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SOME ADVANCES IN SOLID-STATE
FAST NEUTRON DOSIMETRY*

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ABSTRACT

Progress which has been made at ORNL in solid-state fast neutron dosimetry since the last review on this subject was presented in 1972 is summarized with emphasis on the following topics:

1. Further studies on the directional and energy response characteristics of TSEE detectors based on ceramic BeO covered with hydrogenous radiators, and on a new method for extending the dynamic range of gas-flow TSEE readers by two orders of magnitude;
2. Experiments with highly dispersed and sensitive TLD phosphors permanently embedded in high-melting organic radiators such as p-sexiphenyl;
3. Investigations on the automatic spark counting of neutron-induced recoil particle tracks in thin polymer foils after extended etching; and
4. Studies concerning the feasibility of intercomparisons by mail of neutron sources with an accuracy of better than $\pm 5\%$ in the ~ 50 -500 rad range by using fission fragment track etching.

The relative merits and disadvantages of the various methods, for personnel monitoring and other applications, are discussed briefly, and a few other promising approaches are mentioned.

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1. Introduction.

The development of sufficiently sensitive, stable, accurate and inexpensive integrating dosimeters for fast neutrons remains one of the more important, but essentially unsolved problems in radiation protection and research. In personnel monitoring, considerable efforts have been devoted in recent years to "albedo" techniques which are based on the moderating and backscattering properties of the human thorax. Despite all the development and testing, mostly of $^6\text{LiF}/^7\text{LiF}$ TLD pairs in a wide variety of combinations with various absorbers and moderators (for a recent compilation see ref. 1), it has not been possible to overcome the intrinsic problems of this technique, namely a rapid loss in sensitivity for neutron energies exceeding 10-100 keV; a pronounced directional dependence of response; and the strong influence of the distance between body surface and detector on the results. Such a detector should, therefore, be considered only as a semi-quantitative supplement to fast neutron dosimeters in situations where a large intermediate energy neutron contribution to total dose is to be expected.

The best neutron energy response so far, excellent sensitivity, stability as well as a wide dynamic range can be obtained with thermal neutron detectors in the center of large Bonner-type moderator spheres or cylinders. Unfortunately, such devices are very bulky and heavy, and their spatial resolution is poor. Their use is restricted to area monitoring, and perhaps some calibration situations.

More promising, but also restricted in their practical use by the large neutron to gamma dose ratio which is required for sufficiently precise measurements are fast neutron dosimeters which are based on the detection of differences in the readings of two detectors, usually either TLDs or TSEEs, only one of which contains hydrogen and responds consequently to the neutron-induced recoil protons. The high-LET to low-LET (or recoil proton to gamma radiation) response ratio of all known solid-state dosimeters, which are not based on track registration or nuclear interactions, amounts (depending on their electron trap density) to between about 0.05 and 0.5 on a rad/rad basis. No significant improvement of this figure is to be expected, if one accepts the best available model of the processes involved⁽³⁾ as correct. Assuming a QF for fast neutrons of 10, and a precision in the individual reading of a detector of about $\pm 5\%$, it is obvious that the measurability of neutron doses and subsequent evaluation in dose equivalent (rem) is limited to neutron to gamma ratios higher than one. As the gamma radiation contamination in many fast neutron sources is small, detectors of this type may, however, have some future in radiobiology and neutron therapy.

Finally, there are detectors based on threshold reactions, usually nuclear track formation, which are essentially insensitive to low-LET types of radiation. In the long run, they appear to be most interesting for personnel as well as radiobiological fast neutron dosimetry. This paper discusses some work on the two latter approaches which has been carried out by the ORNL Solid-State Dosimetry Research Group since late in 1972, when the last review on our work in integrating fast neutron dosimetry was presented at an IAEA Symposium on Neutron Monitoring in Vienna⁽⁴⁾.

2. Thermoluminescence.

Various investigators have attempted to increase the low inherent fast neutron sensitivity of inorganic thermoluminescent (TL) materials by mixing them with liquid or solid (5; and others) organic recoil proton radiators, but all the systems described so far require the separation of the constituents prior to evaluation. This is necessary because most hydrogen-rich organic compounds evaporate and/or disintegrate at temperatures well below those which are required for the read-out and annealing of the more stable TLB materials. Covering of TL phosphor layers with radiators results in a strong neutron energy dependence. Organic TL phosphors fade rapidly and/or are very insensitive.

