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SECOND AEC WORKSHOP ON PERSONNEL
NEUTRON DOSIMETRY

Battelle

Pacific Northwest Laboratories
Richland, Washington 99352

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SECOND AEC WORKSHOP ON PERSONNEL NEUTRON DOSIMETRY

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INDEX

	<u>Page</u>
Foreword	1
Participants.	2
Summary	3
Oak Ridge National Laboratory	5
Lawrence Livermore Laboratory, Livermore.	25
Chalk River Nuclear Laboratories, Chalk River, Ont.	29
Brookhaven National Laboratory.	33
Battelle-Northwest.	41
Rocky Flats	42
Savannah River Plant.	54
University of Wisconsin	77
Stanford Linear Accelerator	83
Spark Counting Survey Summary	95

SECOND AEC WORKSHOP ON PERSONNEL NEUTRON DOSIMETRY

FOREWORD

The first AEC Workshop on Personnel Neutron Dosimetry was held September 23-24, 1969, in Bethesda, Maryland. In the approximate two years since the first meeting, research and development activities directed toward the practical use of an improved neutron dosimeter were continued at many facilities. The results of these programs are summarized in this report.

This meeting followed the precedent set in the first meeting by providing a fruitful and uninhibited exchange of current personnel neutron dosimeter development activities.

This report presents an overall summary of the papers and the meeting discussions followed by the detailed summaries. The editor has prepared the Summary of Discussions and presents it as his review of the the discussions - not the papers presented. It was not reviewed by the participants. Readers of this report are urged to read the participants' summaries for specific information on personnel neutron dosimeter developments and to contact the participants directly for the best current status of any particular program. Note also that all graphic materials were reproduced directly from the copy provided to the editor and vary widely in reproduction quality.

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SUMMARY OF DISCUSSIONS

Neutron dosimeters utilizing the Albedo neutrons were described by representatives from several facilities. These systems require a fixed or generally well known spacing between the dosimeter and the body of the individual. Usually direct contact is recommended. Energy dependence is still an important problem, but the dosimeters can give good results when properly worn and when calibrated with neutron spectra approximating those to which they will be exposed.

Throughout the meeting it was clear that additional dose rate data for standard neutron sources is required. Specifically, rem/hr or rad/hr values versus neutron source size need to be standardized and uniformly used by all developing or comparing neutron dosimeters. Some of the variations of performance results noted for neutron dosimeters is believed to be primarily related to the neutron dose assigned to various neutron sources and to variations in energy spectrum.

Some study of anthracene for use as a neutron dosimeter were reported. It appears that there may be some LET response capability for this material. It was also pointed out that when light emissions in the red spectrum range are to be measured, photodiodes might be better light integrators than photomultiplier tubes. The photodiodes are slow in response but are very stable.

Fission fragment dosimeters are currently used as fixed area monitors. Around high energy accelerations, care should be taken when using fission fragment dosimeters to avoid interference that may result from high energy photofission events.

Considerable discussion on the use of spark counting equipment for track counting to interpret fission fragment dosimeter was presented. Emphasis was placed on the importance of detector materials, etching techniques, electronic discharge problems, and overall system performance factors to the use of these methods. Track size was reported to be a function of neutron energy to some degree and studies in this area may hold a potential for neutron spectrum measurement.

Considerable discussion developed relating to the use of Thorium-232 for fission fragment dosimeters. Thorium was preferred to uranium by some due to the limited surface oxidation, but others believe the beta-gamma radiations from the foils were excessive for personnel dosimeter use. The question left unanswered is, "What is the dose rate from small foils of Thorium-232 and how significant is this dose rate compared to the improved neutron dose measurements a thorium foil fission fragment dosimeter system may provide"?

The use of Neptunium-237 fission fragment dosimeters was also thoroughly discussed. Some gamma dose also originates within this detector foil. It was reported that a neptunium aluminum alloy containing only 1/2 milligram of neptunium has a 90% chance of detecting a 40% mrem dose over a one month period.

New TLD neutron dosimetry systems that are now used routinely were described. Generally with proper care in wearing, calibrating and readout, good results were attained. Some difficulty in TLD neutron dosimetry systems may arise due to the heating technique that develops large temperature gradients across the TLD chip and, hence, may not completely readout all stored radiation dose information. The use of hot gases for more uniform heating of chips was discussed. In reality, most heat transfer even in the lot planchet system is by hot gases, but primarily from only a single side. No work is currently known about the potential use of microwave heating for readout of TLD chips.

Recent advances in thermally stimulated exoelectron emissions (TSEE) systems were discussed, including research studies that seem to establish silicon as the activating material. Encouraging dosimetry results were reported with these systems. The detection of 0.1 rad of neutron dose in a dosimeter sensitivity layer not exceeding a few tens of Å was reported. Commonly these systems yield 150,000 counts per mrad. Small spherical pocket type TSEE devices are being used in both Germany and Russia. A small external heat coil provides the heat for readout and a counting wire through the dosimeter provides the readout signal in these foreign systems. As with TLD's the readout heat provides the necessary system annealing and the dosimeters can be used repeatedly.

PERSONNEL NEUTRON DOSIMETRY RESEARCH AT ORNL*

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1. INTRODUCTION

The exceedingly poor stability of the conventional NTA Neutron Monitoring Film at higher humidities strongly suggests that its use should be discontinued in many cases where it is used presently, even if no replacement is available. Due to its rapid fading (in a recent test, Figure 1, 50% fading occurred in 1.5 days),⁽¹⁾ the readings of this film lead to a dangerous underestimation of the fast neutron dose. Additional sealing of the day film in thin polymer foils usually only delays the penetration of humidity by a few days.

Work on neutron dosimetry of the Applied Dosimetry Group during the last 18 months since the previous workshop has focused on two techniques, namely thermally stimulated exoelectron emission (TSEE) and track etching. Much progress was made in the development of highly sensitive, stable and reproducible TSEE detectors based on a non-toxic ceramic BeO (Brush Beryllium Co. Therma-lox 995).⁽²⁻⁴⁾ It was, for example, discovered that sensitivity and stability can be optimized by a 1400°C heat pretreatment followed by a few hours of storage in liquid water. Using highly efficient proportional counters to count the electrons, sensitivities of 10^4 counts/mR/cm² for gamma radiation have been obtained. Si⁴⁺ in the form of a Be₂SiO₄/BeO eutectoid is responsible for the main TSEE peak at ~325°C. High-temperature peaks (~450 and 550°C) can be increased by Li⁺ doping or exposure to a high flux of heavy charged particles.

