide discs) were only available as discs slightly smaller in diameter than the graphite electrodes.

4.2 Optimum working conditions

Experience with this electrode assembly showed that the saturation voltage required across the sample to ensure full charge collection could not be reached before E.H.T. breakdown occurred or the maximum voltage obtainable from the E.H.T. supply was reached. For many samples the limiting voltage was set by the onset of leakage currents
round the edges of the sample discs. To avoid the possibility of damage to the input circuit of the current to pulse rate converter being caused by bursts of leakage current a relatively robust moving mirror galvanometer was used to monitor the sample current during the setting up procedure, and the converter was only connected to the sample electrode after the E.H.T. had been reduced well below the point at which leakage currents first appeared to be observable.

An improved design of electrode assembly using some sort of guard ring structure to prevent surface currents being recorded would probably enable the readings to be repeated with substantially larger applied E.H.T. voltages. However, calculations indicate that for most samples it is likely dielectric breakdown will be reached before the TSC reaches saturation. The mean distance travelled by an electron after being released from a trap will be given by \( d = E \mu T \) where \( E \) is the field, \( \mu \) the electron mobility and \( T \) its mean life. This is not readily evaluated as both \( \mu \) and \( T \) may be expected to be temperature dependent but it can be seen that to reach saturation with a sample of thickness \( t \) requires a value of \( E \gtrsim \frac{t}{\mu T} \) for all values of temperature reached during readout. Using order of magnitude values for \( \mu T \) (~\( 10^{-9} \text{ cm}^2 \text{ V}^{-1} \)) and for the dielectric strength of the sample (~\( 10^5 \text{ V/cm} \)) the estimate of maximum sample thickness for which charge collection under saturation conditions is theoretically possible is typically of the order of 1\( \mu \)m, which is far below the minimum value practical with the electrode system described.

Initial experiments were therefore carried out to determine the variation of TSC signal with polarising voltage. Using a standard radiation dose of about 10 rad it was invariably found that within the limits of experimental error there was a linear relationship up to the point at which leakage currents become detectable (which appeared to be at about 10 kV per cm). Subsequent measurements were then all made for a fixed voltage which came
well below this point. The voltage usually adopted corresponded to a standard field gradient in the sample of 7 kV/cm in order to facilitate the intercomparison of measurements on different sample materials.

Figure 4.2 shows the linear relationship referred to above for PTFE and BeO subjected to field gradients of up to 7.5 kV per cm.

4.3 Materials for TSC dosimetry

The ideal TSC dosimeter is probably a thin slice of a single crystal of tissue equivalent material. Such a slice was prepared from a LiF crystal but was not found to be suitable for TSC measurements because the signal was masked by excessive normal (intrinsic, ionic and impurity-or extrinsic) conductivity. However it was found that commercial TID dosimeter discs consisting of lithium fluoride in a teflon matrix (Conrad type D-LiF-7-0.4) gave useful measurements. Suspecting that the teflon binding matrix was responsible for most of the signal, further tests were carried out with samples of other pure plastics which had an acceptably high melting point and low normal conductivity. These were found to show generally comparable behaviour but in all cases arbitrary changes in the TSC sensitivity of individual samples were repeatedly encountered. From among the samples tested PTFE had a great advantage in that (as can be seen from Figure 4.3) the onset of intrinsic conductivity is only important at temperatures well above those required for the readout of the principal TSC peak.

Further experience showed that it was nearly impossible to obtain consistently reproducible results over a wide range of doses using plastics - this was assumed to be due to changes in polymerisation affecting the mean electron mobility and lifetime in the sample. Such changes could be produced both by the effect of irradiation and by the heating associated with the readout cycle. It was concluded from this that the choice of samples for practical dosimetry must be restricted to samples of greater physical stability, such as single crystals or sintered compounds. Polycrystalline materials were also thought to be unlikely to readily yield
samples of consistent sensitivity and no attempt was made to evaluate their TSC characteristics.

Experimental and theoretical considerations point to the conclusion that single crystals are unlikely to form good TSC dosimeters. With these the obvious requirement is for a sufficiently large energy gap between the valence and conduction bands to give very low intrinsic conductivity at temperatures of up to 250°C. In silicon the energy gap is 1.12 eV, and in LiF it is 14 eV. However with practical samples the conductivity is dominated by the effect of impurities and the principal problem in producing suitable samples is probably obtaining sufficient freedom from impurities and lattice defects to give acceptably low intrinsic conductivity without the loss of the metastable traps on which the TSC process depends. Zone refining and annealing techniques as used with semiconductors for electronic devices should ensure acceptable intrinsic conductivity but probably only at the cost of very low residual TSC sensitivity.

This leaves sintered samples as likely to be of greatest promise for TSC measurements and BeO is one obvious near tissue equivalent dosimeter material suitable for preparation in this form.

4.4 Observations obtained with TSC dosimeters

With the PTFE dosimeters the problems of reproducibility discussed in the last section were best attacked by taking all results from the first readout of samples cut from the same sheet of plastic. Repeated use tended at first to increase sensitivity (this was attributed to the formation of additional trapping centres) but after a few readout cycles the sensitivity rose to a maximum, characteristic of the individual dosimeter, and then underwent erratic changes on further re-use. To obtain a realistic measurement of any fading of the TSC signal twenty freshly cut dosimeters were irradiated to 10 rad each, half of them were read out promptly and the other half after a week's storage in the dark. On average the reading from the second
group was 14% lower, suggesting that even if the other problems associated with these dosimeters could be overcome fading would remain a serious drawback.

Next a series of measurements were made on the group of sintered beryllium oxide samples obtained from Consolidated Beryllium Ltd. for the TSEE studies already reported. The main limitation of these samples was found to be the onset of normal conductivity at an undesirably low temperature (about 180°C). The TSC peak shown on figure 4.3 comes at about the same temperature as the corresponding TLD and TSEE peaks (140°C). The early onset of normal conductivity makes it difficult to separate the integral TSC count in the reader from the thermal background but some measurements of the TSC sensitivity of predosed BeO ceramic discs were made and these form part of the work on supralinearity which is reported in the next chapter.

It would appear that studies of normal conductivity in other BeO samples from different sources (particularly a systematic study of the effect on the normal conductivity of variations in the exact composition or the temperature at which the material is sintered), might enable samples to be obtained for which this separation could be made more effectively. This would appear to be a worthwhile subject for further investigation.

The dose response curves obtained with these materials are shown on figure 4.4. For both BeO and PTFE they take the form:

\[ R = k D^n \]

where \( R \) is the TSC response, \( D \) is the dose in rad, \( k \) is a constant and \( n \) has a value of 0.9.

This form of relationship would normally be taken to imply progressive saturation of the trapping states with increasing dose, but the fact that the index 0.9 appears to be the same for such fundamentally different sample materials as a plastic and a ceramic would make it worth looking further into this effect using a wider variety of materials.
A similar relationship between radiation dose and TSEE signal has been attributed to electrostatic effects inhibiting the escape of exo-electrons, and compounding the samples with graphite to improve conductivity has been shown by Kramer to lead to a more linear response (Section 3.3). Such an explanation cannot be readily extended to cover comparable TSC observations and it seems desirable to remain open minded about the possibility of a more general explanation based on the polycrystalline structure of the various granular, plastic or ceramic samples with which observations have been carried out.

The sheets of PTFE used for the production of TSC dosimeters are prepared by sintering material originally supplied in a granular form and photomicrographs of sections of typical extruded material before and after processing (1) show clearly that this granular structure with voids always persists to some extent after manufacture of the sheet. Further changes in the microstructure following thermal cycling must therefore be regarded as almost inevitable and ready reproducibility of results involving re-use of dosimeters should not be expected. Furthermore ICI* Ltd report that observed conductivity can depend as much on the electrode structure as on the properties of the bulk material. Observed intrinsic conductivity is attributed to 'a very low frequency relaxation loss phenomenon' rather than to steady state charge transport (steady state conductivity is not established within an elapsed time of fifteen minutes). They recommend the use of evaporated gold electrodes (1); a method which has been reported to drastically reduce the observed TSC signals (2). Evaporated electrodes were not used for any of the observations reported in this chapter and some of the variations in the expected response are almost certainly due to electrode contact effects.

* Imperial Chemical Industries, Plastics Division, Welwyn Garden City, Herts.
4.5 Conclusions and comparison of solid state dosimetry systems

For practical dosimetry applications the relatively marked fading of the signal which was discussed in the last section, represented a serious drawback. It has been suggested that this may be an inherent problem associated with non-thermal fading resulting from overlap of the wave functions characteristic of the electron trap and the ground state.\(^2\) Another major difficulty was the very erratic response which was invariably encountered when attempts were made to repeat particular sets of observations made with plastic TSC dosimeters. Partly as a result of these problems the work reported in this chapter was discontinued towards the end of 1973 after an evaporation unit for the production of thin layer TSEE dosimeters had been constructed. At this time the erratic results obtained with plastic dosimeters were attributed to changes of polymerisation resulting both from irradiation and heat cycling of dosimeter discs, effects that would mean such dosimeters would be unlikely to have any important practical applications. The author's attention has subsequently been drawn to the possibility of stabilising the dosimeters before use by a suitable annealing procedure \(^4\). It also now appears in the light of later reports that many of the difficulties experienced were due to the applied polarising fields being frozen into individual crystals within the dosimeters during the cooling following readout. This can lead to marked sensitivity changes on re-use but the problem is readily avoided if the applied voltage is removed and the dosimeters are short circuited before cooling.

The more recent work carried out by the groups under Harper and Thomas at Cardiff \(^5\), \(^6\), Moran in the U.S.A. \(^7\), \(^8\) and Charlesby at Shrivenham \(^9\) have re-established general interest in the potential applications of TSC dosimetry because if the various practical problems can be overcome the method has several important advantages:
(1) TSC dosimeter discs are likely to be much cheaper than TLD dosimeters.

(2) The reader assembly is in many ways much simpler – particularly as a light tight readout chamber is not necessary.

(3) The output from a TSC dosimeter varies linearly with the applied voltage (even this variation could be avoided if the dosimeters could be read out under saturation conditions). By contrast the output from the photo-multiplier tube in a TLD reader varies with quite a high power of the applied voltage, which consequently requires to be more highly stabilised.

(4) TLD dosimeters are not very suitable for the measurement of neutron doses. It is difficult to adjust the composition of the dosimeters to give tissue equivalence for neutrons and their sensitivity for fast neutrons tends to be disappointingly low. TSC dosimeters appear to be able to offer advantages in both these respects, and can probably be 'tailored' to offer almost exact equivalence to specific body tissues.

Because of these factors both the Department of Health and Social Security (on behalf of radiation users in the N.H.S.) and various commercial companies have now begun to take a serious interest in the likely eventual potentialities of the technique. The author nevertheless remains of the opinion that the capital and research investments which have been made in TLD dosimetry systems mean that this technique is unlikely to be supplanted for routine beta-gamma dosimetry for a very long time to come. By comparison thin layer TSEE dosimeters have potential research applications for which the thickness of TLD or TSC dosimeters renders them unsuitable. It appears that they may also help solve the problem of fast neutron dosimetry in mixed radiation fields by offering unusually good discrimination against gamma rays. It will be very interesting to see whether future developments in fast
neutron dosimetry exploit the possibilities of relatively new but known methods such as TSC, TSEE or lyoluminescence (10), or whether some other totally new dosimetric process is introduced to help meet the problems that arise in this area.

Moran et al recently prepared a table (11) (reproduced here as table 4.1) giving an instructive 'state of art' comparison of the current performance to be expected from TL, TSEE and TSC dosimetry systems. The authors personal results and conclusions correlate well with those to be drawn from this table.
CHAPTER 4: REFERENCES

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TABLE 4.1

A comparison of radiation sensitivities for techniques actually proposed for or utilised in dosimetry measurements. Dosimeter response compared for devices 1 cm² in cross-section and 1 cm thick. Response measured in terms of primary detected charge (before amplification) during readout, integrated over the strongest thermogram peak.