In our studies,⁽⁶⁾ finely powdered ($<4\text{ }\mu\text{m}$), highly sensitive and stable TL phosphors such as $\text{CaSO}_4:\text{Dy}$, $\text{CaSO}_4:\text{Tm}$ or $\text{Mg}_2\text{SiO}_4:\text{Tb}$ have been embedded into high-melting organics, for instance p-quaterphenyl or p-sexiphenyl (M.P. $\sim 450^\circ\text{C}$), for fast neutron dosimetry via recoil proton registration. If exposed to the mixed radiation field of the Health Physics Research Reactor (HPRR), such detectors exhibit a response three times higher than Teflon-embedded phosphors having the same gamma radiation sensitivity, the response difference being due to the recoil protons produced by fission neutrons. This indicates about 50 percent efficiency of the recoil protons in producing a TL signal in the phosphor. For 14 MeV neutrons from the (d,T) reaction, the response ratio is 10. The detectors are prepared by hot-pressing the constituents into re-usable pellets, followed by out-gassing in a vacuum furnace prior to first use. An increase in the visible light sensitivity was observed only in the p-sexiphenyl-embedded (not in the Teflon-embedded) material. Other authors (20) have recently obtained a rad-equivalent response for 14 MeV neutrons using a similar system, but radiation cross-linked polyethylene (melting point 350°C , $\sim 14\%$ hydrogen) as a proton radiator.

It should, incidentally, be noted for applications of TLB in intercomparisons or in area monitoring, which both may involve relatively long storage of exposed detectors prior to their evaluation, that the widely used LiF:Mg , Ti (TLD-100, 600, or 700) should not be assumed to be perfectly stable.

It was shown in field tests involving three months of detector storage in tropical climates that other phosphors such as $\text{CaSO}_4:\text{Dy}$ and $\text{Mg}_2\text{SiO}_4:\text{Tb}$ are more stable⁽⁷⁾.

3. Thermally Stimulated Exoelectron Emission.

As has been reported previously^(9,11), the difference in the response of pairs of ceramic BeO discs can be used for the integrating measurement of fast neutrons between at least 0.1 and 20 MeV (the face of one of the discs is covered with a hydrogenous radiator such as polyethylene, and of the other with a non-hydrogenous low-Z material such as graphite or Teflon). In the 1972 and 1973 ORNL Dosimetry Intercomparison Studies, the TSEE results for neutron and gamma radiation measurements at the HPRR reactor were encouraging, and reproducibilities of $\pm 3\text{-}5\%$ are now feasible with well-stabilized and calibrated detectors⁽¹⁰⁾. In further studies on this method, the directional response of simple and more complex multi-detector units has been investigated.

For a single radiator-covered detector, the response drops rapidly, as expected, once the angle of neutron incidence approaches or exceeds $\sim 90^\circ$ (Fig. 1). The directional response becomes less pronounced with increasing neutron energy. Another arrangement consists of placing one radiator between two BeO disks. With the two radiator-detector interfaces being read out, a much less directional dependent response is observed. In addition, information on the direction of neutron incidence can be obtained from the evaluation of such a dosimeter⁽¹¹⁾. An even more accurate localization of the direction of neutron incidence is possible with a cubical or cylindrical unit containing six detectors. It may be possible to reduce the number of detectors necessary by making use of the effect that the TL and TSEE peaks in ceramic BeO are well separated.⁽¹⁰⁾

Information on the average energy and energy distribution of fast neutrons up to high energies can be obtained by either measuring the neutron response as a function of radiator thickness (Fig. 2), or by placing absorbers of different thickness between the (thick) radiator and the TSEE detector. One could, perhaps, also design a dosimeter covering a wide neutron energy range based on a combination of albedo techniques for low and intermediate neutron energies, and recoil proton registration for higher neutron energies⁽¹¹⁾.

The measurement of high radiation doses with TL or TSEE readers is frequently limited not so much by the response characteristics of the detector (saturation), but by the maximum light-intensity, or count-rate, which the readout instrument is able to process. In a GM counter, as it is used routinely for low-dose TSEE measurements at ORNL, for example, a "breakdown" in the counter performance occurs already at gamma radiation levels of 0.1 rad, corresponding to neutron radiation doses of about one rad (Fig. 3). This makes it necessary to read higher doses with another type of readout instrument such as a proportional counter with much shorter lead-time, or with an ionization chamber. It is, however, also possible to "reconstruct" the full peak, and to determine the area under it by using one of several computational methods(12).

For example, it is safe to assume that the glow-curves scale approximately linearly both in X and Y coordinates. A standard curve is adopted, its area determined, then the scaling factors required to transform an arbitrary TSEE curve to the standard curve can be used to find the area under the former curve. To determine the appropriate scale factors when the values near the peak are unreliable, an Elograph* digitizer interfaced to a programmable calculator is used to determine a fit to the two wings of the curve (Fig. 4). Using a parabola $Y = a(x-x_0)^2$ to describe the low temperature side and a straight line $Y = a(x-x_0)$ to describe the approximately linear region of the high temperature side, one obtains

$$\text{X-Scale factor} = \frac{x_1 - x_0}{x'_1 - x'_0}, \quad \text{Y-Scale factor} = \frac{a}{a'} \left(\frac{x_1 - x_0}{x'_1 - x'_0} \right)^2,$$

where unprimed values refer to the standard curve and primed values to the arbitrary curve under examination. As can be seen in Figs. 5 and 6, this method enables one to extend the useful range of GM and proportional readers by about two orders of magnitude towards higher doses. An alternative method, based on the assumption that the curves are approximately gaussian in shape, leads to very similar results.

