RAPPORTEUR PAPER
THERMOLUMINESCENT DOSIMETRY

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INTRODUCTION

Although several groups of workers were interested in the phenomenon of thermoluminescence well before the last war, it is only in recent years that suitable techniques have been developed for applying the thermally released luminescence in suitable crystalline materials to the dosimetry of the exciting radiation. The great interest in this subject of thermoluminescent dosimetry (abbreviated to TLD) was demonstrated conclusively last year at the First International Conference on Luminescence Dosimetry held at Stanford University when 260 people from 17 countries discussed the subject exhaustively for three days. That Conference was stimulating not only for the high quality of the papers presented but also because some attempts were made to explore possibilities for the future development of the subject. One of the main points highlighted in the final discussion was the need for further work to illuminate our understanding of the thermoluminescent mechanism and the parameters on which it depends. A second requirement was for further practical experience of the method in order that TLD could take its rightful place in the armoury of those concerned with dosimetry, including the health physicists.

The six papers which it is my pleasure to review this morning are concerned with one or other of these aspects of the subject. I will start by considering experiments carried out to investigate the mechanism of the TL phenomenon.

REPORT

Dr. Ehrlich from the National Bureau of Standards, Washington, has studied the shape of the response curve of lithium fluoride as a function of radiation *exposure*, *exposure rate* and *photon energy*. ^{60}Co gamma radiation was used at six different exposure rates in the range 100 to 7 million R/hr, as well as X-radiation of half value layer about 5 mm Al at an exposure rate of 7000 R/hr. The range of total exposure studied was 1000 to 20 million R, i.e. that part of the response–exposure curve showing "superlinearity" and also that region of the curve in which the response passes through a maximum. Most of the work was done with Harshaw TLD-100 LiF powder, annealed for 15 min at 400°C prior to exposure and heated for 15 min at 100°C after exposure in order to ensure emptying of shallow traps. The powder was exposed in 1 mm thick polyethylene vials thickened with plastic sleeves to ensure electronic equilibrium when irradiated with ^{60}Co gamma rays. Enough powder was used to allow two read-out values which agreed within 3%.

The results of the irradiations with ^{60}Co gamma-rays at the three highest exposure rates are shown in Ehrlich's Fig. 1. Typical glow curves which are not significantly rate dependent, are shown for three values of exposure. Within the limits of the experimental accuracy (about $\pm$ 15%) no change in the shape of the response curve with exposure rate was found over the whole range of exposure rate or exposures studied. This confirms the work of others, notably Karzmark *et al.*, and Ehrlich suggests that the thermoluminescence of TLD-100 is associated at least mainly with centres other than F-centres, the formation of which are known to be rate dependent.

Ehrlich's Fig. 2 shows the response curves obtained for the two different photon energies. The gamma-ray curve also supplements the previous diagram in showing the negligible effect of exposure rate for the three lower rates. It will be seen that for gamma-rays superlinearity sets in at around 350 rad whereas for soft X-rays not until 2000 rad. The slope of the superlinearity region is steeper for gamma-rays than for X-rays although the curves have the same slope above about 20,000 rad. These results confirm those obtained by Naylor for lower exposures but do not support the findings of Morehead and Daniels in which considerable difference in curve shape with radiation energy was found at high level exposures. However the importance of the present work is that the possible effect of exposure rate as a parameter has been eliminated, the difference in slope being shown to be due solely to photon energy.

Dr. Ehrlich concludes that the decrease in the slope of the response curve with photon energy supports Cameron's theory that superlinearity is due to the creation of additional electron traps by the radiation.

Turning to new applications in the TLD field, Monsieur Van Espen in his paper, discusses improved systems of operation of the calcium fluoride TL dosemeters manufactured by the Belgian firm of M.B.L.E. He points out that erasing the dose effect in the read-out process is a disadvantage of TLD in that it is not possible to recheck a suspect dose-reading or to measure the integrated value of a series of doses if the latter have been read out previously.