2. TSEE NEUTRON DOSIMETRY WITH BeO

BeO has a low inherent sensitivity to thermal and fast neutrons. The thermal neutron response can be increased easily to about 30-40 times the gamma response (on a rad/rad basis) by covering the sensitive surface with a suitable

* Research sponsored by the U.S. Atomic Energy Commission under contract with the Union Carbide Corporation.

converter, for instance LiF , $\text{Li}_2\text{B}_4\text{O}_7$, BN or Cd . It should be noted that due to an LET dependence of the TSEE peak ratios in BeO , the high-temperature peaks will be enhanced if (n,α) converters are used. Also, exposure to Thermalox 995 to total charged particle doses exceeding 500 to 1000 rad creates some measurable semi-permanent changes in its radiation response characteristics which require temperatures around 1400°C for annealing.⁽¹⁾

It has been shown in previous studies⁽⁵⁾ that TSEE dosimeters, covered with polyethylene or graphite, exhibit a difference in response due to the effect of recoil protons from the polyethylene. Such a detector pair has been used initially for fission neutron dosimetry in the mixed radiation field of the fast, unshielded HPRR reactor.⁽⁵⁾ Because the extremely thin sensitive layer in TSEE dosimeters permits the registration of rather low-energy recoil protons, more advanced detectors and instrumentation have been employed in a detailed study of this system.⁽⁶⁾

The recoil proton "radiator" can be composed of either gaseous (e.g. an organic gas), liquid, or solid hydrogenous materials. Submersion of ceramic BeO discs in $\text{C}_2\text{H}_5\text{OH}$ as a liquid radiator, for example, did not affect gamma sensitivity or fading properties; $\text{C}_2\text{H}_5\text{OH}$ can be removed by drying the detectors prior to reading for several minutes at 100°C . In most of the experiments, however, solid radiators have been used. Sets of detectors were covered with one mm of polyethylene and one mm of reactor grade graphite respectively, and exposed to a fluence of 6×10^6 to 2×10^7 n/cm^2 of monoenergetic, monodirectional fast neutrons from the reactions $^7\text{Li}(p,n)$ (60 and 100 keV); $^3\text{H}(p,n)$ (0.2, 0.5 and 1 MeV); and $^3\text{H}(d,n)$ (16 MeV) at a distance of 10 cm from the target of a Van-de-Graaff generator. Fluences were determined by means of a calibrated long-counter.

No significant absorption of recoil protons was anticipated in the thin air layer between radiator and detector in this energy range. For lower neutron energies, however, a more intimate contact with liquid or gaseous radiators may be preferable. During linear heating, the exoelectron emission was recorded as a function of temperature. Figure 2 gives the results obtained with similar combinations of teflon and polyethylene covered BeO discs.⁽²⁾ The net counts

of the detectors were weighted by their calibration factors for ^{60}Co gamma radiation, averaged, and the difference expressed in R gamma response equivalent, then divided by the absorbed dose for fast neutrons in tissue as calculated from kerma.

The results, as summarized in Figure 3, indicate an increase in the fast neutron sensitivity of this system from ~ 0.18 at 100 keV to ~ 0.28 of the gamma radiation response at 16 MeV.

Obviously, the system is potentially more sensitive than any other integrating fast neutron detector, but suffers from a general drawback of all solid-state dosimeters, namely their decreasing sensitivity with increasing LET. It is not known yet to what extent this problem can be overcome by the use of other TSEE detectors which have such a LET dependence of their peak ratios that partial reading (annealing of a certain peak) may permit a discrimination against low-LET radiation. Work along these lines is in progress.

3. USE OF FISSION FRAGMENT TRACK ETCHING IN PERSONNEL DOSIMETRY

Obviously, the easiest way to proceed in the application of track etching would be the use of fissionable converter foils and evaluation of the etched thin foils by spark counting. However, this method has several drawbacks. Although above approximately 2 MeV, the fission cross section of ^{238}U agrees within better than $\pm 30\%$ with the ICRP recommended neutron rem curve, in the region below ~ 2 MeV a rapid drop in sensitivity occurs. The same is true for thorium. However, ^{237}Np has a lower neutron energy "threshold" at about 0.6 MeV, and a higher cross-section insures a higher sensitivity (at 3 MeV, the ^{237}Np fission cross-section is twelve times higher than in ^{232}Th). On the other hand, ^{237}Np is more expensive and more difficult to handle than natural U or Th.

Radiotoxicity makes the use of plutonium impractical in personnel monitoring. A high spontaneous fission rate in ^{238}U causes a considerable "background" increase corresponding to a dose of about 44 mrad/month of 2 MeV neutrons which would seriously interfere with low-level measurements over long periods of time (from the spontaneous fission point of view, ^{232}Th with a spontaneous fission half-life of $>10^{21}$ years is superior to ^{237}Np with $>10^{18}$ y, ^{235}U with $\sim 2 \times 10^{17}$ y, and ^{238}U with $\sim 6.5 \times 10^{15}$ y).

nitrate in acetone. As can also be seen in Figure 4, this reduces the sensitivity by about 20%, but also offers not much protection because the protective cover is subject to radiochemical damage and becomes brittle after a while with a tendency to peel off.

3. Sealing of the fissile material in a 2 μ m polycarbonate (Kimfol) foil. This is the best of the three methods resulting in a good protection and a tolerable reduction in the sensitivity of $20 \pm 5\%$ (Figure 4), as compared to direct contact with the unprotected fissionable material. If the coated surface is smooth, the danger of perforations in the protective foil is small, and little radiation damage is to be expected in polycarbonate.

As can also be seen in Figure 4, the sensitivity range of polymer-protected ^{237}Np combination covers well the range required for personnel dosimetry. The "background" spark count is zero, and 1 mrad (10 mrem) of fission neutrons result in ~ 6 spark counts. If necessary, the sensitivity can be further increased,

1. by a factor of two by sandwiching the Kimfol between two ^{237}Np foils;
2. by increasing the detector area; and
3. by another factor of about 2 by increasing the thickness of the ^{237}Np .

The precision and reproducibility of spark counts is usually about $\pm 10\%$, unless limited by the counting statistics. If a number of potential sources of error are eliminated and multiple counting of the same foil is done with great care, this error can be reduced to about $\pm 2-3\%$.

It has been shown previously that only little fading of the "latent" fission fragment tracks in polycarbonate foils takes place in dry air at temperatures not exceeding about 60°C . More recently, some experiments seemed to indicate an acceleration of fading in a humid environment: After 12 hours at 90°C , 20% fading occurred in dry air, but 45% fading at 80% relative humidity. Therefore, a fading test was carried out at 60°C and 77% relative humidity (Figure 5). 25% fading took place within two weeks, indicating that no significant fading should be expected at the temperatures encountered in personnel dosimetry even at high humidities.

In order to study the directional response of the detectors on the body, an elliptical plastic thorax phantom, filled with water, was equipped with detectors and their response was measured as a function of the orientation to a point neutron source. In Figures 6 and 7, the results obtained for 14 MeV neutrons and the HPRR fission spectrum are given. As expected, the directional response is much more pronounced for the fission spectrum. Due to the different low-energy cut-off energies for the two detectors, there is less directional dependence for ^{237}Np than for ^{232}Th . Varying the distance between the detector and the surface of the phantom had no significant effect on the sensitivity with one exception, namely the ^{237}Np detector on the back of the phantom exposed to fission neutrons. In this case, increasing the distance from 0 to 2 cm reduced the response by a factor of three.

