Luminescence Dosimetry

In June 1965, an international conference on luminescence dosimetry was held at Stanford University [Science 150, 391 (1965)]. In the more than 3 years since, the understanding and practical use of luminescence and related trapping phenomena for integrating radiation measurements have undergone a rapid expansion. This expansion was reflected at the second international conference on this topic, held in Gatlinburg, Tennessee, 23-26 September 1968, sponsored by the Oak Ridge National Laboratory. A total of 142 scientists from 11 countries attended the conference.

In the first session, on new thermoluminescent (TL) materials, a report by T. Yamashita et al. (Osaka) was particularly interesting. By activating the most sensitive phosphor known, calcium sulfate, with dysprosium or thulium instead of the previously used manganese, they succeeded in the production of a material which combines high luminescence efficiency (about 2 percent of the absorbed radiation), good linearity of response (10⁻⁴ to 5 \times 10³ roentgens), and relatively good stability (about 5 percent fading during the first month after exposure). Another rare earth activated phosphor, $CaF_2: Dy$ (TLD-200), showed good sensitivity but greater fading (W. Binder et al., University of Wisconsin). For accident dosimetry, either the TL of natural materials occurring in the environment, such as quartz (Y. Ichikawa and T. Higashimura, Nara and Kyoto) or TL materials which can be used as building components such as Al₂O₃ bricks (G. Portal et al., Fonteney-aux-Roses) may be employed. Some materials with rapidly fading low-temperature glowpeaks also permit an estimate of the time at which an accidental exposure occurred (M. Sidran, Grumman Aircraft).

An interesting new TL material, lithium borate, was discussed in the next session. The TL properties of $Li_2B_7O_4$:Mn, first described by J. H. Schulman *et al.* (NRL) at the Stanford conference, have been studied in great

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detail by several groups (P. Christensen, Risö; C. A. Jayachandran et al., London; R. H. Wallace and P. L. Ziemer, Purdue University; and C. R. Wilson and J. R. Cameron, University of Wisconsin). Despite the fact that its sensitivity is somewhat less than that of sensitive LiF (TLD-100), the easy and inexpensive preparation process and tissue-equivalent energy response make this material very attractive for the dosimetry of low-energy photons. The Li₂O, B₂O₃ system can also be used as a base for new types of radiophotoluminescent glasses which combine a low-energy dependence with high thermal stability; their weathering resistance has been improved by additions of BeO (K. Becker and J. S. Cheka, ORNL).

In the following session on nonluminescent dosimeters based on trapping phenomena, conferees became acquainted with one of the most interesting new developments in solid-state dosimetry. As was discovered by J. Kramer (Braunschweig), the optically or thermally stimulated emission of low-energy (< 5 ev) electrons (TSEE) from the surface of irradiated ionic crystals can be used as a method of dosimetry. He reported γ -ray exposure measurements between a few microroentgens and many megaroentgens, with unactivated materials such as CaSO₄ and BaSO₄. Dosimetric proper-ties ["glow-curves," lunar energy transfer (LET) and fast neutron response, fading characteristics, factors affecting the sensitivity] of many other TSEE detector substances such as BeO, LiF, SrSO₄, and Li₂ B_4O_7 , and new possibilities for their use were reported (K. Becker, ORNL). High sensitivity, easy preparation of the detectors, extremely small sensitive layers (< 100 Å), adjustable energy response, higher reading temperatures, and a number of other advantages make TSEE detectors suitable for many dosimetric applications. A great number of questions related to types of trapping centers, the role of activators (Z. D. Spurny, Prague), and others still remain unanswered. However, with a rapidly increasing number of scientists

working in this field, interesting developments can be expected during the next few years. Other nonluminescent processes with potential dosimetric applications include the stimulated "current-glow" and pyroelectric processes in ionic crystals (H. K. Henisch and D. Bose, Pennsylvania State University).