4. Spark Counting of Neutron-Induced Recoil Particle Tracks in Polymers

Among the more promising dosimeters with a selective fast neutron response are track etching dosimeters (for recent reviews, see Refs. 1 and 13). There have been essentially two approaches with different advantages and disadvantages. One possibility is to record fission fragments from ^{237}Np , ^{232}Th , etc., preferably in thin polycarbonate foils. This permits fast automatic evaluation of the resulting perforations in the foils by spark counting, but

*Elographics, Inc., Oak Ridge, Tenn./U.S.A.

also requires the use of fairly large quantities of radioactive materials which may restrict their large-scale use.

The other possibility is based on the detection of the recoil nuclei tracks which the neutrons produce by interaction with foil (or foil cover) constituents such as carbon and oxygen (it is more difficult to record recoil proton tracks). The recoil tracks could, however, so far not be counted efficiently with the spark counter due to their short range. We found recently that spark counting of heavy recoil particles is possible if the foils are etched to a point where electrical breakdown begins to occur even in unexposed foils⁽¹⁴⁾.

As can be seen in Fig. 7, the background spark count of such an "over-etched", unexposed commercial cellulose nitrate foil (Kodak-Pathé LR 115 strippable) is increased by exposure to fission neutrons. There is no well-defined plateau in the spark-count vs. voltage diagram and the count increases during repeated sparkings of the same foil. Optimum etching conditions have to be established separately for each foil by determining the signal-to-background ratio as a function of sparking voltage and etching time. The sensitivity increases with increasing foil sensitivity and neutron energy, but even in less sensitive Kimfol polycarbonate foils, 3 rad of fission neutrons produce a count substantially higher than background (Fig. 8).

The count was consistently higher in Teflon-covered foils than in polyethylene-covered ones for different foil materials and neutron energies, indicating that few, if any, recoil proton tracks are sparked. A similar acceleration in electric breakdown is produced by very high ($\sim 10^4 - 10^6$ R) gamma radiation doses. This does, however, not interfere with most dosimetric applications of this technique. The method shows promise in personnel dosimetry as well as for special studies such as depth-dose measurements and bone-tissue interface dosimetry.

5. Precision of Fast Neutron Dosimetry by Track Etching.

In the past years, emphasis of our track etching fast neutron dosimetry work has been on the detector sensitivity, with the measurability of the low doses encountered in personnel dosimetry being the most important goal and the precision of secondary importance. As it became apparent since, there is frequently a need for the accurate measurements of fairly high doses in the 50-500 rad range, for example, in accident dosimetry, radiobiological or radiotherapeutical work, or for the intercomparison of neutron sources by mail. The latter application is the subject of a Research Agreement between ORNL and IAEA. As track etching detectors are most likely to meet the re-

quirements for mailable detectors (they are light, inexpensive, not breakable, have a high spatial resolution, and exhibit no fading under the conditions to be expected), it had to be determined if accuracies consistently better than $\pm 5\%$ can be obtained in neutron flux (or dose) measurements by track etching.

So far, the following methods have been studied, using fission neutrons from the HPRR and monoenergetic neutrons between 0.5 and 15 MeV.

1. Lexan disks, glass microscope slides, and commercial cellulose nitrate* track detectors in direct contact with a thin (0.05 mm) thorium metal foil, evaluated after etching by microscopic track counting at 100 to 400 times magnification;
2. The same three detector types in direct contact with a foil consisting of 2 mg/cm² of a mixture of 5% ²³⁷Np and 95% Au (by weight) on a 0.16 mm Ag backing and covered by a protective gold film 0.45 mg/cm² in thickness,** evaluated also by microscopic counting; and
3. Thin (10 μ m) Kimfol foils exposed in contact with a thick layer of a material consisting of 2% ThO₂ and 98% W. The foils are evaluated by spark counting (presparking at 760 V, counting by three sparkings at 500 V after 3 h of etching at $43 \pm 0.5^\circ\text{C}$ in KOH of 1.239 g/cm³).

With repeated exposures and etchings of 10 foils each over a period of several months, standard deviations of 2 to 3.5% have been obtained for both spark counting and visual counting by different persons. The time required for equally precise measurements was less for visual than for spark counting at the relatively high track densities ($\sim 10^2 - 10^3$ tracks/mm²) encountered in high dose measurements ($\sim 50 - 500$ rad). Also, the visual counting is much less dependent on small variations in etching and evaluation conditions.