There have been some special applications of fission fragment registration in personnel dosimetry, for example, in the hand monitoring of persons working with plutonium in glove boxes. The ring dosimeter (Figure 8) contains a thin ^{232}Th foil attached to the top, and the 10 μm Kimfol which is glued to a retainer ring and pressed gently against the Th by a lead disc acting also as a shield to reduce the gamma dose to the skin.

4. AMPLIFICATION OF DIRECT INTERACTION TRACKS IN POLYMERS

If a sensitive polymer is exposed to fast neutrons, numerous tracks of recoil nuclei, mainly carbon plus some alpha tracks from (n,α) reactions at higher neutron energies, can be revealed by extended etching. The technique is attractive because it avoids many of the disadvantages of fissionable materials, but has the disadvantage that recoil tracks are small, more difficult to count visually than fission fragment tracks, and do not lead to perforations in thin foils which can be spark-counted.⁽⁸⁾ It would, therefore, be highly desirable to enlarge these tracks in order to simplify their visual or automatic counting.

Among the various methods which have been advanced to enhance the etching speed, sensitivity, and/or the size of the etched region, one which was recently proposed by Tommasino⁽⁹⁾ appeared to be promising and has been studied in some

detail. It is based on the conductive energy loss in tracks at high frequencies in an electrical field of sufficient strength, resulting in local heating of the etchant.

The foil to be etched is placed between two plastic containers filled with 28% KOH in such a way that it electrically separates the two containers (Figure 9). Platinum electrodes are placed in the two etchant containers and a H.V. function generator, producing sinusoidal or square waveforms, is connected to the electrodes. Voltage and waveform are monitored with an oscilloscope. The field breaks down whenever the foil becomes perforated. This enhancement principle can, therefore, not be used for samples to be spark-counted.

Experiments have been carried out at 2000 V, and at kHz and 250 kHz (1 kHz is preferable). In the cellulose acetobutyrate backing of dosimeter films, recoil nuclei tracks could be enlarged by 3 hours of this treatment to such a size that they become visible with the unaided eye (Figure 10). Visual counting of such detectors can be done at a very low microscopic magnification (10 X) or, even simpler, by projecting them on the screen of a microfilm reader. By doing this, an area of several cm^2 can be scanned at low track densities within a few seconds without moving the detector film. It should be noted that this method can be used also for the precise evaluation of overexposed, fogged, faded, or otherwise unreadable Kodak NTA films after the photographic emulsion has been removed. One rad of fission neutrons results in 1.2×10^4 tracks/ cm^2 , or $\sim 3.6 \times 10^4$ tracks in 3 cm^2 . Even by measuring the optical density only, doses around 100 mrad can be detected. Further work on this technique and its potential use in other areas, for example, in the ORNL radon progeny personnel dosimeter for miners,⁽¹⁰⁾ is presently in progress.

Acknowledgments

Most of the work reported in this review has been carried out by various members of the Applied Dosimetry Research Group, in particular by Dr. R. B. Gammage, K. W. Crase and J. S. Cheka (TSEE), and M. Sohrabi and T. Clark (tracks).

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Figure Captions

- Figure 1. Fading of the Latent Image in Kodak Personal Neutron Monitoring Film, Type A
- Figure 2. TSEE Curves of Ceramic BeO Discs ($\sim 1 \text{ cm}^2$) Exposed to 100 mrad 14 MeV Neutrons, if Covered with Polyethylene and with Teflon
- Figure 3. Neutron Energy Dependence of the Fast Neutron Response of Ceramic BeO TSEE Detectors which are covered with Polyethylene
- Figure 4. Spark Count of 10 μm Kimfol, Etched for Two Hours at 60°C in 28% KOH, as a Function of HPRR Fission Neutron Dose in rad ($1 \text{ rad} = 4 \times 10^8 \text{ n/cm}^2$), for Uncovered ^{237}Np and ^{232}Th , and Different Protective Covers Between Radiator and Sensitive Foil
- Figure 5. Fading of Latent Fission Fragment Tracks in Kimfol at 60°C and 77% Relative Humidity
- Figure 6. Directional Response of Detectors Attached to a Water-Filled Thorax Phantom for 14 MeV Neutrons
- Figure 7. Directional Response for HPRR Fission Neutrons
- Figure 8. ORNL Fast Neutron Finger-Ring Dosimeter (on Hand and Open, Showing ^{232}Th Foil in Cover, Kimfol on Retainer Ring, and Lead Disc in Base)
- Figure 9. Cell for High-Frequency Electrochemical Etching
- Figure 10. Recoil Nuclei Tracks in Fast Neutron Exposed Cellulose Acetobutyrate, Enhanced by Electrochemical Etching

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5

Fig. 8

Fig. 9

Fig. 10

PERSONNEL NEUTRON DOSIMETRY LAWRENCE LIVERMORE LABORATORY EFFORT*

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OPERATIONAL PROGRAM

LLL has approximately 5,000 employees with an additional 1,000 permanently badged personnel. Neutron producing facilities include a reactor, a 14 MeV ICT, a 100 MeV Linac, and heavy element handling areas in addition to other miscellaneous activities.

All personnel monitoring is done with TLD dosimeters which are processed using an automatic reader. A TLD-100 reading is compared with TLD-700 to detect neutron exposures, with investigation and evaluation by the health physicist following any significant readings. The Anderson-Braun neutron remeter and the Victoreen Model 400 survey meter are used in the field to establish neutron/gamma ratios. An LLL design activation foil type Nuclear Accident Dosimeter (NAD), similar to the SRLCND, was issued to 120 employees in 1970. It replaces the commercial system that had been used previously.

CURRENT DEVELOPMENT

Within the last two years, we have increased our dosimetry research and development effort significantly. This has been primarily due to construction of a new calibration facility, which features a large, low-scatter irradiation room, and the acquisition of a 10^9 n/sec ^{252}Cf source which can be used with a number of moderating shields to give us a wide variety of neutron spectra.

The areas of investigation currently include a TLD albedo dosimeter, track registration techniques, TSEE, and NAD badge improvements.

* Work performed under the auspices of the U.S. Atomic Energy Commission.

TLD DOSIMETER

The TLD albedo dosimeter consists of the standard 3-chip LLL dosimetry packet covered on the back and edge by .02" cadmium and on the front by .062" borated plastic enriched in ^{10}B . Directly over one of the TLD-600's a .312" diameter hole has been cut in the borated plastic and this hole is covered with a .02" Cd button. The packet, therefore consists of one TLD-600 covered by .02" Cd, one TLD-600 covered by borated plastic and one TLD-700 for gamma exposure correction covered by borated plastic. All three chips are shielded from the body by Cd. This was done to enhance the higher energy albedo relative to the thermal albedo.