<table>
<thead>
<tr>
<th>Technique</th>
<th>Material</th>
<th>Response (C/rad)</th>
<th>Routine-use reliably detected minimum dose in Rads</th>
</tr>
</thead>
<tbody>
<tr>
<td>RITAC (conductance or polarization)</td>
<td>Ultrahigh-purity Al₂O₃; uv-grade sapphire</td>
<td>$5 \times 10^{-10}$</td>
<td>$3 \times 10^{-4}$</td>
</tr>
<tr>
<td>Thermoluminescence</td>
<td>Harshaw Chemical Co., CaF₂ (TLD-200)</td>
<td>$2 \times 10^{-12}$</td>
<td>$2 \times 10^{-4}$</td>
</tr>
<tr>
<td>Radiophotoluminescence (pulsed uv readout excitation for 100 sec)</td>
<td>Silver activated metaphosphate glass</td>
<td>$3 \times 10^{-12}$</td>
<td>$2 \times 10^{-3}$</td>
</tr>
<tr>
<td>TSC</td>
<td>TL doped phosphore or nominally high-purity materials</td>
<td>$5 \times 10^{-14}$</td>
<td>20-100 (noisy readout)</td>
</tr>
<tr>
<td>TSEE</td>
<td>BeO ceramic</td>
<td>$10^{-14}$</td>
<td>$10^{-4}$ (extremely noise-free readout)</td>
</tr>
</tbody>
</table>

Reproduced from Moran et al Medical Physics 1, 157 (1974)

FIGURE 4.1a

Photograph of TSC reader assembly

Reproduced from Moran et al Medical Physics 1, 157 (1974)
FIGURE 4.1b

SCHEMATIC DIAGRAM OF T.S.C. READIER ELECTRODE ASSEMBLY

Spring loaded cantilever arm for ensuring contact between sample discs & electrodes. E.H.T. CONNECTOR.

Insulating plates
Brass electrode cup

GRAPHITE ELECTRODE PADS

Kapton insulation
Copper disc to provide good thermal contact with heater assembly

T.S.C. CURRENT

Fixed base plate and heater assembly
'O' ring seal

Thermocouple
Heater Connections

Field kV/cm

Each point mean of first readout from ten discs.

FIGURE 4.2

VARIATION OF TOTAL TSC CHARGE WITH ACCELERATING FIELD

Disc samples 1.0mm thick
Test dose 10 rad
FIGURE 4.3
TYPICAL TSC THERMOGRAMS

TSC SIGNAL

FIGURE 4.4
TSC DOSE RESPONSE CURVES
Disc samples 12mm diameter x 1mm thick

Each point mean of first readout from four discs.
CHAPTER 5 : SUPRALINEARITY OF THERMOLUMINESCENT PHOSPHORS

5.1 Introduction

This chapter describes investigations carried out into possible causes of the supralinear response of some thermoluminescent phosphors and includes a brief review of some theoretical work on this topic. A reader for research into the properties of phosphors requires rather different features to one that has been optimised for us in routine dosimetry. Table 5.1 sets out these differences and shows that the Conrad 5100B reader is not very suitable for phosphor research even after the modifications described in chapter 2.

There are two different approaches to the problem of designing a reader specifically for phosphor research, one is to prepare a complete instrument with all the various special features required built into it and the other is to use a modular assembly in which individual units can be modified or replaced to meet the needs of specific projects. A good example of a reader of the first type is the Pitman(a) 'Toledo' which can be supplied with a simple plug in control unit for repetitive routine work, or a very much more sophisticated alternative intended to meet the needs of particular research projects. Probably the most widely used examples of the modular class of reader are the various versions of the Harwell 2000 series TLD system originally developed by Perry at Winfrith. This comprises a reader head containing little more than the sample heating chamber and photomultiplier assembly, together with a group of standard and special electronic modules from the Harwell 2000 series. Perry's original reader was modified from an AERE alpha counter type 1483A, and the special modules developed for the TLD reader comprised a 'DC to pulse rate' converter for digitising the PM tube current, a temperature indicator and trip unit for interfacing the reader head to

(a) D.A. Pitman Ltd., Jessamy Road, Weybridge.
the remainder of the reader system, and a temperature control module designed to permit the use of either linear or plateau heating cycles.

The reader system was completed by standard 2000 series powered bins, an EHT module, a scaler, a ratemeter and a recorder — with the optional addition of a print controller and an Addo-X printer if required. In each case any of several different modules could be used according to the particular application.

The latest commercial version of this reader, now marketed by Cirtronic Instruments, utilises a very sophisticated reader head in which the sample is heated from both sides simultaneously and the photomultiplier is cooled thermoelectrically. Improved versions of the electronic modules developed for the control of this reader have now been grouped into one unit in the format of the new Harwell 6000 modules, and this is marketed under the reference number M 8571.

In this form Perry's reader is particularly suitable for phosphor research as so many of the parameters involved are completely under the operators control and modifications can readily be made when necessary by interchanging alternative modules. For the work described in this chapter an 'in house' version based on the original Perry instrument was designed and built by the author at Aberdeen. This is described in the following section.

5.2 The Aberdeen reader

Since the range of standard Harwell 2000 modules required for this reader were already available in Aberdeen it was decided to build two special modules and to construct a simple phosphor readout head to complete a system similar to Perry's original design. It was planned to use this reader head for phosphor studies leaving the Conrad 5100B reader permanently free for routine work. In addition once the control modules were known to be operating

(b) Cirtronic Instruments Ltd., St. Peter's Road, Furze Platt, Maidenhead
correctly the intention was to purchase one of the latest Cirtronic readout heads so that this system could eventually form a modern replacement for the Conrad reader. The remainder of this section describes the development of this reader system, and figures 5.1 and 5.2 show the reader head which was constructed and the complete reader system.

The 1483 alpha counter which was modified to form the reader head is fitted with a photomultiplier mounted on a hinged plate in such a way that an iris diaphragm automatically closes to protect it from the light when the instrument is opened. For TLD measurements the PM tube anode current is monitored between anode and earth so a negative E.H.T. supply has to be reconnected to the cathode end of the dynode chain through an additional co-axial cable socket. The original sample holder was removed and replaced by a heating tray with its temperature monitored by a thermocouple. A 0.8 V 100A transformer controlled by a relay is used to heat the sample tray, and facilities for an inert gas purge of the sample chamber are also built in. Figure 5.3 shows the internal circuitry for this new reader head.

The basic control unit is a double width 2000 series module containing the D.C, to pulse rate converter and the temperature indicator and trip unit. The circuit board for the converter was built to the published circuit (1) and found to work very satisfactorily. It covers six decades, converting P.M. tube currents between $10^{-10}$ amp and $10^{-4}$ amp into pulse trains of frequencies between 2 pps and $2 \times 10^6$ pps. The unit constructed was calibrated using a scaler and Keithley electrometer. Figure 5.4 shows that the response was completely linear over the range above. No detailed published circuit of the original temperature indicator and trip unit was available but this part of the standard system consists of a chopper stabilised amplifier used to convert the thermocouple output into a moving coil voltmeter display showing the tray temperature, and simultaneously controlling the temperature channel of a Rustrak recorder. This temperature signal is also fed to two
discriminators which inhibit the scaler from recording except between upper and lower preset temperatures selected by the operator by means of helipots. Figure 5.5a shows the circuit developed at Aberdeen to reproduce these requirements which employs a commercial operational amplifier with two integrated circuit level detectors.

The two channel Rustrak recorder used in the standard form of the 2000 series TLD reader was not regarded as necessary and was replaced by a free standing single channel Rustrak recorder or optionally by an X-Y plotter as required.

The remaining non-standard module required for a full TL reader system is a temperature control unit. The original design was due to J.B. Rae (2) and gives a choice of a fast temperature rise followed by isothermal readout at a predetermined temperature or a linear servo controlled temperature increase. It employs a differential operational amplifier to compare a reference voltage obtained from either a potentiometer controlled D.C. supply or a linear ramp generator, with the amplified thermocouple voltage. The heater transformer is controlled from this amplifier by thyristors operating on alternate half cycles of the A.C. supply to the primary so that the former arrangement gives a rapid temperature rise followed by isothermal readout whilst the latter gives a linearly increasing temperature.

With this arrangement the circuitry controlling isothermal readout can be readily modified to give a multi-plateau type heating cycle incorporating preheat, read and anneal periods. This involves only using three integrated circuit timer control chips (Rastra Electronics type 555) to control the total time for each of the three heating plateaux, and three D.C. reference voltages to control the peak temperatures attained. All six of these controls can then be adjusted independently.

Figure 5.6 shows the circuit of this modified heater control module and with this refinement the complete reader assembly can form a versatile routine TLD reader
as well as a research tool. One of its special features is that it can take the same graphite sample planchets as those used for the TSEE studies reported in chapter 3. The modified reader described above was used for the investigations into supralinearity which are described in section 5.4.

5.3 Theories of Supralinearity

It has already been explained in section 1.3 that there are two possible processes which can lead to supralinearity. It can arise either through an increase in the probability of trapping at centres leading to luminescence, caused by the irradiation; or through an increased probability of luminescent recombination of the charge carriers released from traps during readout. Either of these basic processes can in turn be caused by several different types of mechanism. The most important of these can be classified as follows:

(i) Mechanisms associated with an increased probability of trapping.

(a) Creation of new traps at radiation induced lattice defects.

(b) An increase in the capture cross-section of traps following irradiation.

(c) The reduction or removal of competitors for TL traps (e.g. alternative traps from which radiationless transitions can occur).

(d) A decrease in the capture cross-section of these competing traps.

(ii) Mechanisms associated with an increased probability of luminescent recombination.

(a) The creation or activation of new luminescent centres by irradiation.

(b) An increase in the capture cross section of luminescent centres following irradiation.

(c) The removal of competing centres which lead to non-radiative recombination. (This removal may in practice be a direct consequence of such centres becoming filled up first due to
their greater cross-section).

(d) A decrease in the capture cross-section of these competing recombination centres).

Any satisfactory mechanism which is postulated must also be able to explain the irreversible falling off in sensitivity which is universally observed beyond the supralinear region in the case of very large absorbed doses and is quite distinct from the reversible effects of trap saturation. This is probably due to severe radiation damage to the crystal lattice which cannot be annealed out.

The thermoluminescent sensitivity of a phosphor can be expressed as the ratio:

\[
\text{Sensitivity} = \frac{\text{Increased TL output for small additional test dose}}{\text{Additional test dose}}.
\]

or in symbols:

\[
s = \frac{\Delta L}{\Delta D}.
\]

where \(\Delta L\) = increase in light output and \(\Delta D\) = additional dose absorbed.

The initial value of the sensitivity depends among other things upon the previous irradiation history of the sample. Once supralinearity has set in following the absorption of a large dose (either on a single occasion or cumulatively) the sample will retain an enhanced constant sensitivity during further exposure and readout as long as the additional accumulated dose is relatively small but the initial sensitivity can normally be restored again by an extended period of high temperature annealing. Phosphors which have had their normal sensitivity enhanced in this way are often said to have been pre-dosed. Measurements of the change in TL sensitivity following the predose can help differentiate between the two mechanisms for the production of supralinearity discussed above. If mechanisms of type (i) apply the number of electrons trapped per unit of absorbed dose will increase with the build up of trapping centres (or the destruction of competing traps) which takes place as a large dose is being
absorbed. Consequently on reading out this dose the phosphor will show a mean sensitivity which is a weighted average of its initial and final values.

In contrast if mechanisms of type (ii) apply the enhanced probability of luminescent recombination produced by the dose will be fully operative before readout commences and hence the final luminescent sensitivity shown will apply to all the trapped electrons. (Essentially the sensitivity of the phosphor depends upon the product of two probabilities - the probability of trapping and the probability of luminescent recombination. Either or both these probabilities may be a function of dose). If therefore a phosphor is subjected to a dose large enough to take it well onto the supralinear part of its response characteristic and its total TL light output on readout is determined, the mean output per rad (the integral sensitivity) can be expected to be about half the value of its final differential TL sensitivity where a mechanism of type (i) applies but equal to it where a mechanism of type (ii) applies.

The principle behind this method is further explained in Section 5.4 where consideration is also given to distinguishing between some of the possible mechanisms of each type. This can be done by studying the TSEE and TSC sensitivities of the phosphor and how these are affected by predosing. Section 5.5 summarises the results of experimental work carried out using these methods for the study of supralinearity.

Two important mathematical models for supralinearity have been proposed, one postulated a type (i) mechanism and the other a type (ii) mechanism. They are considered briefly in turn below:

Type (i) Model of Cameron and Zimmerman 1965 (3).

The basic parameters are:

- $R = \text{total radiation dose}.$
- $N = \text{total number of traps (filled or empty) at any level of absorbed dose}.$
- $N_F = \text{number of these traps filled at this dose level}.$
\( N_0 = \) total number of traps initially present.
\( N_M = \) maximum number of traps possible.
\( \lambda = \) proportionality constant for creation of new traps by radiation.
\( \beta = \) proportionality constant for filling of traps by radiation.

