Meetings

Luminescence Dosimetry

Although some knowledge of radiation-induced luminescence dates back almost to the discovery of x-rays and radioactivity, only in the last 15 years has there been a substantial growth in the development and application of luminescence phenomena to radiation dosimetry. This growth was reflected in the recent convening of an international conference on luminescence dosimetry, held at Stanford University, 21–23 June.

Luminescence dosimetry depends on the fact that the electrons released by ionizing radiation in certain phosphors may be trapped at defect or activator sites in the crystal lattice resulting in long-lived metastable states. In thermoluminescence dosimetry (TLD), the irradiated phosphor is heated (thus releasing the trapped electrons), and the emitted light which results is measured. In radiophotoluminescence dosimetry (RPLD) the radiation produces stable fluorescence centers which may be repeatedly stimulated by ultraviolet; the emitted visible light may be measured. Luminescence dosimeters, which are now undergoing rapid development, possess the advantages of small size, high sensitivity, good storage properties, a wide dose range (from perhaps 10^{-5} to 10^{6} rad), and often approximate tissue equivalence.

The chairman of the conference was J. H. Schulman (U.S. Naval Research Laboratory), whose group has played a major role in the development of luminescence dosimetry. Schulman summarized the progress up to the present time in both radiophotoluminescence and thermoluminescence dosimetry and outlined the physical fundamentals underlying these systems. With respect to the most important RPLD system, Ag-activated phosphate glass, he noted, on the basis of optical and electron-spin-resonance studies, that the stable centers responsible for the continuous emission of orange light (5000 to 7000 Å) under 3650 Å ultraviolet excitation are now thought to be Ag^{+2} ions or clusters of silver atoms. Schulman pointed out that a great deal of progress in RPLD research has recently been made, notably by R. Yokota in Japan and K. Becker in Germany. The development of new types of more nearly tissue-equivalent glass, improved readers, and optimum size and surface polish of glass pieces, have all contributed to the present feasibility of measuring γ-ray exposures of tens of milliroentgens, some two to three orders of magnitude below the practical limits of a decade ago.

Schulman's review also dealt with the development of TLD systems, especially CaF_2:Mn and LiF. The thermoluminescence process, including glow curves and the Randall-Wilkins model involving the concepts of trap depth and escape frequency, was described. The influence of the heating rate and thermal history upon the glow-curve peak height and area was indicated.

An interesting historical review of thermoluminescence research at the University of Wisconsin was presented by Farrington Daniels, one of the pioneers in this field. His early work extended into geological and archeological studies and included a dosimetry study (using LiF) at an atomic bomb test in 1952. Daniels noted that when J. R. Cameron (University of Wisconsin) and his co-workers later resumed the study of LiF as a thermoluminescent dosimeter, they found that the current supplies of the material had become too pure to exhibit thermoluminescence, and the proper impurities had to be added deliberately to again produce "TLD-grade" LiF.

The diverse program at the University of Wisconsin was evident from a group of papers presented on the physical and dosimetric properties of LiF powder and of small single crystals. Of special interest were the studies by C. R. Rhyner, D. W. Zimmerman, and J. R. Cameron of pre-irradiation annealing. Their work reveals the strong dependence of the thermoluminescence response to γ-rays upon prior thermal history. For many TLD materials the integrated response curve as a function of dose becomes supralinear above some dose value. D. W. Zimmerman, R. W. Bland, and J. R. Cameron presented a mathematical model of this phenomenon, in which this increased response was associated with the creation of new trapping sites by the radiation. The supralinearity property can be exploited to enhance the sensitivity of the system. For LiF (TLD-100), a pre-irradiation dose of 10^5 rads, followed by partial annealing at 300°C for 2 minutes, increased sensitivity sixfold for a subsequent small dose of radiation.

Solid-state dosimeters may be used as dose transfer standards between organizations. However, a serious limitation in using LiF was reported by C. J. Karzmark (Stanford), J. F. Fowler, and J. White, who presented evidence that a hitherto unreported fading may occur between irradiation and readout. Fading amounted to about 15 percent in 3 weeks, but later readout indicated a return toward the initial value. Similar (often more rapid) fading exists in other luminescence dosimeters.