Several papers dealt with the problem of "spurious" thermoluminescence and its reduction for the measurement of low doses (M. J. Aitken, S. J. Fleming et al., Oxford), and two sessions were devoted to the mechanism of thermoluminescence and related problems of radiation sensitization and supralinearity, mainly in lithium fluoride. Despite the extensive investigations of these effects, particularly of the commercial LiF TLD-100, they remain largely unexplained. There still exists a disagreement as to the identity of the charge carriers released from traps during the heating process in LiF (TLD-100). Mayhew et al. (Dartmouth) presented evidence for electrons, while Claffy et al. (NRL) reported results favoring holes. Alternative explanations were also given for supralinearity in that phosphor. Cameron et al. (University of Wisconsin) proposed that the filling of competing deep traps allowed more charge carriers to reach the normal TL traps at large doses, while Claffy et al. (NRL) suggested that supralinearity is a phenomenon resulting from interaction of charge carriers from neighboring Compton-electron tracks when they become close enough together.

In two other sessions, the relative response of luminescence detectors to x-ray and gamma radiation, to charged particles, and to neutrons was discussed. There appears to be a difference between the calculated and the measured photon energy response in some low-Z materials (BeO, LiF, and $Li_2B_4O_7$) in the 10- to 100-kev photon energy range and a sensitivity drop in LiF above 1.25 Mev which are difficult to explain (C. J. Karzmark and J. Geisselroder, Stanford; P. R. Almond et al., Houston, and others). In some substances such as LiF, the response decreases with increasing LET of the radiation, but in other materials such as BeO it appears to increase (E. Tochilin et al., USNRDL). In radiophotoluminescent (RPL) glasses, high-LET radiation such as caused by (n,α) reactions during exposure to thermal neutrons creates different centers than does gamma radiation. Differences in the RPL spectra and the build-up and

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fading kinetics between gamma and neutron exposed glasses have been observed (K. Becker and J. S. Cheka, ORNL). These glasses also exhibit thermoluminescence (D. F. Regulla, München).

Luminescence characteristics such as the thermal quenching (S. G. Gorbics *et al.*, NRL), the intrinsic sensitivity of TL phosphors (W. H. Lucke, NRL), and the optical spectrum of thermoluminescence (B. G. Oltman and J. Kastner, ANL; A. M. Strash and R. Madey, Clarkson College; S. J. Fleming, Oxford) were subjects of another session. A computer program for the analysis of LiF glow-curves based on the Randall-Wilkins model has been developed (R. M. Grant, Jr., and W. S. Stowe, Denison University).

In the instrumentation field, several new developments have been reported, in particular a fast sample changer for TLD readers (L. Bötter-Jensen, Risö) and a semiautomatic TLD reader based on the use of hot nitrogen gas for heating the sample (K. R. Petrock and D. E. Jones, Livermore). J. Kastner et al. (ANL) improved their technique of predose suppression in RPL glasses by delayed measurement after pulsed excitation through the use of an electrooptical Pockels shutter instead of the expensive pulsed ultraviolet laser previously employed. By allowing the "predose" luminescence to die away before measuring the longer lived RPL signal, the practical range of silveractivated glass can be extended to less than 1 milliroentgen.

Applications of RPL and TL dosimetry in personnel dosimetry are rapidly becoming routine. In the NRTS in Idaho Falls, for example, several thousand workers are regularly monitored with a badge containing LiF (J. P. Cusimano et al.), and in Karlsruhe, glass dosimeters whose encapsulation flattens the energy response or makes it similar to that of critical organs, are used for the same purpose (E. Piesch). Both TL and RPL dosimeters were shown to be more accurate and less expensive than film badges. More sophisticated solid-state badges (A. R. Jones, Chalk River) and automatic reading equipment are under development in several laboratories.

Another application of thermoluminescence is that of dating ancient pottery. New results on the improvement of the methods were reported from Risö (V. Mejdahl) and Oxford (D. W. Zimmerman). The head of the Oxford group, M. J. Aitken, was the banquet



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speaker and gave an impressive description of other methods used for the dating of ancient pottery and the detection of antique forgeries.