We can define \( \lambda \) and \( \beta \) by the equations:
\[
\frac{dN}{dR} = \lambda (N_M - N)
\]
\[
\frac{dN_p}{dR} = \beta (N - N_p)
\]

On integration this gives:
\[
N_p = \frac{N_0 \beta}{\lambda - \beta} \left( \exp\left(-\beta R\right) - \exp\left(-\lambda R\right) \right) +
\]
\[
N_M \frac{\lambda(1-\exp(-\beta R)) - \beta(1-\exp(-\lambda R))}{\lambda - \beta}
\]
\[
N = N_0 \exp(-\lambda R) + N_M (1-\exp(-\lambda R))
\]

For the case of lithium fluoride type TLD 100 the experimental data fitted these equations with the following values for the parameters:
\[
N_0 = 1.2 \times 10^{15} \text{ traps/cm}^3 \quad N_M = 6.0 \times 10^{15} \text{ traps/cm}^3
\]
\[
\lambda = 1.0 \times 10^{-4} \text{ rad}^{-1} \quad \beta = 1.1 \times 10^{-5} \text{ rad}^{-1}
\]

Cameron and Zimmerman showed that conjugate solutions to these equations would also exist such that \( \lambda \) and \( \beta \) are interchanged and \( N_0 \) is replaced by \( N_0 \beta /\lambda \). This ambiguity could be resolved by experimental determinations of \( N/N_0 \). Experimental observations on TLD 100 were shown to fit the equations using the parameters quoted above to within 3% at all dose levels, (see Figures 5.7a and 5.7b). Despite this very close agreement Cameron and Zimmerman pointed out that it appeared intrinsically unlikely that newly created traps produced in this way would have exactly the same depth distribution as those originally present. Since
the sensitivity of TLD 100 can be increased by up to six times with no change in the shape of the glow curve being observable they admitted that their model was probably not a completely accurate explanation of the observed supralinearity.

Type (ii) Model of Dobson and Midkiff 1970 (4)

Claffy in 1967 attempted to correlate the luminescence of lithium fluoride with absorption F-centres acting at a wavelength of about 3800A. (5) This enabled an estimate to be made of both the number of trapped electrons and the number of emitted photons per unit volume. It showed that for low doses only about one per cent of the released electrons undergo a luminescent recombination. Consequently large changes in TL sensitivity can be produced if there is a small absolute radiation induced change in the probability of luminescent recombination. (The sixfold increase of sensitivity possible with TLD 100 could for example be produced by an increase in this probability from 1% to 6%). A change of this magnitude could be readily explained by the filling up of alternative competing non-luminescent recombination centres with larger capture cross-sections.

Later Claffy et al (6) proposed a more sophisticated explanation in which luminescence is affected by the separation of the tracks of the ionizing particles. On this model supralinearity sets in when the dose is high enough for significant overlapping of tracks to take place. It is assumed that the luminescent centres are activated by the absorbed radiation and therefore produced only along the particle tracks, but that the original trapping centres can only be associated with the impurity activator atoms (Mg in TLD 100) and remain numerically constant. Claffy postulated that the response remains linear whilst holes released from the trapping centres on heating can only undergo luminescent recombination at centres produced along the track of the same ionizing particle which created them, once recombination at luminescent centres associated with
other incident particles becomes possible supralinearity sets in.

Dobson subsequently put this model on a quantitative basis (4). He supposed that there are a total of \( M \) trapping centres per unit volume and that, after exposure to a dose \( R \), \( m \) of these are occupied. Each secondary electron produced by the incident photons is assumed to contribute an average dose \( r \) to the phosphor and to create an average of \( n \) electron-hole pairs. An increase \( dR \) in the absorbed dose will then produce an extra \( n \, dR \) holes for which the probability of trapping will be \( r \) proportion to \( M-m \).

The increase in trapped holes for the dose contribution \( dR \) will therefore be:

\[
dm = \chi (M - m) \frac{n \, dR}{r}
\]

where \( \chi \) is a constant of proportion.

Integrating this gives:

\[
m_R = M \left(1 - \exp \left(-\frac{\chi n R}{r}\right)\right)
\]

If each hole has a probability \( p \) of interacting with a luminescent centre along its own track and if there are \( K \) other tracks with luminescent centres near enough for the hole to interact with, it will have a total probability of interaction \( P \) given by:

\[
P = p + p \left(1 - p\right) + p \left(1 - p\right)^2 + p \left(1 - p\right)^3 + \ldots
\]

\[
= 1 - (1 - p)^K
\]

The number of tracks sufficiently close for interaction is assumed to vary with dose in a linear manner. Hence:

\[
K = 1 + \frac{R}{R_0}
\]

where \( R_0 \) is the dose for which the probability of interaction with a centre in a second track is equal to that for
interaction with a centre in the original track.

Denoting $X_n R$ by $\lambda$, the total integrated light output
at saturation by $I_{10}$, and that for dose $R$ by $I$ these equations
yield the relationship:

$$I = I_0 \left(1 - X K\right) \left(1 - \exp \left(-\frac{\lambda R}{R_0}\right)\right)$$

$I_0$ will be directly proportional to the total number
of trapping centres $M$.

Dobson fitted this model to Cameron's experimental
data with the assumption that tracks only begin to inter-
act at separations of the order of $10^{-8}$ to $10^{-9}$ m. This
 corresponds to a photon flux of $10^{12}$ to $10^{14}$ cm$^{-2}$. Taking
an average of these values indicates that the value of
$R_0$ should be about 4000 rad. In the case of lithium
fluoride this is not far into the supralinear range as
can be seen from figure 1.17 which shows Cameron's data
for $^{137}$Cs 660 keV gamma rays. Dobson therefore assumed
that the equation for $m_R$ could be approximated to by a
linear relationship $m_R = \lambda M$ with $\lambda$ constant. $I_0$
was shown to be 478,000 photons/cm$^2$ for TLD 100 and a least
squares fit for the other parameters gave:

$$\lambda = 0.215$$
$$R_0 = 5,135 \text{ rad}$$

Track separation for dose $R_0 = 3 \times 10^{-9}$m
Probability of luminescent recombination for
thermally released holes at low doses = $p = 5\%$.

As with Cameron's model there is a fundamental
difficulty in accepting this model. It gives an almost
entirely geometrical explanation for the point at which
supralinearity would be first observed and is therefore
not readily adapted to explaining why supralinearity in
different phosphors can be observed at dose levels above
minimum values which may vary over the range from about
200 rad up to 10 kilorad or more.

A detailed review of the track interaction model
for lithium fluoride has been given by Attix (10).
5.4 Application of Sensitivity measurements to the study of supralinearity

\[ M = \frac{\text{Integral TL response for dose } D}{\text{Total measured TL output}} = \frac{\text{Total measured TL output}}{\text{Total absorbed dose}} \]

or in symbols \( M = \frac{L}{D} \)

\[ S = \frac{\text{Differential TL Sensitivity}}{\text{Measured TL output for small test dose}} = \frac{\text{Test dose absorbed}}{\text{Test dose absorbed}} \]

or in symbols \( S = \frac{\Delta L}{\Delta D} \)

The value of \( S \) depends, among other factors, on the previous irradiation history of the sample. Denote the initial value for a previously unirradiated sample by \( S_I \).

Then \( S_I = \frac{\Delta L}{\Delta D} \) when predose \( D_p = 0 \)

Similarly denote the value for a sample previously subjected to an absorbed dose \( D \) and thermally drained, by \( S_D \)

Then \( S_D = \frac{\Delta L}{\Delta D} \) (\( D_p = D \))

Either of two basic mechanisms (i) or (ii) which can be responsible for supralinearity and were discussed in section 5.3 may lead to \( S_D \) differing from \( S_I \).

Typically due to one or other of these mechanisms it is found that for many TL phosphors which show supralinearity the differential TL sensitivity increases linearly with the absorbed dose. This gives a relationship:

\[ S_D = S_I + kD \] (\( k = \text{constant} \))

Ratio of final to initial sensitivity:

\[ \frac{S_D}{S_I} = 1 + \frac{k}{S_I} \] \( D \) \( \ldots \ldots \ldots \) Eqn. A
If mechanism (i) is operative more charges are captured at luminescent centres per unit dose as the radiation exposure to which the phosphor has been subjected increases.

The total stored luminescence for a dose \( D_0 \) will be given by an expression of the form:

\[
L = \int_{D=0}^{D=D_0} dL = \int_{D=0}^{D=D_0} S_D dD
\]

where \( S_D \) is the TL sensitivity after exposure to the dose \( D \)

Substituting for \( S_D \) gives:

\[
L = \int_{D=0}^{D=D_0} (S_I + kD) dD = S_I D_0 + k D_0^2 \]

\[
\frac{L}{D_0} = \text{Integral TL response for dose } D_0 = S_I + k \frac{D_0^2}{2}
\]

\[
\frac{M_{D_0}}{S_I} = 1 + k \frac{D_0}{2S_I} \quad \text{Eqn. B}
\]

A comparison of equations A and B show that in this case the integral TL response for samples subjected to a large dose will only increase half as rapidly with dose as the differential TL sensitivity following a corresponding predose.

Now consider the effect of mechanism (ii)

Total TL response for dose \( D_0 \) is given by

\[
L = S_D D_0
\]

i.e.

\[
M_{D_0} = \frac{L}{D_0} = S_D = S_I + kD
\]

or

\[
\frac{M_{D_0}}{S_I} = 1 + k \frac{D_0}{S_I} \quad \text{Eqn. C.}
\]
A comparison of equations A and C show that in this case the integral TL response for samples subjected to a large dose will, increase at the same rate as the differential TL sensitivity following a corresponding predose.

It follows from this analysis that a detailed study of the TL response of a particular material can indicate the broad process by which supralinearity arises. Further studies involving intercomparisons of the TL, TSEE and TSC properties of the material can help elucidate the specific mechanism which is responsible in each particular case.

Possible type (i) mechanisms are:
(a) Creation of additional traps.
(b) Increase of capture cross-section of existing traps.
(c) Destruction or saturation of competing stable traps which do not lead to TL emission.
(d) Reduction of capture cross-section of such competing traps.

Possible type (ii) mechanisms are:
(a) Creation or activation of additional luminescent recombination centres.
(b) Increase of capture cross-section of luminescent centres.
(c) Destruction or saturation of non-radioactive competing recombination centres.
(d) Reduction of capture cross-section of such competing recombination centres.

For type (i) mechanisms effects (a) and (b) should lead to more rapid trapping and hence a lower carrier density during irradiation whilst effects (c) and (d) should lead to the reverse. It should in principle be possible to distinguish between these effects by conductivity measurements. Any such mechanism should lead to an enhancement of both TSC and TSEE sensitivity corresponding with that for the TLD sensitivity. (Assuming the surface traps respond in the same manner as those in the bulk volume). For type (ii) mechanisms effects (a) and (b) should lead
to more rapid luminous emission during readout and hence to a lower carrier density at this time whilst effects (c) and (d) should lead to the reverse. In these cases, where the carriers are free electrons, there should be an associated decrease or increase in both the observed exo-electron emission readout and the TSC signal as these should be directly related to the free electron density in the sample.

This method was originally introduced by J. Thompson for the study of supralinearity in quartz (7). It is applied here to three types of sample, lithium fluoride, sodium fluoride and beryllium oxide. The results appear in the next section.

5.5 Results for particular phosphors
5.5.1 Lithium Fluoride

Both differential and integral TL sensitivity grow approximately linearly with predose up to about 30 krad. Beyond this the response saturates and the sensitivity falls. For larger doses it is well known that there is an irreversible loss of sensitivity due to radiation damage to the crystals. Below 30 krad figures 5.8 and 5.9 show that as the dose increases the mean TL signal per rad (integral sensitivity) grows at a rate very close to half that for the differential sensitivity, pointing strongly to the formation of new trapping centres as the explanation for the supralinearity. This explanation is supported by the observation that the TSEE differential response also increases with predose and, although the change in TSEE response is less marked than that in the TL response for doses below the level at which saturation sets in, it continues to increase with absorbed dose up to at least 1 Mrad. It follows that the effect of radiation damage must be limited to the recombination kinetics not the trapping centres. Furthermore there is no exact correlation between the behaviour of the exo-electron sensitive surface layers of the crystals and the bulk properties. It appears that the formation of new trapping centres in
these surface layers take place very much less readily than in the interior of the crystals so that much larger doses are needed to produce a given degree of TSEE suprilinearity. The presence of a relatively insensitive surface layer in which trapping centres appear to be formed less readily, has previously been postulated for lithium fluoride crystals from the reduction in TL sensitivity and change in glow curve structure observed with small grain samples (8), (9).