The boron-shielded TLD effectively sees those incident neutrons with energies greater than about 10 eV. The Cd-shielded TLD effectively sees those incident neutrons with energies greater than .3 eV. Both TLD's see the same albedo spectrum. Thus, some spectrum discrimination can be achieved by observing the difference in response of the two chips (energy between .3 eV and 400 eV) relative to the boron shielded chip.

The badge was initially calibrated to a free-air spectrum of ^{252}Cf . The badge was then exposed to various calibrated spectra including free air, scattered, and moderation provided by polyethylene, water, steel, and graphite. Over-responses ranged from 1.5 to 2.6 with most grouped in the region between 1.5 and 2.3. If a new calibration point is taken among this grouped data, then the new correction factor is 780 TL units per Rem versus 459 units with a bare source calibration. This means that if no consideration is given for spectral corrections all the exposures tested with the exception of nonscattered spectra would be within 40% of the average. The bare unscattered response would be 50% low. This indicates that this badge may have a flat response for all spectra normally encountered in our facility.

The minimum detectable neutron dose is, of course, a function of the gamma dose. This relationship is actually

$$MDD = 3 D_{\gamma}$$

MDD is the minimum detectable neutron dose

D_{γ} is the observed gamma dose

For example, a gamma dose of 400 mrem would yield a MDD of 60 mrem.

An advantage of using Cd backing is that the badge is not strongly dependent on body albedo. This means that the badge need not be held against the body although it is certainly more desirable to do so.

Work is continuing on slight modifications of the badge for increasing sensitivity and wearability. When these tasks are completed, field tests will begin.

TRACK REGISTRATION

The track registration work includes use of cellulose nitrate for registration of alphas from the $^{10}\text{B}(\eta, \alpha)$ ^7Li or $^6\text{Li}(\eta, \alpha)$ ^3H reactions or use of fission fragment tracks from a fission foil.

The use of cellulose nitrate for registration of lithium or boron produced alphas is being investigated as a personnel dosimeter. Initial experiments with spark counting alpha tracks in thin films of cellulose nitrate have been unsatisfactory, primarily because of the high background that results when the films are etched long enough to detect the 1.6 MeV $^{10}\text{B}(\eta, \alpha)$ or 2.0 MeV $^6\text{Li}(\eta, \alpha)$ alphas. However, Eastman Kodak is producing a colored cellulose nitrate on an experimental basis. The 8-micron layer of deep red cellulose nitrate is supported by a plastic base. When exposed to even weak alphas and etched, the tracks appear as very distinct pinholes in the red background, particularly when illuminated with green light. The tracks are about 10 microns in diameter. In exposures on a phantom using a TLD-600 chip as a radiator, we found track densities in excess of $10 \text{ cm}^{-2} \text{ mrem}^{-1}$, even for a bare ^{252}Cf spectrum. Exposed to a highly thermalized spectrum, the density was more than an order of magnitude greater. When exposed without radiator to a 14 MeV spectrum, the track density was about $75 \text{ cm}^{-2} \text{ Rem}^{-1}$. We are now investigating automatic track counting and sizing techniques.

ENVIRONMENTAL MONITORS

In the search for a long-term, low-level, neutron environmental area monitor, we have placed a thin foil of ^{235}U with a registration plastic in an Anderson-Braun remeter moderator. The polycarbonate film is etched and the track density counted on a spark counter. Using a 3 cm^2 foil, with 5.5 mgm/cm^2 ^{235}U , we have about 15 counts/mrem for .5 mil film and 50 counts/mrem for .25 mil film, both with a background of essentially zero. This implies a detection level approaching 0.1 mrem. We are beginning calibration and long-term response studies on 8 units for laboratory perimeter survey.

TSEE

The TSEE study has just begun, with reader research currently in progress.

NUCLEAR ACCIDENT DOSIMETRY

As a result of computer studies during the past year, we will be issuing a modified NAD badge, with both indium and copper foils enclosed in borated plastic rather than cadmium.. This reduces the effect of low-energy neutrons (1-100eV) and makes the copper foil act more like a threshold detector. It also suppresses the $^{116\text{m}}\text{In}$ activity in the indium foil, allowing early analysis of the $^{115\text{m}}\text{In}$ threshold detector activity.

PERSONNEL NEUTRON DOSIMETRY

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TLD Dosimetry Reader

Although the TLD reader development at the Chalk River Nuclear Laboratories (CRNL) is for use in γ and β dosimetry, the detail of the TLD heating mechanism is reported here because of the possible interest to those using TLD for albedo neutron dosimeters.

Unlike most other TLD readers in which the temperature of the heating element itself is controlled, in the CRNL reader the temperature of the upper surface of a TLD chip is monitored with a small diameter thermocouple which lies on it while a heating anvil is raised from below to come in contact with the chip (supported on a heat resistant plastic film). In this way a reproducible heat treatment is achieved and there is no concern over the constancy of the quality of the thermal contact between the chip and the heater.

Two chips (1/8" square) are mounted on a aluminum plaque, the size of a dosimeter film package.

One chip (0.035" thick) is located between two aluminum plates (0.080" thick) in the existing film badge and is to be used to determine penetrating radiation dose and the other (0.015" thick) is obscured only by an opaque paper and is to be used to estimate total skin dose from β and γ radiation.

The automatic reader will handle 200 dosimeters per batch and units are identified by a teletype code stamped in the aluminum plaques.

Experimental trials are proceeding and full scale use is expected by mid-1972. Full details will be reported by A. R. Jones at the Third International Conference on Luminescence Dosimetry in Riso, Denmark, in October 1971.

Use of $^{103}\text{Rh}(n,n')^{103\text{m}}\text{Rh}$

Details were described in presentation to the Health Physics Society in Chicago, 1970 (H. Ing and W. G. Cross).

This detector is useful only for accident dosimetry because of the short half life of the isomeric state (57 min) which is the main defect even for accident work. However for doses exceeding 60 rad there is a 10 hour period in which to recover the dosimeter and estimate its activity. The estimate may be made within 1 hour of an incident unlike most other activation dosimeters.

The performance realized in one ORNL and two IAEA intercomparison studies has been very satisfactory with the results from Rh activation alone as accurate as we can derive from any combination of activation or damage track dosimeters.

However because of the short half life of the product activity, other devices must be available in case the dosimeter recovery is delayed.

Damage Track Detectors

No major advances have been made since the report to the 1969 Health Physics Society meeting. Developments in details include the following:

- (a) Etching temperature has been reduced from 60° to 40° for 3 hours as a convenience since it is easier to control the process at the lower temperature.
- (b) Main effort has been directed towards improvement of reproducibility with the following results:
 - (i) < 1% reproducibility for repeated counts on the same Makrofol film
 - (ii) $\pm$ 2% spread (in addition to statistical fluctuations) between different foils irradiated with the same neutron dose.