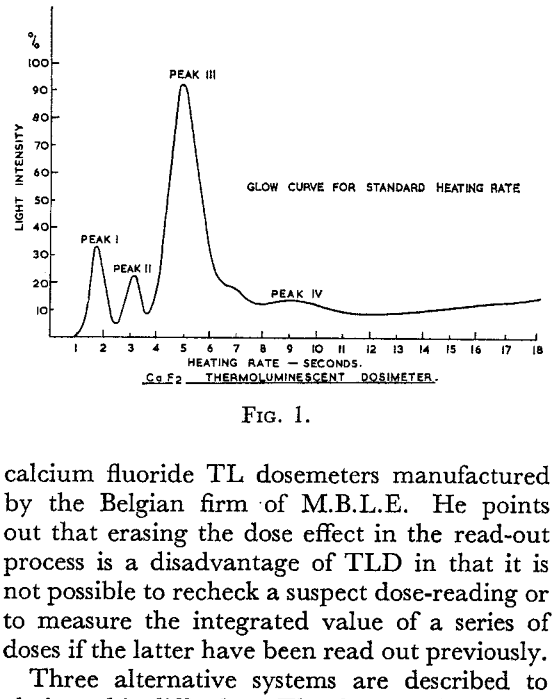

Fig. 1.

Three alternative systems are described to obviate this difficulty. The first consists essentially of two dosemeters in the same glass envelope; by means of twin heating cathodes it is possible to read out doses successively from the two dosemeters, or alternatively to read out individual doses by heating one cathode and to use the other dosemeter as a dose integrating device. These dosemeters are sensitive to doses as low as 100 mR.

The second system depends on a single dosemeter which may be read at two different read-out temperatures. Fig. 1 of this report shows a typical glow curve of M.B.L.E. calcium fluoride. Individual doses are read out by emptying trap II of the glow curve which occurs at about 180°C. Check doses or integrated dose values may be obtained by reading out at a temperature of 375°C which empties traps III′, (and III) of the glow curve. The sensitivity of this system allows integrated doses in the range 1–1000 R to be measured.

The third method depends on the use of UV light to transfer a fraction of the energy accumulated in deep traps into shallower traps which are emptied by read-out at normal temperatures. In M.B.L.E. calcium fluoride the main trap (peak III) shown in Fig. 1 is normally emptied at a temperature of 265°C. Electrons trapped at levels corresponding to peak V in the glow curve, which occurs at about 500°C (not shown in the diagram) are not significantly released at 265°C. Hence in a series of irradiations the energy stored in trap V may be used as a measure of the integrated dose level. It is impracticable to tap this energy directly by reading out at 500°C but if the dosemeter is exposed to UV light of wavelength about 3300 Å for 5 min under standard conditions and is then read out at 265°C, about 0.25% of the total dose received by the dosemeter is recorded with an accuracy of 15% or better. The transfer procedure can be repeated a number of times, the results being reproducible within a few per cent, but at present the method is restricted to total doses greater than 20 R.

The measurement of finger and hand doses incurred when radioactive material is handled has always given rise to difficulty particularly when the dose gradient is steep such that ring or wrist dosemeters are of limited value. Drs. Bjarngard and Jones of Controls for Radiation Inc. (Conrad, for short) describe experience with their firm's TL dosemeters consisting of Teflon discs incorporating LiF, which were introduced over a year ago.

The dosemeters, shown in Bjarngard's Fig. 3, are 12.5 mm diameter and 90 mg/cm^2 thick and 28 mg of lithium fluoride is incorporated uniformly throughout the disc. The lightproof polyethylene pouch is 7 mg/cm² thick. Read-out is conventional and in nitrogen but annealing at 300°C is necessary rather than 400°C since Teflon softens at about 320°C. The response of the disc is a measure of the energy absorbed in it, that is, at effective depths between 7 and 97 mg/cm². Although the biologically important dose is usually taken to be at 7 mg/cm² depth, for the palmar surfaces of the hand and fingers this is usually a gross underestimate, and the average dose to the basal layer is effectively measured by the disc. However, for soft beta radiation a small correction may have to be applied.