In principal it should be possible to further confirm the model for suprilinearity proposed above by demonstrating that the TSC response of the crystal also increases with predose as a result of the increased trapping. To attempt to check this a disc of lithium fluoride in the form of a single crystal was tested, but the check proved impractical because the high intrinsic conductivity of lithium fluoride at elevated temperatures masked the changes in TSC to the point where no useful readings could be obtained.

5.5.2 Sodium Fluoride

The total TL output begins to saturate at a dose level of 1 krad and reaches a maximum at 100 krad. This means that the integral sensitivity falls off steadily for doses above 1 krad. However the differential TL sensitivity still continues to increase with predose up to 10 krad and in this instance is associated with an even larger increase in the TSEE sensitivity, which also reaches a maximum for predoses of about 10 krad. These results are shown on Figures 5.10 and 5.11.

This points to the enhanced sensitivity being due to the reduced likelihood of non-luminescent recombination during the readout process - probably as a result of some of the recombination centres being filled or destroyed during the predose procedure. Although the changes in TL and TSEE sensitivity correspond qualitatively there are again quantitative differences indicating that the properties of the bulk crystals differ in some respects
from those of the exo-electron sensitive surface layers.

It was not possible to confirm these conclusions by TSC measurements as sodium fluoride samples could not be obtained in a suitable form for observations of their TSC properties.

5.5.3 Beryllium Oxide

Both the differential TL sensitivity and the integral TL sensitivity increase at the same rate, approximately linearly with dose up to about 10 krad. This points to supralinearity arising from increased luminescent efficiency. The TSEE sensitivity falls with predose, this is compatible with a model in which supralinearity arises from the creation by the incident radiation of additional luminescent centres which compete for the electrons freed by heating. These results are shown on Figures 5.12 and 5.13.

These TL and TSEE observations were made on powdered beryllium oxide samples. TSC measurements were also carried out using sintered ceramic beryllium oxide discs in order to find out how the sensitivity of the samples changed after predosing and thermal draining. The difference in sample composition prevents anything except general qualitative conclusions being drawn from these studies but the results, with the other discussed in this section, are all summarised in table 5.2.

5.5.4 Conclusion

The general conclusions from the observations on these three sample materials is that supralinearity is not a simple phenomenon with a single consistent explanation, but one which must be investigated in detail for each phosphor in which it can be observed.
CHAPTER 5: REFERENCES

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5.7 THOMPSON, J. D.Phil. Thesis Oxford University 1970
5.9 CHAN, F.K., BURLIN, T.E. Health Phys. 18 325 1970
<table>
<thead>
<tr>
<th>TABLE 5.1</th>
<th>TABLE 5.2</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>COMPARISON OF TL READER REQUIREMENTS FOR ROUTINE</strong></td>
<td></td>
</tr>
<tr>
<td><strong>Dosimetry and Phosphor Research</strong></td>
<td></td>
</tr>
<tr>
<td><strong>Unit or</strong></td>
<td><strong>Routine dosimeter</strong></td>
</tr>
<tr>
<td><strong>component</strong></td>
<td>reader</td>
</tr>
<tr>
<td><strong>Heat controller</strong></td>
<td>Multi-plateau readout with preheat, read, and anneal zones</td>
</tr>
<tr>
<td><strong>Signal collection time (read-out time)</strong></td>
<td>As short as possible</td>
</tr>
<tr>
<td><strong>Readout chamber design</strong></td>
<td>Infallibly reproducible geometry and heat transfer with geometrically identical samples</td>
</tr>
<tr>
<td><strong>Current measuring system</strong></td>
<td>Linearity of response over six or more decades necessary with very high stability</td>
</tr>
<tr>
<td><strong>Temperature monitoring</strong></td>
<td>Not necessary</td>
</tr>
<tr>
<td><strong>Glow curve facility</strong></td>
<td>Not necessary</td>
</tr>
<tr>
<td><strong>Adjustment of controls</strong></td>
<td>Should not be too easy. Any departure from normal settings must be drawn clearly to the operators attention. Precise resetting of controls after any variation is essential</td>
</tr>
<tr>
<td><strong>Reference light source</strong></td>
<td>Must offer long term stability - must be readily brought into check position</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Measurement</th>
<th>LiF</th>
<th>NaF</th>
<th>BeO</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Mean TL per rad</strong></td>
<td>Increases up for large doses to 30 krad</td>
<td>No increase.</td>
<td>Increases up in at 1 krad and is complete at 100 krad</td>
</tr>
<tr>
<td><strong>TL Sensitivity</strong></td>
<td>Increases with preheat rate at twice the preheat to rate for the mean TL response</td>
<td>Increases at same rate as mean TL response</td>
<td>Increases with increasing predose</td>
</tr>
<tr>
<td><strong>TSEE Sensitivity</strong></td>
<td>Rises more slowly than TL sensitivity with increasing predose</td>
<td>Falls with increasing predose</td>
<td></td>
</tr>
<tr>
<td><strong>Integral TSC Sensitivity for large doses</strong></td>
<td>Not measurable sample</td>
<td>Decreases with increasing dose from very low dose levels</td>
<td></td>
</tr>
<tr>
<td><strong>Differential TSC sensitivity</strong></td>
<td>Not measurable sample</td>
<td>Decreases with increasing dose</td>
<td></td>
</tr>
<tr>
<td><strong>Origin of supralinearity</strong></td>
<td>Trapping process</td>
<td>Recombination process</td>
<td>Recombination process</td>
</tr>
<tr>
<td><strong>Suggested mechanism</strong></td>
<td>Creation of new traps by radiation damage</td>
<td>Saturation of non-luminescent centres</td>
<td>Creation of non-luminescent centres</td>
</tr>
</tbody>
</table>

**Note:** The LiF used for these measurements was 'Extrupure' grade supplied by British Drug Houses Ltd. The crystalline NaF and BeO were also supplied by B.D.H. Ltd. The ceramic BeO discs used for the TSC measurements were supplied by Consolidated Beryllium Ltd. of Milford Haven.
FIGURE 5.1
Photograph of 1483 TLD reader head

FIGURE 5.2
Photograph of Aberdeen 2000 Series TLD reader
FIGURE 5.3
Circuit of 1483 TLD Reader Head

FIGURE 5.4
Response of current to pulse rate converter board built into Harwell 2000 TLD Reader control module.
Circuit of readout control module built for Aberdeen TLD reader

FIGURE 5.5b
Photograph of readout and heating cycle control modules
FIGURE 5.6

Schematic circuit of heating control module for Aberdeen TLD reader.

FIGURE 5.7a

Cameron and Zimmerman's model for supralinearity

$^{137}\text{Cs-}\gamma$

$\text{x-ray 60keV}$

Experimental values of sensitivity ratio for LiF (TLD 100)
From Dopke & Cameron 1966.

FIGURE 5.7b

Trapping centre ratio $N/N_0$ for LiF (TLD 100)
From Cameron and Zimmerman 1968.

ALTERNATIVE HEATING CYCLES GENERATED

FIRST TIMER: Closes R1 from 0 to $t_1$ sec. Starts timer T2. Starts count.
SECOND TIMER: Closes R2 from $t_1$ to $t_2$ sec. Starts timer T3. Stops count.
THIRD TIMER: Closes R3 from $t_2$ to $t_3$ sec. Starts timer T4.
FOURTH TIMER: Operates reset relay at $t_4$ sec. Operates 'End of cycle' indicator.
FIGURE 5.8 - Relative Integral Sensitivity of Lithium Fluoride

FIGURE 5.9 - Relative Differential Sensitivity of Lithium Fluoride
FIGURE 5.10 - Relative Integral Sensitivity of Sodium Fluoride

FIGURE 5.11 - Relative Differential Sensitivity of Sodium Fluoride
Figure 5.12 - Relative integral sensitivity of Beryllium oxide.

Figure 5.13 - Relative Differential Sensitivity of Beryllium Oxide
CHAPTER 6: MISCELLANEOUS INVESTIGATIONS CARRIED OUT USING SOLID STATE INTEGRATING DOSIMETERS

6.1 Introduction

This chapter contains reports of a number of relatively short investigations carried out in the Department of Medical Physics of Aberdeen University during the past four years which have involved the use of solid state integrating dosimeters. Most of these have related to dose measurements made in connection with medical radiological procedures but studies of the variation in the angular responses of personnel dosimeters and measurements of environmental radiation doses are also reported.

6.2 Angular response of personnel monitoring dosimeters

In connection with the TLD-film badge intercomparison reported in chapter 2 it was useful to know the directional sensitivity of the two types of dosimeter. A simple perspex irradiation jig was therefore constructed for holding dosimeters at a known angle to an incident radiation beam, (Figure 6.1). It was used under as scatter free conditions as possible with both the film badges and TLD dosimeters and also with TSEE evaporated layer dosimeters.

Measurements were made with 60 kVp X-rays from a Siemens Heliotron portable X-ray unit with a 1 mm Al filter, and with 2.27 MeV (max) beta particles from a $^{90}$Sr-$^{90}$Y radioactive source. All results are shown on the polar diagrams of figure 6.2. With X-rays the TL and TSEE readings show a minimum response on the polar diagram for angles of incidence of the radiation on the surface of the dosimeter of zero and 90°. The minimum for normal incidence can be explained by the higher probability of radiation passing through the dosimeter without interaction, that for angles of incidence of 90° is due to the effect of self shielding which arises when the absorption length of the radiation is comparable with
the detector dimensions. For film the X-ray response is more complex the response curve showing an additional minimum for angles of incidence of about 60° which is not so readily explained.

It is interesting to note that this anomalous film response was previously observed by Brunskill (1) who was inclined to attribute it to experimental error and commented that the film badge response "does not appear to follow any well defined pattern". The explanation of the observed 60° minimum may be associated with the angular distribution of the photo-electrons and Compton scattered electrons in the emulsion, the sensitivity being a minimum when both photo-electrons and scattered electrons have tracks orientated obliquely across the emulsion rather than lying in its plane.

For the beta radiation the response of all three types of dosimeter decreases with increasing angle of incidence. The variation proved to be surprisingly large in the case of the films but was even larger for the TSEE dosimeters. This is to be expected as the very thin sensitive layer of the latter must make them more sensitive to track orientation and outscattering.

For personnel moving around in an area with a complex radiation field configuration variations in the angular response of dosimeters will have little or no effect; but where repetitive 'production line' type operations are involved it may lead to a regular over or under estimation of the dose received. This applies equally to the photon response, if for example the individual concerned is normally handling gamma emitters in a fume cupboard with a standardised experimental layout; and to the beta response, if for example a beta emitting radiopharmaceutical is being routinely injected. Although these effects are not likely to be a serious problem in routine personnel monitoring, additional work to further elucidate the complex response of the film badge would clearly be of interest.
6.3 Environmental dose measurements

Aberdeen is noted as a city with a relatively high radiation background level, largely because of the extensive use of local granite for both private and public buildings. Many enquiries have been made, both to the author and to his department, about these levels and how they compare with corresponding values in other parts of the U.K. It was therefore decided to carry out a limited survey from which some definite figures could be obtained. Two principal factors contribute to additional personal radiation exposure of individuals living or working in granite buildings; these are the gamma rays emitted from the long lived uranium and thorium isotopes present in the granite and the inhaled radon released from it into the air breathed by these individuals. Both of these factors were investigated, measurements of the former are directly relevant to the subject matter of this thesis. These were carried out by placing integrating thermoluminescent dosimeter elements at points of interest for periods of approximately eight weeks. Simultaneously similar measurements were carried out in a few other parts of the country where relatively low background readings would be expected.

A small supply of sintered dysprosium activated calcium sulphate TL dosimeter pellets (kindly made available by the Danish Atomic Energy Commission at Riso) were tested with the modified Conrad 510OB TLD reader and found to have excellent characteristics for environmental dose measurements - a combination of very high sensitivity, low fading and relatively low background. These were used in conjunction with Conrad CaSO₄(Dy)-teflon disc dosimeters for the series of environmental dose measurements reported in table 6.1.