These improvements were realized after

- (i) The introduction of pulse shaping circuit

between discharge and the scaler. This enables a higher setting of the scaler discriminator and excludes spurious pulses.

- (ii) Careful control of pressure holding aluminized Mylar and Makrofol together while sparking.
- (iii) Introduction of several high voltage discharges followed by quick rinse of Makrofol in dilute KOH to remove aluminum debris.
- (iv) High voltage discharges done with two sizes of electrodes and increasing voltage to avoid isolation is necessary only if track density exceeds $300/\text{cm}^2$. (At this track density accuracy is no longer limited by statistics so the procedure is a worthwhile improvement.) In routine estimation of small doses this treatment is not required.

- (c) Techniques for handling 11 x 15 mm foils have been developed. This is not the ideal size but it matches a receptacle in the existing film badge holder. Small scale issue of Makrofol-Thorium combinations has begun.

Nether Th nor Np foils (as alloy with Al) interferes seriously with other gamma sensitive dosimeters in badge. A constant background of about 10 mrem in two weeks is easily subtracted.

Attempts to Use Neptunium

Developments of a 1% Np in Al alloy appears to be successful. The contamination on swipe is about the same as that from Th or U metal. The alloy presently available has a small concentration of a thermally fissile element (probably ~ 100 ppm of ^{235}U) and gave readings about a factor of 2 high in tests on the HPRR at ORNL.

Lithium-6 Shields

Shields of ^6LiF have been pressed from powder in a simple pill-box shape to provide protection against exposure to the thermal neutrons for dosimeters to be used to estimate gamma radiation doses. A filter of Sn (0.006" thickness) on the outside of the shield improves the gamma radiation response below 100 keV.

Publications

- (a) Alternatives to Nuclear Track Emulsions for Fast Neutron Personal Dosimeters, W. G. Cross, Health Physics Society New York, 1971.
- (b) Use of the $^{103}\text{Rh}(n,n')^{103m}\text{Rh}$ Reaction for Personal Neutron Dosimetry, H. Ing and W. G. Cross, Health Physics Society, Chicago, 1970.
- (c) Neutron Spectra in Criticality Accidents, W. G. Cross and H. Ing, Health Physics Society, New York, 1971.
- (d) The Spark Replica Technique for Measuring Neutron Doses with Damage Track Dosimeters, W. G. Cross and L. Tommasino, Health Physics Society, Pittsburgh, 1969.
- (d) A Rapid Reading Technique for Nuclear Particle Damage Tracks in Insulating Foils, W. G. Cross, Intern. Topical Confer. on Nuclear Track Registration for Insulating Solids and Applications, Clermont-Ferrand, France, May 1969.

SUMMARY OF PERSONNEL NEUTRON DOSIMETRY INVESTIGATIONS AT BNL

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I. INTRODUCTION

Improved personnel neutron dosimetry involves both human and physical factors. Present technology employs an NTA system which suffers from energy dependence and human reader variability. Additionally, the neutron observations are fogged by gamma background masking neutron exposure with moderate to high gamma exposure.

Three different systems were investigated. They are:

A. Albedo Techniques Employing TLD Materials. Albedo techniques were considered in a model badge configuration. Ensuing investigation proved a large variation in response with badge-to-person separation for a constant exposure field. This problem could be reduced by employing a belt carrier. However, the processing problems of routinely changing large numbers of belt badges was discouraging. Further, the ideal dosimeter location is at head height due to beam height at the Alternating Gradient Synchrotron (AGS). Since about 80% of the neutron exposures occur at the AGS, a belt badge location would not properly report a head exposure.

B. TSEE Methods. A system employing thermally stimulated exo-electrons emission (TSEE) is in the earliest stages of consideration. At this time a reader has been constructed and is under adjustment for acceptable response. The exo-electrons emanate from a dielectric substance about 50 Angstroms thick when thermally stimulated. A Be oxide material commercially known as Thermolox 995 by Brush Beryllium Company will be used together with (N-P) polyethylene and (N- α) ^6Li , ^{10}B radiators. This system is a subtractive method since the trapped exo-electrons are contributed by both the radiator and the presence of an external gamma field.

C. Fission Pit Dosimetry. The fission pit method of detecting neutrons has the principle advantage of an acceptable neutron sensitivity with low background. The neutron sensitivity directly relates to known cross section allowing a straightforward calculation of dose for any given spectrum. The method is a nonsubtractive method allowing the monitoring of neutron exposure in the presence of any gamma exposure. Photo-fission is a competing reaction. However, the cross section is trivial for energies below about 8MeV. Additionally, the accelerator produced gamma component of the exposure is limited to 50% of the total exposure. The energy spectrum is expected to be below 8MeV.

II. PRESENT RESULTS

A number of investigations have been conducted to outline the parameters of the fission pit system. The following is a listing of some of the parameters investigated:

A. Development of a spark counting system. Appendix 1

B. Response of different plastics to fission fragments. 40 gauge Kimfol is now being used.

C. Variation in etching temperature-time was investigated and 75 minute etching in 6N KOH at 65°C was found to offer the best signal to background ratio.

D. The effect of sparking anode weight on both sensitivity and background. 100gm/4.46cm² stainless steel anode has produced consistent results. The cathode is 40 gauge aluminized Kimfol.

E. Background characteristics of Kimfol, both in contact with, and separated from a fission radiator were studied. The spontaneous fission rate of ²³⁸U produced a 5 pit/day background within a 4.46cm² area. The Thorium background fission rate is about .25 pits/4.46cm²/day and the non-time dependent foil background averages about 1 pit/4.46cm².

F. The sensitivity of ²³²Th and ²³⁸U were compared empirically and mathematically. The ²³²Th is calculated to be 0.286 times as sensitive as the ²³⁸U for an AmBe spectrum. Experimentally the sensitivities are

$$^{238}\text{U} = \frac{2.3 \pm 0.2 \text{ mrem}}{\text{pit}/4.46\text{cm}^2}, \quad \text{Th} = \frac{6.9 \pm 0.6 \text{ mrem}}{\text{pit}/4.46\text{cm}^2}.$$

G. The fission pit neutron detection method has already been used as an area monitor. Appendix 2. Eleven widely different locations were monitored within the AGS experimental area. In each case the exposure was characterized by a number of other detectors and the fission pit method favorably compared. The comparison was made by assuming that the neutron spectrum followed a $1/E$ dependency with spectra limits taken as 10^{-8} to 10^2 MeV. Appendix 3. The fission pit response to a $1/E$ spectrum was calculated by following the fission cross section with energy for an integral neutron fluence.