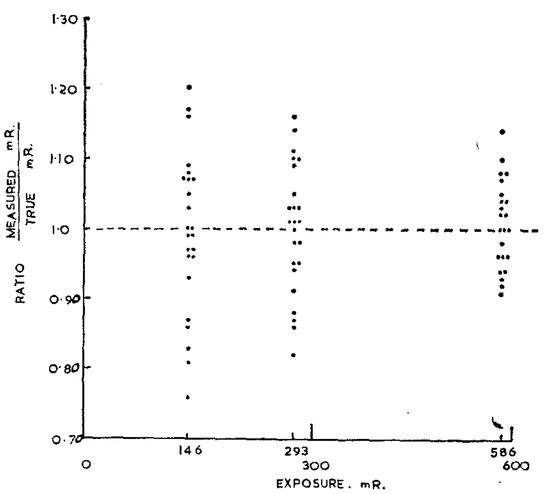

FIG. 2.

Bjarngard's Fig. 4 shows the precision attainable with known doses of ^{60}Co gamma radiation down to 10 mR. At this dose level the standard deviation from the mean of 20 determinations is 20% but decreases to 3% for doses above 3R. It is interesting to compare Bjarngard's results with those obtained by Burton, Foster and Townsend from the U.K. Central Electricity Generating Board. Following similar experiments with Conrad Teflon-LiF discs, these authors found poorer reproducibility, as shown in Fig. 2 of this report, unless the discs were recalibrated after each exposure when similar results to those of Bjarngard and Jones were obtained. However, the British authors underline the need to avoid measurement of the low temperature peak which, on account of its short half-life, can give rise to poor reproducibility of results unless a rigid time schedule is adopted between irradiation and read-out, or the low temperature peak is reduced to insignificance by post-irradiation annealing at say 80°C for several minutes or by delaying 24 hr or so before read-out. Glow curves taken before and after the partial elimination of this peak are shown in Burton's Figs. 4 and 5.

Bjarngard and Jones point out that the background reading associated with unirradiated discs is 1.5 times the net TL signal associated with a dose of 20 mR. This background is made up of the photomultiplier dark current and a component due to what the authors term "spurious thermoluminescence". It has been shown that the latter is not significantly affected by friction or mechanical shocks applied by shaking the discs. The possible effect of visible or ultra-violet light has been investigated and the latter, present in fluorescent laboratory lighting, has been shown to produce definite TL response, as shown in Bjarngard's Fig. 6. It would be interesting to know whether these experiments were undertaken on freshly manufactured discs having no ionising radiation history or whether they had merely been annealed at 300°C following previous irradiation. In the latter case the so-called light excitation may really represent the transfer to shallow traps of stored energy from previous ionising irradiation present in deep traps not emptied in the annealing process, much as in the method employed by Monsieur Van Espen described earlier. However, light excitation is presumably not the cause of the spurious thermoluminescence in the present situation since the discs are used in light-proof pouches. The authors suggest that unirradiated dosemeters should be included for background determination in any measurement series.

There is no doubt that devices of this type have an important place in health physics practice. The next two diagrams show the relative doses received by the hand and fingers in handling a radium needle (Bjarngard, Fig. 1) and a uranium plate (Bjarngard, Fig. 2) and demonstrate the facility with which the system may be used. However, the relatively high cost of these components—25 shillings each ($ 3.50)

—is a limiting factor to their more widespread use—at any rate in the U.K. at present.

Burton, Foster and Townsend have also measured finger-tip doses incurred in the handling of radioactive materials but, instead of Teflon discs, have used LiF powder in PVC sachets. At the same time the total doses to the wrist were measured with wrist film badges, the contributions from gamma, X- and beta radiation being summed. The ratio of the finger-tip dose to wrist dose was found to average about 7 with a maximum value of 22. The types of radiation source are not defined but the large value of the dose ratio clearly arises from the inhomogeneity of the irradiation field.