It will be seen from this table that the background gamma dose received in a granite house is typically more than double what is to be expected in one built from materials of lower natural radioactivity, and that consequently the average natural background dose received in a
year may be almost twice as great in Aberdeen as in most other parts of the U.K. However, this difference is of little radiobiological consequence. Even in monazite sand areas where the background radiation levels are an order of magnitude greater than normal there has only been one serious report suggesting any observable effect on the health of the local population (2); and houses built of bricks formed from uranium mine tailings or containing some types of contaminated gypsum plaster board or cement aggregate have been shown to have much higher gamma dose levels inside than any granite building without perceptible adverse effects. (3); (4), (5).

A more serious problem is likely to be the release of radon into the building from the granite walls. In poor ventilation conditions a relatively high radon level can build up. It leads not only to a direct gamma ray dose to lung tissues but also and more important to the deposition of long life alpha emitters when the radon decay takes place within the lungs. It was therefore decided that a series of radon level measurements would be made so that this hazard could also be evaluated. One method of radon measurement now used in mines is by means of a TSEE dosimeter which integrates the alpha dose from the radon decay products collected on a filter paper through which a known volume of air has been pumped (6). This method has the advantage that it can give a direct reading of the atmospheric working levels, which are defined in terms of the total alpha energy emitted by radon daughter products in each litre of air (1 WL corresponds to any combination of daughter products which will ultimately release $1.3 \times 10^5$ Mev of alpha energy (7).) Unfortunately attempts to use it failed because the maximum area samples which could be accommodated in the TSEE reader were too small to give an acceptable signal to background ratio for samples collected in buildings. Instead an M.Sc. project student at Aberdeen (Miss M. Money) set up a conventional radon monitor using a surface barrier solid state detector to count the alpha activity of the daughter products collected
on the filter paper. Accurate estimates of radon levels from such results involve a measurement of the ventilation rate and relatively complex calculations which are frequently computerised (8), (9), (10), but for the purpose of comparing the radon and external gamma ray hazards from granite it can be assumed that radioactive equilibrium exists, that the counter has 50% efficiency (2n counting) and that there is complete collection of the daughter products from the air sampled. (Unattached daughter ions, usually estimated to be at least 10% (10), (11), will not be collected on the filter paper). All these assumptions mean that the figures reported in table 6.1 will be underestimated. Nevertheless it can be seen that radon build up under poor ventilation conditions can be quite high. Calculations of the dose to which it can give rise depend upon the lung model assumed, two recent estimates of the annual dose to lung tissue from continuously breathing air containing a radon concentration of 1 pCi/litre are 0.11 rad and 1.38 rad (12), (13). This is predominantly an alpha dose but it is thought that the appropriate quality factor does not exceed three, even so the radon levels reported here imply annual radiation dose levels to the segmental bronchioles of between 1 and 10 rem - at least an order of magnitude greater than the corresponding external gamma dose.

6.4 Calibration of LiF TLD and TSEE Dosimeters for beta dosimetry

For much of the work described in this thesis the response of both TLD and TSEE dosimeters to known doses was determined using a beta source with the specially constructed irradiation jig shown on figure 6.3. This consisted of a perspex dosimeter holder (forming an approximately tissue equivalent backscatter plate of infinite thickness), a series of known dimension perspex spacer rings and a perspex source holder designed to accommodate a Radiochemical Centre 20 mCi $^{90}$Sr beta plaque source in an accurately defined recess immediately over the sample being irradiated.
The successful use of this jig requires full knowledge of the response of the dosimeter elements to both photon and beta radiation. This was therefore studied both theoretically and experimentally. Calibrations were carried out with both a 60Co source and an X-ray machine using an EIL 37C ionization chamber dosimeter; as well as with the beta source using a specially constructed extrapolation chamber.

The X-ray calibrations were carried out at 70 kVp with a filtered (1 mm Al) Watson diagnostic X-ray set. Electronic equilibrium in the dosimeter elements was ensured by placing these in a near tissue-equivalent irradiation chamber (Figure 6.4) machined from reactor grade graphite and covered with a pivoting graphite lid. Using this the ratio of the energy absorbed per unit mass in the dosimeter element relative to that in soft tissue is given by the ratio of the mass (energy) absorption coefficients. With gamma radiation of 1.0 MeV photon energy these coefficients are 0.0258, 0.0268 and 0.0306 for LiF, teflon and muscle tissue respectively* so that a dose of one tissue rad would correspond to an energy absorption of 100 x \( \frac{0.0258}{0.0306} \) ergs per gm. in the LiF, i.e. to 84.3 erg per gm. With a diagnostic quality X-ray beam the effective mean photon energy is somewhat uncertain and the cross-sections of the activator impurities in LiF (magnesium and titanium) vary sharply with energy so that it is not realistic to quote a precise value for the mass (energy) absorption coefficient of LiF. As Attix has pointed out the relative amounts of \(^6\)LiF and \(^7\)LiF will also have a significant effect on this coefficient (14). Typically however, a dose of one tissue rad will produce an energy absorption in lithium fluoride of between 95 and 110 erg/gm. It follows that if 1 MeV photon energy radiation is used to calibrate the dosimeters the TL emission associated with the absorption of 84.3 ergs per gm of radiation energy is interpreted as corresponding to a

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radiation dose of 1 tissue rad, but that when making measurements of X-ray doses the actual energy absorbed per tissue rad may differ appreciably from this.

The corresponding figure for the beta dose calibration depends upon the thickness of the dosimeter elements. For thick dosimeters in which the beta particles are completely stopped the total energy absorbed per unit area (gm rad/cm²) will be independent of the medium. This means that where a dosimeter replaces tissue, although the range of the beta particles (expressed as mass per unit area), is about 10% greater in both LiF and teflon than in tissue the total energy absorbed per unit area of surface must be the same. Therefore for thick layers (i.e. greater than the beta range) of tissue and LiF, of equal mass per unit area, the mean dose to both will be identical so that a one rad mean dose to tissue will correspond to a dosimeter dose of 100 erg/gm, compared with the 84.3 erg per gm for photon irradiation. Thus a photon calibrated dosimeter will appear to over-read by 100/84.3 or 1.18.

For low and medium energy beta emitters this situation is approximated to by 0.4 mm thick LiF-teflon dosimeters, the approximation becoming perfect where the range of the maximum energy beta particles is less than the thickness of the dosimeter elements. In practice this applies where the maximum energy of the beta particles is not greater than about 100 keV. For the 90⁰Sr-90⁰Y source which was used for most of this work the range of the maximum energy beta particles is about 350 mg/cm² and the exit dose in a LiF-teflon thermoluminescent dosimeter disc is about 50% of that at the front surface. The irradiation conditions are then more close to those which arise when using low energy beta emitters in conjunction with evaporated layer LiF TSEE dosimeters. With these the dosimetric layer is so thin that the bulk of the energy absorption always takes place in the substrate. Assuming there is not an abnormal backscatter contribution to the dose in the evaporated layer this will then be given by the product of the soft tissue dose and the ratio of the mass stopping power.
(m.s.p.) of the LiF to that of the tissue. The value of these mass stopping powers varies appreciably with the beta particle energy but their ratio \((\sim 0.82)*\) does not \((15)\). The energy transferred per gm to the LiF in the layer for a one rad dose to the tissue is therefore, within experimental error, the same as in the gamma calibration procedure so no correction is required to the observed dose (based on TL output) in this instance.

To check these assumptions direct measurements of the beta ray dose rate were made with a Failla type extrapolation ion chamber \((16)\) designed for use with the EIL 37O electrometer dosimeter. This chamber is shown on figures 6.5 and 6.6. It has an aluminised rear electrode of 3.5 cm\(^2\) area surrounded by a guard ring and deposited on a perspex absorber. The front window is aluminised mylar approximately 1 mg/cm\(^2\) thick mounted onto a perspex tube in which the rear electrode assembly fitted. Instead of using the conventional micrometer screw head for varying the separation, a simple spring loaded fitting presses the rear electrode forward so that lugs on it are located against a perspex spacing piece in any one of five positions which can be selected by drawing the electrode back and rotating it. These give automatic inter-electrode spacings of 0.5, 1.0, 1.5, 2.0, and 2.5 mm. Normally the first four of these are used and measured currents are normalised to the value for the 1 mm spacing where the chamber volume is 0.35 cm\(^3\) before they are extrapolated to zero. Doses can then be obtained directly from the 35 cm\(^3\) gamma ray chamber calibration of the EIL electrometer by multiplying the scale reading by 0.01. Corrections for temperature and barometric pressure variations can be made equally easily by using the chart provided with the EIL instrument.

With small extrapolation chambers an additional correction for groove depth is sometimes needed \((17)\) but

\[
\frac{\text{m.s.p. (LiF)}}{\text{m.s.p. (tissue)}} = \frac{1.534}{1.874} \text{ at } 1 \text{ MeV}
\]
the chamber described above is not small enough to make this correction necessary for the work described here.

Two sets of measurements were made with this new extrapolation chamber. First a depth dose curve for the $^{90}\text{Sr}$ beta ray source was determined using a series of layers of 50 mg/cm$^2$ tissue equivalent absorber (in this instance, polythene). This is shown in figure 6.7.

Next absolute measurements of the dose rate in air produced by the source were made with 1.5 and 10 cm separation between the front electrode and the surface of the source. (These distances corresponded to the three principal spacers available with the beta irradiation jig shown in figure 6.3). The results form table 6.2. They enabled sets of five TLD and TSEE dosimeter discs to be irradiated to a standard one rad mean dose using the beta source and irradiation jig and allowing for attenuation by use of the depth dose curve already prepared. Further sets of five dosimeters made up from the same batches were irradiated to the same dose using the $^{60}\text{Co}$ source and the diagnostic X-ray unit respectively. With the latter the doses were measured using the EIL 37C dosemeter and standard 35 cm$^3$ photon ionisation chamber. A comparison of the readout obtained with these three groups of discs showed that for both the TLD and the TSEE dosimeters the response was, within experimental error, the same.

Although the theory above indicates that the X-ray calibrated dosimeters should over-read when compared with the gamma calibrated group the situation is complicated by the fact that the ionisation chamber measures rontgen and the $f$ factor (rad/rontgen) for soft tissue falls by about 5% for the X-rays. Thus although with X-rays a tissue rad corresponds to a larger energy absorption in the dosimeter than with gamma rays, it also requires a greater exposure in rontgen, giving a larger ionisation chamber response to partly compensate the greater dosimeter signal. This helps explain why no significant difference could be observed between the mean readout from these two groups of
Comparing the beta and gamma irradiated groups it is clear that both the TSEE and TLD dosimeters are acting as thin layer devices for which equivalent responses to both radiation sources would be expected. It follows that when comparing results obtained with these three sources of radiation the only correction which is necessary is for attenuation of the beta radiation in thick samples.

6.5 Skin dose measurements in mammography using TSEE dosimeters

Mammography is unquestionably a valuable diagnostic aid for the confirmation of breast cancers but it requires exceptional radiographic skill and care to obtain good mammograms which can be interpreted accurately, and quite commonly a large proportion of repeat examinations are found to be necessary. Consequently there is considerable controversy over how far its use should be extended to the screening of asymptomatic groups of women. Advocates of this recommend a six monthly mammographic examination of all women from early middle age until about 60, to cover the period where the risk is greatest. This would lead to a very large accumulated skin dose even when the best facilities and techniques were employed and it is felt by other experienced radiologists that the overall effect of such a programme could well not be beneficial.

A recurrent problem with mammography is to ensure that slipshod techniques do not creep in. It is difficult to check the doses concerned routinely with TL dosimeters as even the bare dosimeter disc casts an appreciable shadow on a mammogram. The use of the ultra-thin evaporated layer TSEE disc dosimeters would clearly be an advantage in this respect and an investigation was therefore made into the suitability of these devices for the measurement of skin doses in mammographs.

In general a mammographic examination consists of two radiographs of each breast - a supero-inferior (or craniocaudal view) and a lateral view. The positions for
these are shown on figure 6.8. The skin nearest to the X-ray tube receives the highest dose as the soft X-ray beam is rapidly attenuated - some overlap from the different exposures is inevitable.

The mammography unit at the Aberdeen Royal Infirmary employs a Machlett molybdenum anode tube. Until recently each exposure was typically 400 mA sec for an average size breast, the maximum exposures made were twice this.* The usual voltage is 35 kVp but up to 42 kVp is sometimes needed with large breasts. Measurements of the corresponding doses were made on a perspex phantom using TSEE dosimeters formed by evaporating LiF onto a 75 μm thick sheet of aluminised kapton film. Since this phantom was too rigid to enable the usual radiographic compression technique to be used it was not possible to determine the areas of overlap for the different exposures from the phantom measurements. The measured values of the individual doses for each exposure are reported in table 6.2. Both the skin dose and the depth dose under 1 cm of soft tissue were determined. The latter was obtained by covering the dosimeter element with a 1 cm layer of Mix D tissue equivalent wax. It can be seen that skin doses vary between 2.5 and 8 rad whilst depth doses at 1 cm are only about one quarter of this.