This was compared with the calculated fission pit response to an AmBe spectrum of the same integral fluence. The above ratio was normalized to a measured system response to a laboratory AmBe exposure and adjusted to the dose equivalent for a $1/E$ spectrum by use of a spectrally weighted fluence to dose conversion. Measurement of the thermal neutron fluence can also characterize the dose due to a $1/E$ spectrum by use of a calculated integral dose equivalent to thermal fluence ratio. The absolute thermal neutron response of TLD 600 was determined at the BNL Sigma pile. The calibrated TLD dosimeters were then used to characterize the dose of each of the eleven Accelerator positions. Reasonable agreement exists between the fission pit monitor reporting the spectral dose and the thermal neutron monitor. This demonstrates the validity of the $1/E$ assumption and suggests the development of a two-component neutron badge containing a thermal neutron detector and a fission pit system.

III. FUTURE DIRECTION

Future effort will be divided into two phases. The first involves radioactivity reduction and the second final product assembly. ^{232}Th , ^{238}U and ^{237}Np are all useful fission radiators but each suffers from radioactivity. ^{232}Th is the least toxic of the three. Calculations indicate that an unshielded Th foil of $4\text{mg}/\text{cm}^2$ containing 18mg of ^{232}Th would produce a gamma dose rate of 0.48mRad/hr at 2cm in tissue. The 4mg foil is nearly infinitely thick for fission fragments. The fission pit system sensitivity versus foil thickness will be investigated. Preparation of thin foils by coating-firing techniques

and electroplating will be considered. Additionally, shielding will be studied to further reduce the gamma radiation from the radioactive decay of ^{232}Th and its daughters. When an acceptable shield and foil combination is reached construction of a production reading system and badge holder will be started.

IV. SUMMARY

1. A personnel neutron dosimeter has been investigated and has the following characteristics:

- a. Known and acceptable neutron response
- b. Little or no fading, allowing quarterly, semi-annual and annual reading for low risk people
- c. Simple processing
- d. Very inexpensive
- e. Much faster and easier to read than NTA film.

2. Future investigations are directed toward the reduction of radioactivity by the development of thin foils and shielding.

3. A badge will be developed and appropriate semi-automatic production read-out equipment will be designed and constructed.

BNL SPARK COUNTER

APPENDIX I

APPENDIX 2

LOCATION	FILM mR	NTA FILM mrem	MBLE CaF ₂ mR	U PITS 1.9mrem/tr mrem	ALBEDO* BADGE mrem	12" BONNER SPHERE DE mrem
1. Tgt. Desk	150	60	73	135	142	45
2. SEB Cross walk	215	2770	159	1830	690	820
3. 922 Irma	50	200	34	214	138	-
4. P. S. East Hyperon	205	1130	96	595	690	285
5. Collins- Argo	20	33	24	15	15	11
6. Col-Christ	75	380	55	95	173	91
7. Rutgers- Test	80	310	54	173	142	97
8. C-8 Col- Brdge.	295	920	151	940	1250	580
9. HO°Bm Shield	170	270	68	376	490	210
10. Prince- Fitch	80	435	50	210	123	82
11. Muon Trl.	50	10	37	57	23	9

Character of Exposure Location

1. Elevated room detached (150') from the experimental area. Skyshine
2. On top of beam stop. Transmission through shielding.
3. 4' above concrete floor in an assembly building remote (300') from the center of the experimental floor. Skyshine
4. Power supply catwalk about 10' above experimental floor and 20' east of the Hyperon cave. Transmission through thin shielding
- 5, 6 and 7. Open experimental area. Skyshine and scatter from experimental equipment.
8. On platform above roofless secondary beam cave. Source is scatter from underlaying beam lines and skyshine.
- 9, 10. Near experimental trailers in different experimental area. Skyshine is expected source.
11. Near an experimental trailer rich in muons.

* Dose Equivalent due to 1/E spectrum deduced from the (Li(N- α)) thermal neutron end point corrected for the entire spectrum $\left[\frac{0.22m43m}{mR} \right]$.

APPENDIX 3

Consider the expected pit response to 1/E spectrum

$$(P)_{1/E} = \left[\int (\phi_i \sigma_i)_{1/E} dE \right] (N \text{ eff}) \Phi_{1/E}$$

$$\text{where } \int \phi_i dE = 1 \text{ N/cm}^2$$

$$\Phi_{1/E} = \text{Field Integ. flux}$$

$$(N \text{ eff}) = \text{Target sized counting eff.}$$

Both (N eff) and $\Phi_{1/E}$ unknown

$$(P)_{\text{AmBe}} = \left[\int (\phi_i \sigma_i)_{\text{AmBe}} dE \right] (N \text{ eff}) \Phi_{\text{AmBe}}$$

$$(N \text{ eff}) = \frac{(P)_{\text{AmBe}}}{\left[\int (\phi_i \sigma_i)_{\text{AmBe}} dE \right] \Phi_{\text{AmBe}}}$$

substituting

$$\Phi_{1/E} = \frac{(P)_{1/E} \left[\int (\phi_i \sigma_i)_{\text{AmBe}} dE \right] \Phi_{\text{AmBe}}}{\left[\int (\phi_i \sigma_i)_{1/E} dE \right] (P)_{\text{AmBe}}}$$

or

$$\Phi_{1/E} = (P)_{1/E} \left[\frac{\int (\phi_i \sigma_i)_{\text{AmBe}} dE}{\int (\phi_i \sigma_i)_{1/E} dE} \right] \left[\frac{\Phi_{\text{AmBe}}}{(P)_{\text{AmBe}}} \right]$$

$$DE = \Phi_{1/E} \left(\frac{DE_{1/E}}{\text{unit flux}} \right)$$

APPENDIX 3 (continued)

where

$$DE_{1/E} = \int (\phi_i DE_i)_{1/E} dE$$

$$DE = (P)_{1/E} \left[\frac{\int (\phi_i \sigma_i)_{AmBe} dE}{\int (\phi_i \sigma_i)_{1/E} dE} \right] \left[\frac{\Phi_{AmBe}}{(P)_{AmBe}} \right] \left[\int (\phi_i DE_i)_{1/E} dE \right]$$

$$DE = (P)_{1/E} \frac{2.09 \times \frac{5.78 \times 10^6 N/cm^2}{18.6 pit/cm^2} \times 1.29 \times 10^{-5} \frac{mrem}{N/cm^2}}{}$$

$$\frac{DE}{(P)_{1/E}} = \frac{8.41 mrem}{pit/cm^2}$$

Sparker Area 4.46 cm²

$$\frac{DE}{(P)_{1/E}} = \frac{8.41}{4.46} = \frac{1.9 mrem}{pit/cm^2}$$

The Hanford Thermoluminescent Multipurpose Dosimeter

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The Hanford Thermoluminescent Multipurpose Dosimeter is a five element dosimeter using LiF blocks designed to measure personnel exposure to beta particles, photons and neutrons. One ^7LiF block is shielded to provide interpretation of the one centimeter tissue depth dose. A second ^7LiF block is unshielded to provide a derma dose interpretation. Two ^6LiF and one ^7LiF blocks are used for fast and thermal neutron dose interpretation.