The response of luminescence dosimeters to corpuscular radiation was reported by several laboratories. A decrease in sensitivity and a change in glow curve suggest that exposure to electron or heavy-particle radiation involves different trapping centers than for γ-radiation. Lithium fluoride powder is available enriched in either Li^6 or Li^7. The Li^6F has a very large response to thermal neutrons, while Li^7F responds only slightly and therefore can be used for gamma dosimetry in the presence of thermal neutrons. Lithium fluoride (TLD-100) may be used for a wide range of thermal neutron fluence. However, C. L. Wingate, E. Tochilin, and N. Goldstein (NRDL) reported that a higher temperature is needed for thermal neutron readout because of an additional peak in the glow curve. Moreover, the fast-neutron response of Li^7F per tissue rad is only 2 to 16 percent of that for Co^{60} γ-rays, for neutron energies from 0.25 to 14 Mev, respectively. An approximate fourfold increase of sensitivity of LiF to fast neutrons can be achieved by dispersion of the powder in alcohol. K. Becker and J. W. N. Tuyn (Juelich Nuclear Research Establishment) indicated that pairs of glasses with different compositions could be used to estimate neutron energies and also revealed that these glasses can serve as heavy-charged-particle detectors through counting of surface etch-pits produced by these radiations. A study of β-ray response in LiF as a function of beta energy by J. Kastner (Argonne) and R. Hukkoo showed a sharp decrease in sensitivity for low energy emitters such as Ni^{63} and H^3. Such a response apparently cannot be entirely reconciled with the range of the beta particles in relation to the LiF grain size. This necessitates a careful calibration for each isotope.

For measurements of low doses the readout of thermoluminescent powders, such as LiF, is troubled by an interfering spurious signal. J. F. Fowler (London University) and V. Svarcer reported that mechanical vibration enlarges this spurious signal but the presence of oxygen is required for its observance; a large surface-to-volume ratio increases it. This effect is eliminated in LiF by readout in an inert atmosphere, such as nitrogen, as first noted by R. McCall (Stanford), or by immobilizing the powder, as reported by M. J. Aitken, J. Reid, M. S. Tite, and S. Fleming (Oxford), F. H. Attix (NRL), and J. F. Fowler. A. E. Nash, F. H. Attix, and J. H. Schulman (NRL) reported that the spectra of the spurious signals in LiF and CaF_2:Mn are more complicated than the corresponding thermoluminescence signals induced by γ-rays. The basic processes are not understood, but are evidently related to trapping and emitting centers located at or near the crystal surfaces. Single-crystal LiF blocks of millimeter dimensions, although less subject to the spurious signal, are less convenient to use because of the variation in sensitivity from block to block. N. Suntharalingam, D. W. Zimmerman, and G. N. Kenney (University of Wisconsin) devised a novel solution to the latter problem in which a post-readout calibration is performed on every sample using a built-in source of Sr^{90} radiation. I. Berstein (Con-Rad), B. Bjärngard, and R. C. McCall reported that by imbedding LiF powder in teflon the spurious signal could also be avoided and exposures of 10 mr could be measured with about 25 percent accuracy. Elimination of the spurious signal for CaF_2:Mn is usually accomplished by immobilizing the powder in an evacuated glass tube.

Some spirited discussions arose when comparing film dosimetry with TLD and RPLD for personnel monitoring. R. Maushart and E. Piesch (Karlsruhe Nuclear Research Center) described an extensive personnel monitoring system using phosphate glass with a special filter designed to eliminate the effects of orientation and energy. The operational cost was reported to be significantly lower than that of film. J. Cluchet and H. Joffre (Saclay) discussed the energy dependence of several luminescence dosimeters and recommended LiF because of its near tissue equivalence for protection measurements. On the other hand, W. A. Langmead (Harwell) raised a serious objection to simple tissue-equivalent, solid-state systems for use in personnel monitoring situations. He noted that to comply with the recommendations of the International Commission for Radiation Protection, information about the radition quality is also needed. The multifilter film badge supplies the information by acting as a simple spectrometer. One restriction in the use of thermoluminescence phenomena for monitoring radiation in humans is the erasure of dose information resulting from thermoluminescence readout. R. Schayes (M.B.L.E., Brussels) described methods of multiple readout of natural CaF_2. One method involves controlled heating of phosphor layers; another involves transfer of energy by light stimulation from the 550°C trap into the 250°C trap for a second readout.

Many technical improvements in existing systems were reported. R. Yokota (Toshiba) reported that the new low-Z glasses can measure a 10-mR exposure of Co^{60} γ-rays to an accuracy of $\pm$ 10 percent. An inexpensive portable reader for glass dosimeters was described by W. Buttler (Ladenburg). Photomultiplier dark current is often a limiting factor in the detection of low-dose readouts. By chopping the luminescent output and employing a phase-synchronized amplifier, J. Lippert and V. Mejdahl (Danish AEC, Riso) were able to effect a significant improvement in signal-to-noise ratio which allowed measurement of 5 mr in LiF and 5 μr in $CaSO_4$ with $\pm$ 10 percent accuracy.