It is reasonable to conclude that an average mammographic examination will involve a maximum skin dose of at least 5 rad (with doses outside the area where the separate exposures overlap only half this value and those on the exit side very low indeed), but that with large breasts the maximum dose may well be increased to a value of about 16 rad. These figures agree well with TLD measurements made by Palmer et al in the U.S.A. (18) and serve to emphasise the undesirability of unnecessarily frequent mammographs.

A recent review paper by the author (19) estimates

* General use is now made of a Du Pont low dose vacuum packed film-single screen combination specially developed for mammography. With this combination the maximum exposure has been reduced to 300 mA sec, corresponding to a maximum dose of about 0.5 rad per radiograph.
the potential risk of inducing a breast cancer by an annual mammographic examination between the ages of 35 and 60 as being of the order of 1.5 times the natural incidence.

6.6 Depth dose measurements with TSEE dosimeters

An obvious application of thin layer TSEE dosimeters is to the measurement of radiation dose in regions where a steep dose gradient exists. Examples are interface doses (such as for example, the problem of measuring the dose received by the osteo-progenital cells of the periosteum at a bone-soft tissue interface which was discussed in Section 1.4) and doses immediately adjacent to the surface of extended area radiation sources. To illustrate such measurements a build up curve and two central axis depth dose curves, measured for different field sizes on an A.E.I. 'Orbitron' $^{60}$Co radiotherapy unit, are shown. (Figures 6.9 - 6.11). These measurements were made using lithium fluoride TSEE dosimeters evaporated onto kapton film and stacked in a perspex tube inserted into a large block of 'Mix D' tissue equivalent wax. The spacers were teflon discs cut from a rod. It was shown in section 6.4 that teflon has almost identical gamma ray absorption characteristics to lithium fluoride so interface effects between the teflon disc and the sensitive LiF surface of the dosimeter should be minimal.

Radiation dose distributions arising from radiotherapy treatment units have been very intensively studied and are exceptionally accurately known. The results shown here agree closely with the expected values. Their function is to demonstrate the potential value of thin film solid state dosimeters in enabling quick and accurate assessments of such radiation fields to be made. The observations obtained fulfill this function admirably.

6.7 Comparison of male and female gonad doses in diagnostic X-ray examinations of the abdomen

These measurements were made by an M.Sc. project student* using a specially constructed phantom based on a

* H. Ghara'ati M.Sc. University of Aberdeen 1975
fibreglass shell from a dress shop window dummy. A set of pelvic bones were cemented together and placed in their correct position in the shell. This was then filled with a tissue equivalent preparation based on gelatine and sugar with the addition of an anti-fungicide to prevent the development of mould in the medium. Tubes, plugged by polythene rods, were fitted into the phantom to enable dosimeters to be introduced at positions approximating to the female ovaries. This phantom, which is shown on figure 6.12, was then used to determine the ovary dose for anterior-posterior (A-P), posterior anterior (P-A) and lateral pelvic radiographs at a range of voltages from 60 kVp to 130 kVp. Simultaneously other dosimeters were used to determine the skin dose, the male gonad dose and the film cassette dose.

It is commonly assumed that the female gonad dose will increase with increasing penetration of radiation, an effect accentuated by falling film contrast and sensitivity which mean that for corresponding radiographic quality cassette exposure has to be increased with increasing kV. Results confirming this conclusion have been published by various workers but appear to have all been based on work in which a Bucky grid was not employed. The measurements made at Aberdeen indicate that when a Bucky grid is used, and scatter is kept away from the cassette but not from the gonads, this assumption is not necessarily true. The gonad and cassette dose measurements, together with values of gonad dose normalised for a 1 mrem cassette dose, are shown on tables 6.4a and 6.4b for the cases of A-P and P-A radiographs respectively.

6.8 Radiation doses received by nuclear medicine personnel from $^{99m}$Tc

Technetium 99m is by far the most widely used radioisotope for diagnostic purposes, it emits a 140 keV gamma photon, which is ideal for most scanning procedures, and has a radioactive half life of six hours. This short half life minimises patient dose but means that the isotope
has to be produced on site from a precursor supplied as a commercial molybdenum 99 generator column from which pertechnate ions can be eluted and used to label a wide range of radiopharmaceuticals. These procedures lead to doses to personnel at six different stages:

(a) During periodic replacement of the column.
(b) When eluting the column to extract the daughter isotope.
(c) In preparing the labelled pharmaceutical.
(d) Whilst transporting the prepared dose from the 'hot' laboratory to the ward or outpatient treatment area.
(e) When injecting the isotope into the patient.
(f) During the ultimate scanning procedure.

Doses to personnel arising from each of these six procedures were measured at the Aberdeen Royal Infirmary using TLD dosimeters during a typical working period in 1974. The conclusions drawn from these measurements have been reported (20). Previous reports from nuclear medicine centres have tended to suggest that radiologists routinely injecting the radioactive preparations receive the greatest radiation dose. Our measurements showed that doses arising in this way were no higher than those to the hot laboratory technicians and radiopharmacist, thus confirming observations made by Rudavsky (21) who found that both wrist and whole body doses received by radiologists could be held below 100 mrem/month, even if some hundreds of injections were involved.*

At Aberdeen the personnel receiving the greatest exposure were found to be nurses routinely holding patients on the couch during lengthy scanning procedures. It was impossible to achieve adequate separation between the patient and the nurse, and the problem of reducing doses received

* If a syringe shield is not used the beta plus gamma dose to the skin of the fingers holding the syringe becomes the limiting consideration. Our measurements indicate that up to two injections of 10 mCi of "Tc can then be administered daily without exceeding the appropriate maximum permissible dose.

With beta emitters this limitation may be even more stringent.
from this cause proved very intractable. A worthwhile improvement was achieved by providing a floor mounted lead screen to shield the nurses gonads and much of her trunk. The use of such screens is now routine at Aberdeen.

6.9 Measurement of skin doses in dental radiography using TSEE dosimeters

During the early part of 1974 a colleague* and I surveyed and monitored X-ray equipment used by dental surgeons practicing in Aberdeen city and its environs. It became clear during this survey that many dental X-ray units still in regular use are old and do not incorporate the minimum filtration now recommended by the Code of Practice for the Protection of Persons against Ionising Radiations arising from Medical and Dental Use (1.5 mm aluminium equivalent for sets operating at up to 70 kVp). To check the effect of this deficiency a series of TLD skin dose measurements were made on a Rando skull phantom (formed of a silicone rubber moulding over a skull bone complete with a normal complement of teeth) exposed to a normal bite wing radiograph from a Watson Kingsway dental X-ray unit.

From the resulting grid of dose measurements combined with a radiograph made on a large X-ray film to determine the exact field size and width of the penumbra, a set of isodose contours was constructed. These gave a good visual representation of the skin dose received by a patient and showed clearly that the use of the proper filter reduced the skin dose received by a factor of two (with no loss in quality in the resulting radiograph). The two sets of isodose contours resulting from these measurements, together with the other conclusions drawn from the survey, have been reported (22). Subsequent to this work it was decided that it would be of interest to repeat these measurements using thin layer TSEE dosimeters to give a more accurate figure for the dose to the germinal epithelium. A six by six array of discs of lithium fluoride,

* Mr. Brian Heaton
evaporated onto kapton film and inserted into plastic finger badge sachet pockets, was prepared (Figure 6.13) and mounted on the cheek of the phantom using adhesive tape. These were supplemented by a few additional individual dosimeters placed at other points of interest. As dosimeters of closely matched sensitivity could not always be produced they were all individually calibrated.

Standard 0.5 second radiographs were then taken with and without the filter present in the cone of the X-ray set, the array of dosimeters being read out after each exposure. (0.5 second is the typical exposure required to give a good bite wing radiograph of an adult mouth on a fast dental X-ray film).

The results of these measurements are shown on figure 6.14 and can be compared with the previously published TLD measurements. As would be expected the doses recorded on the thin layer TSEE dosimeters are higher but the difference is only significant in the case of the un-filtered beam where there is a higher preponderance of very soft radiation. These observations suggest that the factor by which a filter reduces the skin dose is nearer to 2.5 than to the 2.0 observed in the earlier experiment. They clearly demonstrate the value of being able to make a remote measurement of filtration in a dental X-ray set by following some procedure which can if necessary be carried out by post. Such a method was therefore developed and is described in the next section.

6.10 Measurement of the effective quantum energy of X-ray beams

The effective quantum energy* of an X-ray beam depends upon both the operating potential and the filtration. If one of these is kept constant a measurement of effective energy enables the other to be evaluated. The method described was developed as part of the survey reported in section 6.9 to enable remote measurements of filtration to

* Defined as the quantum energy of a monochromatic beam having the same half value layer.
be made by post for dental X-ray units which normally operate at about 60 kVp. It is in practice the converse of the method for measuring voltage described by Gorbics and Attix (23).

This method is based on the difference in the energy response of CaF₂ and LiF TLD dosimeters which was discussed in chapter 1. The higher atomic number of calcium means that the absorption cross-section of CaF₂ rises rapidly with decreasing quantum energy whereas that of LiF remains almost constant. It follows that, for equal exposures, the ratio of the light emission from LiF and CaF₂ TLD dosimeters decreases in proportion to the change in absorption cross-section for the CaF₂. A calibration graph can be constructed in which this ratio is plotted against the peak voltage applied to properly filtered X-ray generators, if filtration is normal an alternative scale can show the corresponding half value thickness of aluminium (measured with a step wedge and photographic film). Absence of a filter results in a very low value of both the first half value thickness and the LiF/CaF₂ ratio. The measurements shown in figure 6.15 were recorded over a period of time as opportunity offered, using several different diagnostic X-ray sets. This ensured that if a reasonably consistent calibration was obtained it would be applicable to almost any diagnostic X-ray set - not just to one set which might have some abnormal characteristic. It will be seen that in practice only one of the sets tested gave a reading which, when plotted, lay right away from the mean trend line through all the other readings and which, therefore, indicated either an unfiltered generator or one with a 'soft' tube. It can be reasonably assumed that this calibration graph would apply with fair accuracy to the vast majority of such X-ray units.

The readings shown were obtained with small polythene sachets made up to each contain three LiF-teflon and three CaF₂-teflon dosimeter discs, the measured ratio being that of the average response of each set of three discs. To ensure adequate 'build up' of the dose from the X-ray units
the dosimeter sachets were covered with a 2 mm thick perspex sheet before an exposure was made.

In practice the difference in response with these discs is 'diluted' by the teflon matrix surrounding the TL sensitive grains and further readings have shown that greater discrimination could be obtained by using sintered pellet dosimeters of Li$_2$B$_4$O$_7$ (uniform response) and CaF$_2$ (energy sensitive) although a full calibration curve has not yet been prepared for these.

With the teflon disc dosimeters typical dental X-ray sets were found to have a LiF/ CaF$_2$ response ratio $> 1.5$ when properly filtered. An unfiltered unit had a corresponding value of about 1.2 or less (depending upon the inherent filtration of the tube window). Thus it was relatively easy to detect the absence of the proper filter purely from readout of the exposed dosimeters.

6.11 Other applications and general conclusions

The preceding reports have been chosen to illustrate the types of medical problems in which the use of TLD or TSEE dosimetry can be very helpful, but do not attempt to form a comprehensive survey. Further examples of measurements of interest which have been made include surveys of the radiation dose distributions round patients with radium or caesium 137 implants, measurements of the dose distribution in soft tissue round an Yttrium 90 rod source of the type used for pituitary implants, area monitoring round an E.M.I. tomographic brain scanning machine and round one of its radioisotope equivalents, measurements of the radiation doses received by both patients and radiologists during angiographic procedures, dose measurements associated with partial body in-vivo neutron activation analysis, etc. Some of these measurements have been reported elsewhere (24), (25). Only a few years ago it would have been necessary for them all to have been made by other methods, for example, by the use of Baldwin-Farmer type condenser ionisation chambers or film dosimetry. Such alternative methods would tend
to give inadequate spatial resolution of the dose distribution and/or uncertain energy response. The former point is clearly shown by comparing the lithium tetra-borate TLD dosimeter discs intended to be used by the N.R.P.B. later this year in a new survey of patient doses from diagnostic radiological procedures, with the 35 cc ionisation chamber widely used for the same purpose in the survey carried out in 1957-58 for the committee under the chairmanship of Lord Adrian.