The LiF blocks are enclosed in a teflon sandwich placed in a plastic card. A special holder with metallic and plastic filters helps provide the necessary response of the dosimeter to the various types of radiations.

The development and testing program which includes a fully automated computer controlled reader has shown that the multipurpose dosimeter is a significant improvement over the film badge presently in use.

The dosimeter is able to detect 300 mrad of beta-photon radiation with ± 5 percent at the 67 percent confidence level. The lower level detection limit is about 20 mrad ± 50 percent at the 67 percent confidence for penetrating radiation.

For fast neutrons the dosimeter makes use of the backscattering of thermalized neutrons from the human body. This system works well when properly calibrated for most neutron exposure conditions at the Hanford Plant. The lower detection limit for fast neutrons produced by a PuF_4 source is 5 mrad ± 50 percent. For thermal neutrons the detection limit is about 0.5 mrad ± 50 percent. A correction for backscattered incident thermal neutrons makes the dosimeter useful even in exposure geometrics involving moderated fission sources such as ^{252}Cf and plutonium fuels.

NEUTRON DOSIMETRY AT ROCKY FLATS

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INTRODUCTION

At the Rocky Flats Plant, neutron dosimeters are required for personnel engaged in glove-box operations. Various thicknesses and configurations of neutron shielding used on glove boxes result in a variety of neutron spectra. These spectra range from unmoderated to heavily moderated (four inches of lucite) fluences from plutonium spontaneous fission and plutonium (α, n) reactions on various light elements.

Since January, 1971, we have been using a simple albedo neutron dosimetry system, using lithium fluoride thermoluminescent dosimeters (TLD), as a routine personnel neutron dosimeter. Description and calibrations of the system are presented in the report RFP-1581, entitled "A Personnel Neutron Dosimeter Using Lithium Fluoride Thermoluminescent Dosimeters." Briefly, the system consists of two paired sets of TLD-600 and TLD-700 LiF ribbons, obtained commercially from The Harshaw Chemical Company, Cleveland, Ohio. One pair is shielded from the front by 1/64" thick cadmium, and the other pair is shielded from the back by the cadmium. The total system is contained in a plastic holder which can fit in the cavity in our dosimetry badge, which originally held NTA neutron film. The dosimetry badge is then secured to the chest of the wearer by a special pocket in his coverall.

THEORY

The theory for this system is presented in RFP-1591. The calculation of the dose equivalent for the Rocky Flats' system is given by

$$\text{Dose Equivalent} = C \times [(FCd) - f \times (BCd)]$$

where C is a calibration factor in units of mrem per count,

F is a factor determined by calibration to optimize the system response as a function of moderation,

FCd is the net neutron response of the dosimeter pair shielded from the front by cadmium, and

BCd is the net neutron response of the dosimeter pair shielded from the back by cadmium.

The net neutron response of the pair is obtained by subtraction of the response of the TLD-700 dosimeter from that of the TLD-600 dosimeter after adjusting the TLD-700 response to allow for any difference in sensitivity to gamma radiations.

Briefly and very simply, the rationale for this method of obtaining the dose equivalent is that the TLD-600 dosimeter shielded, from the front by cadmium responds to moderated and reflected fast and intermediate and reflected thermal neutrons and to incident fast and intermediate neutrons. The expression $[(FCd) - f \times (BCd)]$ then represents a net system response consisting of a response to $(1 - f)$ of incident fast and intermediate neutrons plus (reflected thermals - $f \times$ incident thermals). If this last term is considered to be zero, then the net system response results from neutrons of non-thermal incident energy. It should be emphasized, however, that even though this may be a good method, the purpose for the practical health physicist is to find a useful correlation of dosimeter response with the dose equivalent received by the personnel being monitored by the dosimeter, and other methods of obtaining the dose equivalent may also be just as good.

DOSIMETER CALIBRATION AND PERFORMANCE

Initially, we experimented with a neutron system using three pairs of dosimeters, the third pair being shielded both front and back by cadmium. Even though this resulted in an improved theory, we found no significant improvement in accuracy for the three pair system over the two pair system. The net response of the FCd pair is the crucial one for dosimetry with the purpose of the BCd pair being to adjust for the thermal component in the incident fluence.

The method and procedure in calibrating the system is also described in detail in RFP-1581. In general, the approach in calibration is to determine the response of the system response to the total spectrum for the sources and range of moderation which the system will encounter in routine operations.

At Rocky Flats we have used a PuF_4 source with a set of spherical polyethylene moderators as our primary calibration source. We also have recently used ^{252}Cf and PuBe sources with a set of cylindrical polyethylene moderators to explore the system response to these spectra. In all calibrations the dosimeter is attached to the chest of a water-filled REMAB phantom. Preliminary results of these calibrations are presented in Figures 1, 2 and 3, which show the response (counts per mrem) versus thickness of polyethylene moderation for the dosimeter pair shielded from the front with cadmium (designated FCd), for the dosimeter pair shielded from the back with cadmium (BCd), and also for the third dosimeter pair shielded front and back with cadmium (2Cd), respectively. The significance of these results is that the response of the system is similar for PuF_4 and ^{252}Cf sources so that the system can be used for mixtures of low energy (α, n) sources and fission sources. However, a separate calibration is needed for neutron spectra from higher energy (α, n) sources, such as from PuBe, and inaccurate results may occur from use of this system in mixtures of spectra from PuBe and from lower energy sources. A new design of the Rocky Flats' personnel dosimeter has provision for a dual NTA film - TLD neutron system as a possible approach to provide more accurate dosimetry coverage for exposure situations which may involve such a range of neutron sources and spectra.

Table 1 gives a summary of the total system response as a function of polyethylene moderator thickness for the PuF_4 , ^{252}Cf , and PuBe sources using a value of $F = 0.39$. Also presented are values for the system dose indication for a given dose of 100 mrem using a calibration factor of 1.00 mrem per count.

TABLE 1. Summary of Total System Response as a Function of Polyethylene Moderator Thickness for $f = 0.39$

Thickness (cm)	System Counts per mrem			TLD Dose Indication for 100 mrem Given Dose		
	PuF ₄	²⁵² Cf	PuBe	PuF ₄	²⁵² Cf	PuBe
0	0.47	0.55	0.25	47	55	25
1	0.73	0.87	0.35	73	87	35
2	0.91	1.06	0.43	91	106	43
3	1.04	1.14	0.47	104	114	47
4	1.09	1.13	0.48	109	113	48
5	1.07	1.07	0.46	107	107	46
6	1.04	1.00	0.43	104	100	43
7	1.03	0.94	0.41	103	94	41
8	1.02	0.89	0.42	102	89	42
9	0.99	0.85	0.44	99	85	44
10	0.97	0.80	0.47	97	80	47

It is seen that the system is about half as sensitive to neutron spectra from PuBe as to spectra from PuF₄ or ²⁵²Cf. The system response is also independent of the amount of moderation within less than $\pm 15\%$ for a range of less than two centimeters to almost ten centimeters for the three sources. One possible way to adjust for the under response for unmoderated spectra is to note the ratio of the BCd to FCd responses and use this ratio as an index of the amount of moderation. An index of about 0.5 would indicate an unmoderated spectrum, for which a different calibration factor could be used.