The remaining papers in this rapporteur session deal with comparative studies of TLD and film badges. Drs. Johnson and Attix of the Naval Research Laboratory, Washington, describe an intercomparison experiment in which 100 NRL personnel wore one or two TL dosemeters in addition to a simple film badge for four one-month collection periods. Quartz fibre pocket dosemeters, specially selected for low electrical insulation leakage, were also worn by some of these staff.

The dosemeters used are shown in Johnson's Fig. 1. The two TL dosemeters, a U.S. Navy experimental prototype (DT-284) and a commercial type designated M, use activated calcium fluoride as phosphor and both incorporate a metal shield which flattens the response per röntgen above 80 keV. The response below 80 keV rapidly falls to insignificance. The film holder is made of stainless steel with a single filter of 1 mm Cd as developed at Oak Ridge. Dual emulsion film packets were used in the holder, the processing and assessment being conventional; the same ^{60}Co source was used to calibrate both film badges and TL dosemeters.

The dosemeters were worn in medium dose-rate areas (although the highest available), only three exposures greater than 100 mR being measured in the monthly periods. Almost all employees wore their film badges on their belts and their TL and quartz fibre dosemeters in their shirt pockets. However, the authors believe that the exposures in the two sites were not significantly different and state that "seldom did any individual wear only part of his complement of dosemeters"! The TL dosemeter readings were corrected for signal fading, shown in Johnson's Fig. 2, and for radiation background, although a large part of the measured background was due to radioactivity present in the dosemeter structures. Johnson's Table 5 summarizes measurements of this radioactivity which contributes some 25 mR/month to the background reading.

Figure 3 of the same paper shows some of the results of the intercomparison in terms of the readings of the DT-284 dosemeter. The points chosen were from among the highest exposures. Considering the low level of the doses being measured, it is surprising that such good agreement was found. In studying this graph it should be borne in mind that the lowest dose measurable with the film badge is 25 mR; smaller doses than this are all recorded as 1 mR on the graph. Further, the authors point out that the largest discrepancies between the film badge results and the TL readings occurred during a failure of the air conditioning system in the NRL Reactor Hall giving rise to temperatures of 27–32°C and relative humidity above 90%; this led the local Health Physicist to disallow the film readings for the purpose of the employees exposure histories. The authors' comment "clearly the film badges were at a disadvantage in these tests", although apparently environmental conditions approaching those described are not atypical of Washington in the summer months. The authors conclude that it is premature to consider replacing film badges by TL dosemeters for personnel monitoring at NRL in the immediate future.

Burton, Foster and Townsend also describe operational trials in which LiF dosemeters are compared with film badges for personal monitoring. These authors used Conrad Type 7 LiF in 45 mg aliquots in PVC sachets; the film badge was the U.K. multi-filter badge. The experiments were performed in two parts. In the first part sachets of LiF powder were *attached to* the film badges which were issued monthly to about 12 members of the staff. In the second part of the experiment the sachets were *not* attached to the film badges, it being left to the operator to wear the sachet as near to the film badge as possible. Burton's Fig. 2 shows the results obtained with the composite dosemeter. Apart from the dose region up to 50 mrad, which is near the limit of sensitivity of both dosemeters, the correlation is reasonably good despite the fact that the operators were working with varying amounts of gamma, X- and beta radiation. The results of the second part of the comparison (Fig. 3 of this report), unlike the results of Johnson and Attix, show very poor correlation, in most cases attributed to the different positions of the dosemeters on the body.