There can therefore be no doubt that the rapid improvements in solid state dosimetry techniques discussed in this thesis are making a very large contribution to the improvement of medical radiation techniques and the control of doses received by both patients and hospital staff.
CHAPTER 6 : REFERENCES


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**TABLE 6.1** BACKGROUND RADIOACTIVITY MEASUREMENTS IN BUILDINGS

<table>
<thead>
<tr>
<th>Building</th>
<th>Gamma dose-corrected for mean outside background. Rad/yr</th>
<th>Radon level</th>
</tr>
</thead>
<tbody>
<tr>
<td>Marischal College - granite</td>
<td>0.153</td>
<td>0.25 pCi/l*</td>
</tr>
<tr>
<td>Medical School - concrete</td>
<td>0.073</td>
<td>0.18 pCi/l</td>
</tr>
<tr>
<td>A.R.I. - granite</td>
<td>0.143</td>
<td>0.54 pCi/l</td>
</tr>
<tr>
<td>Aberdeen house - granite</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(Living room)</td>
<td>0.133</td>
<td>1.5 pCi/l</td>
</tr>
<tr>
<td>(Basement)</td>
<td>0.147</td>
<td>2.9 pCi/l</td>
</tr>
<tr>
<td>Aberdeen house (modern)</td>
<td>0.077</td>
<td>0.24 pCi/l*</td>
</tr>
<tr>
<td>Timber &amp; plaster cottage - Essex</td>
<td>0.042</td>
<td>-</td>
</tr>
<tr>
<td>Brick house - Essex</td>
<td>0.061</td>
<td>-</td>
</tr>
</tbody>
</table>

† Measured over periods of between 6 and 12 weeks

* Measurements made by Dr. K. Cliff of the N.R.P.B. with a portable radon monitor brought to Aberdeen in 1976.

**TABLE 6.2** DOSE RATE FROM $^{90}$Sr-$^{90}$Y 10 mCi Beta SOURCE USING IRRADIATION JIG SHOWN ON FIGURE 6.3

<table>
<thead>
<tr>
<th>Depth (cm)</th>
<th>Dose rate (rad/sec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 cm</td>
<td>1.040</td>
</tr>
<tr>
<td>5 cm</td>
<td>0.0557</td>
</tr>
<tr>
<td>10 cm</td>
<td>0.0216</td>
</tr>
</tbody>
</table>

**TABLE 6.3** RADIATION DOSES RECEIVED DURING MAMMOGRAPHIC EXAMINATIONS

Dose for each X-ray exposure—measured with evaporated layer TSEE dosimeters using a perspex breast phantom.

<table>
<thead>
<tr>
<th>X-ray exposure</th>
<th>35 kVp</th>
<th>42 kVp</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Skin dose</td>
<td>Depth dose at 1 cm</td>
</tr>
<tr>
<td></td>
<td>(Under Mix-D wax)</td>
<td>(Under Mix-D wax)</td>
</tr>
<tr>
<td>400 mAsec</td>
<td>2.6 rad</td>
<td>0.7 rad</td>
</tr>
<tr>
<td>800 mAsec</td>
<td>4.9 rad</td>
<td>1.2 rad</td>
</tr>
</tbody>
</table>
### TABLE 6.4a

Radiation dose (mr) from the primary beam and internal scatter in the range 60—130 KVP. The phantom was irradiated in A-P position, FSD = 100cm field size 23 x 18cm and 2mmAl total filtration.

<table>
<thead>
<tr>
<th>KVP</th>
<th>Skin dose</th>
<th>Male gonad dose</th>
<th>Ovary dose</th>
<th>Mean value of Cassette dose</th>
<th>Skin to M.G. conversion factor</th>
<th>Skin to ovary conversion factor</th>
</tr>
</thead>
</table>
|     | 100 MAS   | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cassette dose | 100 MAS | 1mr Cas
FIGURE 6.1 - Experimental arrangements

NOTE: Free air measurements.
Dosimeters not mounted on water phantom.

FIGURE 6.2
Polar diagram of Personal Dosimeter response

$^{90}\text{Sr}$ $\beta$ Radiation  $\rightarrow$ Angle of incidence  $\rightarrow$ $60$ kVp X-Rays

--- Film (Kodak RM - Estar base)
--- TLD (LiF-teflon discs)
--- TSEE (LiF - Evaporated)
FIGURE 6.3
Photograph of beta ray irradiation jig

FIGURE 6.4
Photograph of graphite gamma ray irradiation chamber

FIGURE 6.5
Photograph of beta extrapolation ion chamber

Without front mylar electrode and with retaining ring removed
FIGURE 6.7
Depth dose distribution for $^{90}\text{Sr}$ beta source

Thickness of LiF teflon disc dosimeter.

Depth mg/cm$^2$
Figure 6.3
Patient positions during mammographic examinations.

- Position for supero-inferior view.
- Position for lateral view.

Figure 6.9
Dose build up for $^{60}$Co radiotherapy in soft tissue.

DOSE IN SOFT TISSUE AS A PERCENTAGE OF THE DOSE AT 1 cm DEPTH.

MEASURED FOR SSD 70 cm, AND SQUARE FIELD 15 x 15 cm.
FIGURE 6.10

Central axis depth dose curve for 60Co Radiotherapy Unit.
70cm SSD. Field size 4cm x 4cm.

FIGURE 6.11

Central axis depth dose curve for 60Co Radiotherapy Unit.
70cm SSD. Field size 15cm x 15cm
FIGURE 6.12
Photographs of phantoms used for dose measurements reported

Breast Phantom. Pelvic Phantom under construction

FIGURE 6.13
Photograph of dosimeter disc array used for isodose contours
FIGURE 6.14
Skin doses in dental radiography
Isodose contours in millirem

Without filter

With filter

FIGURE 6.15
Measurement of effective energy of X-ray beams
7.1 Trapping statistics for lithium fluoride

Apart from the application considered in chapter 5 combined studies of TL, TSEE and TSC properties of phosphors can pose useful tests for existing models of the trapping process. Despite intensive studies of trapping in lithium fluoride a full understanding of the nature of the lattice defects which form the electron and hole traps responsible for TL, as well as those which contribute to luminescent and non-luminescent recombination processes, has only recently begun to evolve and a great deal of further work still remains to be done.

In this section the relevance of the work already reported to presently accepted models for the trapping process in lithium fluoride is reviewed briefly. Lucke has shown that with Harshaw LiF type TLD 100 the intrinsic efficiency for the conversion of incident radiation energy into light is 0.039% (1) and Ellis has calculated that 42,000eV of radiation energy must be absorbed for each photo-electron released from the PM tube cathode (2). These figures are clearly compatible; 0.039% of 42,000eV is 16eV which is equivalent to about five blue light photons of average energy 3eV. A photo-cathode with an efficiency of about 20% could therefore be expected to emit one photo-electron.

The band gap in lithium fluoride is 14eV and typically with both semi-conductors and gases, the energy required to create an ion pair (w) is about twice the band gap. Assuming w to be 28eV about 1,500 electrons would be lifted into the conduction band by 42,000eV of absorbed radiation energy. Assume that the fraction which are trapped (trapping factor) is t and the fraction of trapped electrons which undergo luminescent recombination when thermally released (recombination factor) is r. Then as shown above, 1,500 t_r corresponds to about 5 photons so that
\[ t_r = \frac{1}{300} \] (eqn. A) and either t or r can be calculated if the value of the other is known. In principle
it would appear that $t_0$ could be estimated by irradiating a sample of lithium fluoride in the readout chamber of a TLD reader and comparing the light emitted during irradiation by luminescent recombination of electrons which were not trapped, with that subsequently emitted during normal thermal readout. The author has seen no reports of such an experiment being attempted and suggests it could represent a worthwhile piece of supplementary work, particularly if carried out with both undosed and predosed phosphors in order to demonstrate any change in the value of $t_0$ following irradiation. The result would however, be equivocal, the ratio of the cross-sections for luminescent and non-luminescent recombination is almost certainly temperature dependant. An alternative method for obtaining an order of magnitude estimate of the trapping factor $t_0$ is therefore outlined below:

Mayhugh and Rossiter have independently demonstrated that the luminescent centres in LiF are associated with titanium atoms and that the optimum concentration of titanium for peak light emission is 8 ppm. (3), (4), (5). Similarly the basic electron traps are associated with the magnesium impurity which has an optimum concentration of 80 ppm (6). With complete dispersion of the magnesium atoms (this is generally regarded as unlikely; a proportion of the magnesium is thought to take the form of precipitated MgF$_2$) this could lead to the creation of $2 \times 10^{18}$ divalent electron traps per gram of lithium fluoride. However Dryden and Shuter have shown that the principal traps associated with the main thermoluminescent peak of lithium fluoride appear to be characterised by Mg trimers formed in the 111 plane by a ring of three of the dipoles (7). The maximum number of such traps will therefore be about $6 \times 10^{17}$ per gm for this magnesium concentration. It was shown in chapter 5 that supralinearity of LiF is associated with an increased probability of trapping so that with increasing dose the value of $t$ rises from that of $t_0$ in equation A. The variation in phosphor sensitivity with predose should on this hypothesis also represent the corresponding variations in the trapping factor $t$. Saturation
can be assumed to be beginning to set in when the sensitivity has fallen from its peak value to the initial value in the undosed condition and it can reasonably be assumed that the response will begin to saturate when about half the available trapping sites ($\sim 3 \times 10^{17}$ per gm) are filled. From figure 7.1 it would appear that this corresponds to a total dose of the order of $3 \times 10^{5}$ rad. Each rad corresponds to an energy absorption of $6 \times 10^{13}$ eV per gm so that, for a $w$ value of 28 eV, $2 \times 10^{12}$ electrons should be raised to the conduction band per gm rad. For a total dose of $3 \times 10^{5}$ rad there should therefore be $6 \times 10^{17}$ electrons per gm available in the conduction band for trapping. The mean trapping factor $t_m$ over this range is therefore $3 \times 10^{17}$ or 0.5. Over this range of doses reference to figure 5.8 in chapter 5 indicates that the mean trapping factor $t_m$ will be about 1.5 times $t_0$, the initial value in the undosed condition. Putting $t_0$ equal to 0.33 in equation A yields a corresponding value of the recombination factor $r$ of 1% which agrees exactly with the estimate made by Zimmerman and Cameron in 1968 (8).

The analysis above has assumed that the relevant traps are all associated with Mg trimers and it is not immediately obvious how this can be reconciled with the work described in chapter 5 which demonstrated that supralinearity arises from the creation or activation of additional electron traps during irradiation. A possible answer is that irradiation leads to an enhanced vacancy density in the crystal and only when there are sufficient lattice vacancies can the Mg atoms completely disperse and group to form the trimers which constitute effective traps. Such a model provides a simple explanation for the loss of enhanced sensitivity by a predosed phosphor when subjected to a high temperature anneal. It also explains why the response of LiF to high LET radiation shows very much less supralinearity, since the lattice defects produced by the high LET radiation will tend to be concentrated along particle tracks rather than dispersed and therefore may well be
geometrically separated from local clusters of Mg atoms in the form of precipitated MgF$_2$.

This brief survey demonstrates that the physical model of trapping in lithium fluoride proposed above is not contradicted by any of the experimental work reported elsewhere in the thesis. The extent to which it remains applicable in the case of the surface layers (where the TL sensitivity has been shown to be relatively low (9)) can be assessed by considering the TSEE data in a similar way.

Figures for thermally stimulated electron emission from luminescent grade lithium fluoride show typical values of about 4000 exo-electrons per rad from 10 mg samples and values of the same order are obtained from chemically 'pure' lithium fluoride. Assuming a particle size of about 100 µm such samples would contain about $10^4$ grains of LiF with a total surface area of about 3 cm$^2$. Experiments with varying sample sizes suggest that except for highly dispersed samples only part of this area (perhaps ~10%) can act effectively in the emission process (see figure 7.2). This is probably due to the electrostatic space charge effect adjacent to the surface of the grains. If a correction is allowed for this effect one can estimate the maximum total emission per cm$^2$ as about 10,000 electrons per rad. The total number of trapped electrons which would be expected to be released from traps within a 10 mg sample has been shown to be about $2 \times 10^{10}$ per rad i.e. about one electron in a million actually escapes through the crystal surface after detrapping.