Encouraging research is in progress with other materials such as $CaSO_4$, Al_2O_3, carbonates of Ca, Mg, Sr, and Ba, and a thermoluminescent glass. A promising new thermoluminescent material, lithium borate, was described by J. H. Schulman, R. D. Kirk, and E. J. West (NRL). Like LiF, it has little dependence on photon energy, and low production costs may allow the powder to be discarded after one use. It emits in the orange, however, and cannot be used below 100 mr because of competition from infrared radiation. R. J. Ginther (NRL) described a thermoluminescent terbium-activated glass which emits strongly in the violet, thus permitting discrimination against infrared. G. Oster, G. Gabor, and A. Zelechow (Polytechnic Institute of Brooklyn) presented preliminary results on some organic thermoluminescent materials, principally dibenzanthracene. However, the low glow-peak temperatures preclude practical application for dosimetry at present. Similarly the work of K. Miyashita and H. K. Henisch (Pennsylvania State University) in exploring the phenomenon of field-effect enhancement of thermoluminescence is so far limited by glow-peak temperatures below 20°C, very-thin-film phosphors, and electroluminescence. Nevertheless, they have demonstrated the existence of enhancement factors of up to 100. E. N. Sanborn and E. L. Beard (Livermore Radiation Laboratory), described the preparation and characteristics of Ca, Sr, and Mg sulfides, which exhibit luminescence under infrared stimulation. A theoretical discussion of infrared-stimulated luminescence was given by P. Bräunlich, D. Schafer, and A. Scharmann (University of Giessen). Infrared-stimulated systems offer high sensitivity if the fading problem can be solved.

The outstanding advantages for each of the widely used luminescence dosimeters follow: Radiophotoluminescent glasses allow convenient multiple readout. Lithium fluoride is the least energy-dependent for low-energy γ-radiation. Calcium fluoride has high sensitivity and a linear response over the largest dose range, and suffers little radiation damage.

Numerous interesting applications of luminescence dosimetry were described. J. F. Fowler (London University), V. Svarcer, T. J. Deeley, and E. Shuttleworth discussed the results of measuring exit doses from the lungs and pharynxes of patients by means of LiF packets taped to their skin. Substantial departures from water phantom measurements were observed. Thus accurate calculation of dose delivered to a tumor requires measurement of the exit dose for each patient and situation. Moreover, shrinkage of a tumor could be measured during a series of treatments. A. C. Lucas (E.G. & G.) and N. R. French described a radiation ecology study in which miniature TL powder dosimeters, about 1 mm in diameter and 6 mm long, were attached to desert rats near a large, elevated γ-ray source and subsequently recovered. For these small dosimeters, the reported precision of dose measurement of ± 3 percent is remarkably good.

M. J. Aitken and M. S. Tite (Oxford) reported the dating of old pottery (the firing process would have removed any previously stored energy due to natural radioactivity), and T. Higashimura (Kyoto Univ.) presented dose measurements about a nuclear detonation site obtained from the luminescence of roof tiles taken from bombed cities in Japan.

The conference was held under the joint sponsorship of the Atomic Energy Commission, the Office of Naval Research, and Stanford University. A limited number of abstract booklets have been reprinted and are available on a first-request basis from C. J. Karzmark. The conference proceedings are to be published early in 1966 by the Atomic Energy Commission Division of Technical Information, and will be available from the Clearing House for Federal Scientific and Technical Information Springfield, Virginia 22151.

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Forthcoming Events

October

26. American Soc. of **Safety Engineers**, annual, Chicago, Ill. (A. C. Blackman, ASSE, 5 N. Wabash, Chicago, Ill.)

26–28. **Fluid Amplification**, symp., Washington, D.C. (J. M. Kirschner, Fluid Systems Branch, Harry Diamond Laboratories, Washington 20438)

26–28. **Shock and Vibration**, 25th symp., New Orleans, La. (Shock and Vibration Information Center, Code 4021, U.S. Naval Research Laboratory, Washington, D.C. 20390)

26–28. **Spacecraft Sterilization Technology**, natl. conf., NASA, California Inst. of Technology, Pasadena. (L. B. Hall, Code SB, NASA, Washington, D.C. 20546)

26–29. National Soc. for **Clean Air,** 32nd annual conf., Eastbourne, England.