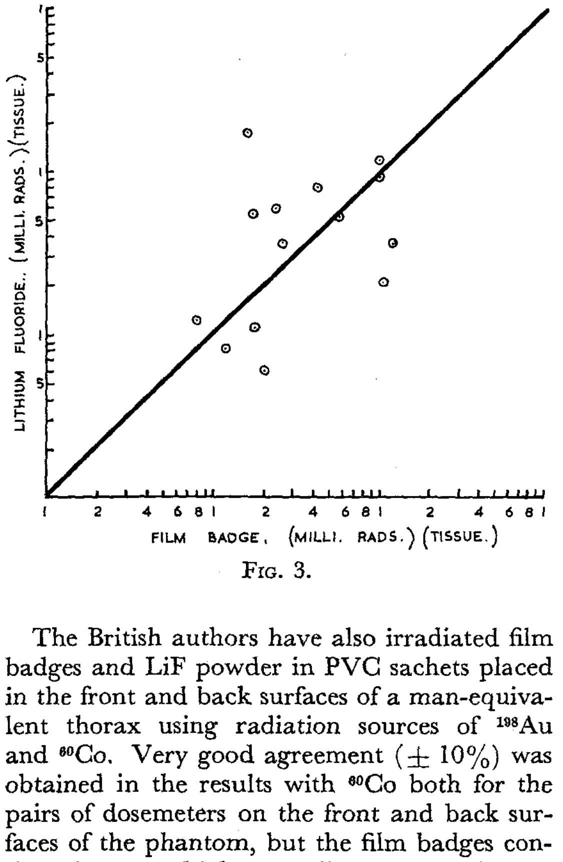

FIG. 3.

The British authors have also irradiated film badges and LiF powder in PVC sachets placed in the front and back surfaces of a man-equivalent thorax using radiation sources of ^{198}Au and ^{60}Co. Very good agreement ($\pm$ 10%) was obtained in the results with ^{60}Co both for the pairs of dosemeters on the front and back surfaces of the phantom, but the film badges consistently gave higher readings amounting to about + 30% with the 0.4 MeV gamma radiation from the ^{198}Au.

The paper by Cameron and Suntharalingam from the University of Wisconsin, describes a laboratory intercomparison experiment using single crystals of LiF (TLD-100) and film badges supplied by commercial companies. The LiF crystals were enclosed in lucite capsules having 4 mm wall thickness and attached to the film holder. The crystals, of mass approximately 20 mg, were cleaved from a single piece of virgin TLD-100 and annealed for 1 hour at 400°C before use in the experiment.

Known exposures of radium and caesium-137 gamma-rays and 140 kVp X-rays, measured with a Victoreen R-meter to an accuracy of $\pm$ 10%, were given to the composite dosemeters. The film badges were returned to the company together with 300 other operationally exposed badges. The company were not informed that a special intercomparison experiment was being undertaken. The LiF crystals were read by the authors using the read-out process described in the literature. In a further similar experiment the irradiations were not made by the authors but by the National Sanitation Foundation Testing Laboratory at Ann Arbor. Twenty composite dosemeters were irradiated with exposures not known to the authors until after they had evaluated the LiF thermoluminescence in their own laboratory. The film badges were evaluated as previously by the commercial company.

Cameron's Fig. 3 shows results of the intercomparison using radium gamma rays. The broken lines represent $\pm$ 20% of the true dose. When mixtures of radium gamma rays and 140 keV X-rays were used the results were still satisfactory (Fig. 4 of same paper) except for dose values of about 20 mR. As the detection threshold for the film badges of one of the companies was stated to be 50 mR, good correlation clearly cannot be expected at these levels. The results of the experiment involving the N.S.F. Testing Laboratory are shown in Cameron's Fig. 7. The LiF results are slightly high and the film readings by and large underestimate the results by a small amount.

The authors claim an overall accuracy of $\pm$ 30% for the single crystal LiF results which can be improved upon if a slight energy correction is applied. The principal disadvantage of these crystals at present is the slow read-out time of 15 min, required because of the need to calibrate each crystal individually.

CONCLUSIONS

In conclusion in view of my earlier restraint perhaps I may use the last minute or two of