Holzapfel (10) has demonstrated that for ionic crystals exo-electrons are all released from surface layers (within a depth of 1 - 10 nm) and that the contribution of electrons diffusing from greater depths as postulated by Bohun and Dolejsi (11) is negligible. Considering first the smaller of these values it is clear that the probability of emission must always be low however near to the surface the electron may be when released from its trap. A 10 mg sample has a volume of
about $4 \times 10^{-3}$ cm$^3$; for the 100 μm grains under consideration the total volume of such a sample within 1 nm of the surface of a grain will be $\sim 3 \times 10^{-7}$ cm$^3$, so even for this thickness layer the number of electrons emitted will only be of the order of 1% of those released in the layer. If a thicker layer is considered the fraction emitted will be still smaller. Three basic mechanisms of exo-electron emission have been postulated. Emission has been attributed to the high energy tail associated with a Maxwellian energy distribution, for which the electrons are capable of overcoming the work function of the surface (12); to chemical reactions on the surface which lead to a transfer of energy from excited chemical bonds associated with adsorbed impurities to conduction electrons (13); or finally to the presence of local surface defects leading to internal electric fields which provide 'emission centres' from which low energy electrons can escape.

The first of these hypotheses cannot readily account for the sharp peaks which can be observed in the energy spectrum of the emitted exo-electrons (14) whilst the second conflicts with observations by Samuelsson who showed that surface impurities do not substantially influence the exo-electron emitting characteristics of lithium fluoride (15). The third appears to be applicable in the important case of beryllium oxide dosimeters where the crystalline structure leads to the establishment of strong pyroelectric fields (16), (17), (18). Nevertheless there is no general evidence in support of a theory of field assisted emission in the case of alkali halides (19) and for such samples none of these theories can yet be ruled out as there is unfortunately no easy experimental way of conclusively distinguishing between them. Assuming that emission from alkali halides is predominantly a thermal process Holzapfel has analysed the effect of surface work function on exo-electron thermograms and shown that it must lead to an increase in the observed temperatures of the emission peaks (20). Although the exo-electron and TL thermograms of many materials (including lithium fluoride) are very
similar, there are usually differences (21) which could be associated either with this effect or with modifications of the trap depth in the surface layers produced by slight differences in the lattice structure. The second theory does not appear to be applicable in the case of LiF and the extent to which the third theory can explain the observed emission involves detailed knowledge of the lattice structure of the critical surface layer, and of the electric field configurations associated with the various defects present. Nevertheless there is increasing evidence of discrete emission centres associated with specific lattice defects or discontinuities rather than of uniform probability of emission over the whole surface (22).

Surface layers must be strongly affected by oxygen and other contaminants as well as being very subject to the effects of abrasion and mechanical damage. It is therefore not surprising if their crystal lattice structure shows defects and activator concentrations different to those in the bulk material. For example with aluminium oxide it has been shown that there is a thin oxygen deficient cubic lattice surface layer superimposed on the hexagonally crystalline bulk material (23). In the light of these considerations it is unrealistic to expect the thermal behaviour of exo-electron dosimeters to correlate closely with that of corresponding TL dosimeters, or to assume that the absence of such correlation implies a totally different mechanism. With many materials, including lithium fluoride, it is remarkable how closely the thermograms of TL and TSEE dosimeters correspond. It is difficult to believe that the primary electron traps are not closely related in such cases.

Although it is felt that the preceding discussion forms a reasonably coherent model for the trapping process in LiF several observed effects not yet adequately explained do still remain, and these indicate valuable lines which can be followed in future work:

(1) There are many types of trap in LiF which normally shows five radiation induced peaks in its glow curve
between ambient and 300°C, and can exhibit two more-one following neutron irradiation and the other following post-annealing. Three additional peaks stimulated by tribo-luminescent effects and apparently associated with surface states, may also be observed.

(2) Evaporation of samples of thermoluminescent LiF leads to a complete loss of TL efficiency although the TSEE efficiency remains. This can be attributed to the titanium luminescence centres not being carried over during the evaporation process but other factors must also be involved as the TSEE curve structure is simplified after evaporation, showing a main peak only. On the model discussed above this would imply that the magnesium evaporates with the lithium fluoride although the titanium does not, and that some simplification of the magnesium traps ensues.

(3) The physical mechanism involved in the exo-electron emission process is still not understood and as with supra-linearity it is probably unwise to assume that it is the same for all types of sample. Apart from the three detailed models discussed on p.210 possible explanations of exo-electron emission include a form of internal conversion process analogous to Auger electron emission but associated with non-luminescent electron hole recombination, and a surface photo-electric effect arising from photon absorption at some appropriate surface emission centre.

(4) The importance of thermal annealing in the restructuring of glow curves and simplification of trapping levels indicates that many of the subsidiary levels (and the corresponding peaks) may be due to lattice defects or discontinuities. It can reasonably be postulated that where such a defect or discontinuity occurs close enough to an Mg F-centre trap for the wave function of an electron in the trap to interact with the discontinuity, the associated trap depth will be modified. Only a limited number of discontinuity configurations will be geometrically possible and some of these will lead to the trap becoming unstable. It therefore appears possible that any specific trapping centre can be associated with several discrete values of trap depth according to the configuration of the surrounding crystal lattice.
7.2 Summary and recommendations for further work

Chapters 2 - 6 of this thesis have all reported research into some aspects of the general problem of solid state radiation dosimetry using thermally stimulated read-out, and most of this work has been open ended with possible lines for further study indicated.

Chapter 2 is largely an account of routine development work. But comprehensive studies of the problems encountered while operating small scale routine personnel monitoring services with LiF loaded teflon discs dosimeters do not appear to have been previously published. Many of the early users of TL dosimeters have become so discouraged by these problems that they have reverted to other methods until the manufacturers of commercial equipment can produce improved readers and dosimeter elements. An account of one system in which these problems have been largely overcome should therefore be of value to other workers contemplating the introduction of regular dosimetry by this method.

Chapter 3 comprises a corresponding account of the problems encountered in developing a reliable reader for TSEE dosimetry. This is a more recent method and so far few practical applications have been evaluated fully. It appears that TSEE could be used to provide a personnel monitoring service which would be (a) cheaper and (b) more sensitive than TLD. The remaining problems are primarily related to achieving the absolute reliability necessary where the loss of even a single reading is unacceptable. Using the reader design described in chapter 3, counter instability was reduced to the point where it was very exceptional for any reading to be affected. However occasional counter breakdown could not be entirely excluded and tended to occur more often towards the end of a period of time (usually of the order of a month) during which the build up of particulate matter in the counter had made dismantling and thorough cleaning imperative. After such an operation sensitivity seldom returned exactly to its former value and the plotting of a new counter plateau
became desirable. Thus the TSEE dosimeter described in this chapter could be readily used for applications (such as those reported in chapter 6) where a series of related readings were expected and failure to obtain this gave prompt warning of a marked change in reader characteristics; but could not be so readily trusted if successive readings were (as in personnel monitoring) totally unrelated.

Although this limitation is still an important problem the very thin sensitive layer of an exo-electron dosimeter provides a valuable compensating advantage for many applications, and if dosimeters are produced by the method of evaporating thin layers under vacuum as described in chapter 3, the full benefit of this property can be obtained. Lithium fluoride was used as the sensitive material for most of the work with evaporated dosimeters actually described but it would be possible to use the technique with almost any other substance which proves to have high and stable exo-electron emission properties. For such very thin layers the question of the tissue equivalence of the dosimeters is governed almost entirely by the properties of the backing film and packaging - there is therefore much greater freedom in the choice of the actual dosimetric material. This method of dosimeter production also minimises electrostatic effects during readout in a more elegant way than by the alternative of trying to produce conducting ceramic discs (24).

An important future line of work will therefore be to search for stable high sensitivity exo-electron emitters suitable for evaporation under vacuum but unrestricted by consideration of their effective atomic number. Other suggestions for further work would include studies of alternative readout procedures (e.g. optically draining the irradiated dosimeter by flooding it with a light beam of gradually increasing quantum energy, or releasing the electrons trapped after irradiation by applying a gradually increasing mechanical stress). Further fundamental and applied research which would be valuable includes for example studies to determine the exact thickness of the
surface layer from which exo-electrons can escape and to relate this to the mean free path of the electrons in the crystal; and work to establish whether a reliable neutron or charged particle personnel monitor with a very low response to background gamma radiation can be developed.

Chapter 4 which discusses TSC, the third of the dosimetry techniques used for the work reported in this thesis, is relatively short. The main aim of the TSC studies carried out was to investigate the effect of large absorbed doses on the TSC sensitivity of samples, and was planned as part of the investigations made into the physical basis for supralinearity effects. The results obtained during this work, which was discontinued relatively early, gave no promise of providing a good alternative method for routine dosimetry but the final part of the chapter surveys later and more promising observations carried out subsequently in other centres where further work in this field is being actively pursued.

Chapter 5 comprises the study of supralinearity involving observations not only of TSC but also of TL and TSEE sensitivities, and this probably forms the principal theoretical contribution made in the thesis. The method appears to be both convenient and powerful; further work using other sample materials could be readily carried out on this basis.

Chapter 6 consists of accounts of a number of separate research projects in which TL or TSEE dosimetric measurements were employed. They help emphasise the versatility of both these important dosimetric techniques. Other projects of a similar nature are already in hand.

The relative priorities which should be allocated to the further work suggested must depend upon the particular interests of the research school concerned. However extensive commercial exploitation of thermoluminescence has been under way for a considerable time now and is likely to continue. Similarly commercial beryllium oxide ceramic disc TSEE dosimeters with a claimed response of \( \pm 6\% \) over the range from \( 10^{-6} \) rad to \( 10^{2} \) rad (eight decades) have recently
been marketed.* The author feels that at this stage commercial interests will probably exploit practical dosimetric applications of the TSEE technique more effectively than research institutes, and that the latter should now concentrate on increasing the basic understanding of the properties of thin layer dosimeters in terms of fundamental solid state physics. Such work would probably be most effectively conducted by both preparing and reading out the dosimeters under vacuum using a channel multiplier as the exo-electron detector.

7.3 Future outlook

Solid state dosimetry is still in its infancy. A bewildering variety of systems are theoretically capable of development. Some, such as thermally stimulated conductivity and lyoluminescence (25), (luminescence released when irradiated solids pass into solution), have been exploited rapidly and within the last few years have passed from insensitive techniques of purely theoretical interest to serious competitors with the established thermoluminescence systems. Others will undoubtedly be exploited in a similar way within the next few years. It nevertheless seems likely that with the present development effort being expended in the field of thermoluminescence other systems will be unable to supersede TLD for general use in the near future, although TSEE may become more widely used for those applications where TLD is either unsuitable or not readily employed. Intensive studies of the properties of phosphors and of their explanation in terms of solid state physics will also undoubtedly continue. For this purpose there will be a continuing need for investigations into the TL, TSEE and TSC properties of the various phosphors in conjunction with the use of all other modern techniques of solid state research.

* Novex Co. Ltd., 1056 Budapest, Marcius 15, ter I.IV, Hungary
CHAPTER 7 : REFERENCES


7.22 BECKER, K. 'Solid State Dosimetry' C.R.C. Press 1973 p.113


FIGURE 7.1
Supralinearity in LiF

FIGURE 7.2
Variation of TSEE response with sample size

Calcium Fluoride:

Photograph of samples above.
Without the help and encouragement of many individuals in many organisations the work reported in this thesis would not have been carried out in its present form. Some of these have already been named in the preface to the thesis, many are not named but have contributed ideas or references during discussions at conferences and seminars. Others who work in the North East London Polytechnic Science Faculty workshop and the University of Aberdeen Medical Physics Department workshop, also played an essential part by helping to provide the necessary hardware. My immediate colleague Mr Brian Heaton co-operated enthusiastically with those parts of the work reported which related to our immediate responsibilities in the University Radiation Protection Service, and some of the experimental arrangements I used for the work reported in chapter 6 were prepared and tested by students as projects which formed part of their University training. Such help has been acknowledged where appropriate in the main text but grateful thanks are expressed to all the individuals who have helped in one or more of these ways.

Apart from this, particular thanks are due to my supervisor Dr W.B.Gilboy, not only for his constant help and detailed criticisms throughout, and particularly during the final preparation of the manuscript; but also for his understanding and forbearance at times when circumstances made progress difficult or impossible. I must also pay tribute to my wife for her tolerance and encouragement at times when the work interfered with my capacity to carry out normal domestic responsibilities and to my typist Mrs P Watson who typed it so carefully and clearly.