It was observed that although repeated sparking of the same foil may yield reproducibilities within ± 2 counts, the foil to foil reproducibility within one experiment rarely is less than 2-3% for one standard deviation, and deviations up to $\pm 5\%$ occur from one test (involving separate irradiations, etchings, and sparking by different investigators) to the next. Studies concerning the influence of environmental parameters on the characteristics of spark counters have been carried out using a specially designed counter (Fig. 9) which permits exact definition of counting gas, gas pressure, etc.

*8 μ m red dyed C.N. on polyester carrier, LR 115 by Kodak Pathe, France.

**Nucl. Radiat. Developm., North Grand Island, N.Y.

Apparently, normal fluctuations in ambient temperature, air pressure and humidity have only little effect, and less influence of the counting polarity on the sparking plateaus has been found⁽¹⁴⁾ than was reported by other investigators.⁽¹⁵⁾

In the visual counting of fission fragment tracks in polymer foils, one of the more important factors limiting the precision, in particular at higher neutron energies, is the overlap in the size distributions of the smaller recoil and alpha particle tracks and the larger fission fragment tracks. This is schematically illustrated in Fig. 10. Obviously, depending on etching time, magnification, observer, etc., the counting threshold will vary slightly, amounting to variations up to several percent above the expected statistical counting errors. This problem can be overcome by using less sensitive detectors (glass) in combination with a collimating device which narrows the distribution in energies and incidence angles of the fission fragments on the detector. Further details on this work will be published elsewhere⁽¹⁶⁾.

6. Other Potentially Promising Systems.

It is difficult to anticipate or judge the likely course and value of future research and developments, but I would like to conclude by outlining a few other areas which, besides the ones already discussed, may deserve some attention during our long march towards the perfect solid-state fast neutron dosimeter:

1. It is apparently possible now to etch recoil proton tracks with good efficiency in cellulose nitrate⁽¹⁷⁾, and whether this method could be developed into a sufficiently reliable, automatically readable system should be investigated;
2. Silicon diodes, if made more uniform in response, annealable and/or much less expensive, could become a versatile dosimeter for the dose range of interest in radiobiology and medicine;
3. For dose levels above ~10 rad, the measurement of stable free radicals in neutron-irradiated organics could also provide an excellent, apparently almost rad-equivalent fast neutron dosimeter; there are at least two methods, EPR and lyoluminescence⁽¹⁸⁾, which could provide simple and sensitive measurements of the radiation-induced free radicals; and
4. Radiation-induced polarization or depolarization phenomena in organics, similar to those currently being investigated in inorganic compounds (see, for example, ref. 19), could turn out to be valuable tools if the present problems in sensitivity, stability and reproducibility are surmountable.

We can, in summary, look forward with some anticipation to further interesting progress in this field.

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Figure Captions.

Fig. 1. Directional Response of a ceramic BeO disk TSEE dosimeter, covered with a 2 mm polyethylene layer as recoil proton radiator, for different neutron energies.

Fig. 2. Relative fast neutron response of TSEE dosimeters as a function of the thickness of the polyethylene radiator, for fission and 14 MeV neutrons.

Fig. 3. Breakdown in the capacity of a GM counter to record the complete TSEE curve at higher dose levels (1 rad of gamma radiation corresponds to about 0.1-0.2 rad of fast neutrons).

Fig. 4. Schematic diagram for parabolic-linear correction of TSEE or TL glow curves.

Fig. 5. Dynamic range of a GM counter for TSEE measurements, without and with computational "reconstruction" of incomplete glow curves.

Fig. 6. Same as Fig. 5, but dose range extending to higher dose levels because of reduced dead-time losses in a proportional counter.

Fig. 7. Spark counts in "overetched" commercial cellulose nitrate foils as a function of sparking voltage without, and after exposure to 3 rad fission neutrons.

Fig. 8. Replica of foil perforations in aluminized Mylar after sparking of polycarbonate (10 μ m Kimfol) without neutron exposure (top), and after exposure to 3 rad of fission neutrons; 9 h of etching at 40°C in 28% KOH, sparking voltage 500 V.

Fig. 9. Device for track spark counting under conditions of controlled atmospheric composition, pressure, temperature, and humidity.

Fig. 10. Schematical presentation of the overlap in the etch pit size distributions from fission fragment and recoil/alpha particle tracks in an etched polymer foil, which introduces a source of subjective error into visual track counting and becomes more pronounced with increasing neutron energy.

Fig. 1

ORNL-DWG 73-5248

PROCEDURE FOR PARABOLIC-LINEAR CORRECTION

ORNL - DWG 73-9450

Fig. 2. Spark counts in cellulose nitrate (LR 115), etched 90 minutes at 40°C in 28% KOH, without irradiation, and after exposure to 3 rad NPPR fission neutrons.

Fig. 27