In a session on the medical applications of luminescence dosimetry, attention was drawn to the high toxicity of ingested LiF (C. M. Dettmer and B. M. Galkin, Stein Research Center) and the changes that occur in nonencapsulated, implanted LiF dosimeters used for in vivo studies. In some circumstances, implanted detectors were totally disintegrated or dissolved in body fluids. Some prepacked LiF dosimeters have been used for clinical routine measurements with excellent precision of ± 3 percent (N. Suntharalingam, Stein Research Center).

In a final panel discussion, moderated by J. H. Schulman (NRL), the conference chairman J. R. Cameron and panel members F. H. Attix, K. Becker, J. F. Fowler, V. Mejdahl, and Z. D. Spurny, summarized their impressions of the conference and outlined desirable future work. Their individual statements may be summarized as follows:

1) It is undesirable and unsatisfying to spend further basic research efforts on commercial LiF materials of unknown composition. Instead, systematic studies with well-defined, reproducible materials should be conducted. The characteristics of commercial phosphors may, however, be worthy of critical study because of practical aspects.

2) The increasing use of TL and RPL detectors for routine applications makes the development of improved devices such as more sophisticated (but easy to handle) personnel dosimetry badges and automatic readers desirable.

3) Unnecessary duplication of work could be avoided and research stimulated by the creation of an information center for the efficient collection and rapid distribution of all relevant information.

4) The problem of sensitive, reasonably energy-independent fast neutron dosimetry needs to be solved. Detectors with a high sensitivity for high-LET radiation should be developed.

5) The promising method of thermally stimulated exoelectron emission dosimetry may in the future provide considerable improvements in solidstate techniques as well as in the understanding of the basic processes involved.

Finally, the reviewers concluded that researchers in luminescence dosimetry



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should meet at approximately 2- or 3year intervals. This small but vigorous conference has, without doubt, stimulated many to change their studies in various ways to make them more productive. The full text of the papers and discussions is available from the Clearinghouse for Federal Scientific and Technical Information, Springfield, Virginia 22151.

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Calendar of Events

Courses

Fluid Mechanics, Urbana, Ill., 11–15 August. It will cover fundamental aspects of fluid mechanics and the principal applications to the paper industry. (H. O. Teeple, Technical Association of the Pulp and Paper Industry, 360 Lexington Ave., New York 10017)

Trace Element Analysis, Harwell, England, 30 June-4 July. Will emphasize nuclear techniques in trace element analysis, including neutron, charged particles and gamma-ray activation analysis, autoradiography, and mass spectrometric isotopic dilution analysis. It should appeal to chemists and metallurgists working in such fields as ferrous and non-ferrous metals, refractories, glass, plastics, and so forth. (Education Officer, Royal Institute of Chemistry, 30 Russell Square, London, W.C.1, England)

Automatic Control Theory, St. Louis, Mo., 11-16 August. The course will cover foundations of modern control theory, finite automatia and dynamical systems, stability-operator theoretic methods, foundations of optimal control theory, differential games and minimax problems, optimal control of distributed parameter systems, linear and nonlinear filtering, stochastic optimal control, and new developments in designing large linear systems. The prerequisite mathematical level of students will be a good M.S. level of preparation; however, the lectures will be conducted in a manner suitable for stimulating even the advanced research worker. (Dr. G. L. Esterson, Box 1048, Division of Continuing Professional Education, Washington University, St. Louis 63130)

Biology of Aging, University of California, San Diego, at La Jolla, 22 June– 11 July. Is intended for senior predoctoral and early postdoctoral students in order to interest them in research on the biology of aging. The course content will include the aging process, environment and aging, mammalian aging, cellular aging, and subcellular and molecular aging. Costs will be paid by the Adult Development and Aging Branch, National Institute of Child Health and Human Development. (Dr. Gabe Maletta, Adult