Consideration of angle of incidence, as presented in RFP-1581, for the PuF₄ source indicated no significant effect on the system response, assuming either a line source or isotropic incidence through 180°. Preliminary results of calibrations using the PuBe source indicate a similar conclusion for that source.

Results of field tests, presented in RFP-1581, indicated an adequate performance under a wide range of source and shielding configurations. Table 2 presents results of one series of measurements for a given neutron dose of 20 mrem in the presence of 0 to 400 mR gamma from a ^{137}Cs source.

TABLE 2. TLD Neutron Dose Indication for a Given Neutron Dose of 20 mrem from a ^{252}Cf Source (moderated by 7.0 centimeters of polyethylene) in the Presence of 0 to 400 mR Gamma Dose

<u>Gamma Dose</u>	<u>Given Neutron Dose</u>	<u>TLD Neutron Dose</u>
0	20	22
0	20	22
0	20	21
0	20	19
50	20	29
50	20	35
50	20	30
50	20	40
100	20	21
100	20	33
100	20	43
100	20	29
200	20	23
200	20	28
200	20	16
200	20	28
400	20	32
400	20	23
400	20	39
400	20	39

USES AND LIMITATIONS

The usefulness of this type of dosimeter depends upon the user's ability to calibrate his holder and TL dosimeters to the sources and range of moderation which would be encountered. Significant differences in both neutron and gamma sensitivity can occur between different batches

for both TLD-600 and TLD-700 dosimeters. Batch-to-batch differences in sensitivity mean that calibrations of each batch of dosimeters must be made. Once the user knows the gamma and neutron sensitivities of his batches of dosimeters, it is necessary for him to determine the response characteristics of the system in his particular holder and dosimetry badge. At Rocky Flats we are in the process of converting to a new dosimetry badge for which the cadmium and plastic thicknesses are essentially the same as our initial TLD badge. Yet the response characteristics of the system in the new badge, especially the FCd response, are somewhat different than that for the old badge to the degree that the overall sensitivity of the new badge is generally 30% greater than that for the old badge, using the same batch of dosimeters.

Our studies indicate that this simple dosimeter is useful for moderated spectra from plutonium fluoride and fission sources or combinations of the two. The system is also useful for moderated spectra from sources of higher energy neutrons, such as PuBe sources, although the usefulness is limited for mixtures of spectra from PuBe with that from PuF_4 or fission.

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The study of personnel neutron dosimeters at the Los Alamos Scientific Laboratory during the past two years consisted of albedo neutron dosimetry and fission fragment damage to polycarbonates using the spark counter technique.

The albedo neutron dosimetry study was designed to determine the effect on the energy dependence and sensitivity of the dosimeter when polyethylene is included in the dosimeter. A series of dosimeters were prepared which had polyethylene thicknesses of 0, 1/8, 1/4, 1/2, and 3/4 inches. The thermal neutrons were detected by pairs of ^6Li and ^7Li TLD's. To determine the best location to place the TLD's in or on the dosimeter, data were obtained and curves drawn for the positions shown in Diagram I. The two dosimeters in the diagram are identical except that the lower dosimeter has no cadmium on the bottom of the dosimeter next to the wearer. Positions 3, 4, and 5 are the same as positions 7, 8, and 9 but because cadmium is placed on the bottom of the upper dosimeter in the diagram, the results obtained at these positions will be different and were assigned different numbers.

The results indicate that there are no significant changes in the energy dependence of an albedo neutron dosimeter resulting from the addition of polyethylene to the dosimeter. There is, however, an increase in the response of the TLD's located at several of the positions in the dosimeter. The albedo dosimeters are very energy dependent and the series of curves we obtained do not indicate that correction, based on a pair of dosimeters, can be used to correct for the energy dependence. It would appear at the present time that albedo dosimeters can be used only with neutron spectra having limited variations in the energy spectra.

The results and curves obtained in this study are being prepared into a LAMS report.

The spark counter technique gave us considerable difficulty in obtaining reproducible results until it was decided to spend additional time evaluating the system. Several things contributed to the poor reproducibility. These included an ~10% change in the number of sparks depending on the orientation of the Kimfol (that is, whether the shiny or dull side of the Kimfol is facing the source during the calibration procedure).

When an etching solution is used repeatedly, a drop in the number of sparks of 4% for the second and 6% for the third etching is found. The stirrer speed must remain constant and the sparking procedure we were using proved to be inadequate. Previously recognized variables such as etching temperature, time, solution concentration, and weight placed on the foil during sparking are important. The new sparking technique consists of a 5 minute precount spark routine at 600 volts for each 500 sparks (with a minimum of three spark counts at 600 volts) followed by two sparkings at 500 volts. The final two spark counts are averaged. The standard deviation of the spark counter technique is now >2.0%. Our procedure in the future will include the etchings of three standard foils with each set of foils being etched. This system will be calibrated in the near future and the study of personnel dosimetry will be continued.

THERMOLUMINESCENT NEUTRON DOSIMETRY USING ^6LiF AND ^7LiF EXTRUDED RIBBONS*

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During the first AEC Workshop on Personnel Neutron Dosimetry,⁽¹⁾ an albedo-type TLND badge was discussed; this badge appeared promising except for its large physical size and the inability to physically maintain a constant geometry between the badge and wearer's body. Since our last meeting both of these problems have been solved by incorporating both hemispheres into a single unit and by providing a means of attaching the badge to the wearer's belt. Although the dosimeter is directionally sensitive and exhibits energy dependence when exposed to unscattered and monoenergetic spectra, its overall accuracy and precision for continuous spectra are better than other types of personnel monitors presently available.

THE TLND BADGE

A "belt" mounted thermoluminescent neutron dosimeter (TLND) was designed and evaluated for personnel monitoring at the Savannah River Plant. The badge (shown in Figures 1, 2, and 3) consists of a 2-inch polyethylene hemisphere that has been sliced one-fourth inch from the rounded end. One TLD-600 (1/8 inch $\times$ 1/8 inch $\times$ 0.025 inch) and one TLD-700 (1/8 inch $\times$ 1/8 inch $\times$ 0.035 inch) extruded ribbon constitute a detector pair. One detector pair is placed in the polyethylene dome section, and one detector pair is placed in the flat section next to the rear plate. The TLD pair in the dome section is completely shielded by cadmium. The curved surface of the hemisphere is covered with 1/32-inch-thick cadmium except for a 1/2-inch-diameter section of the dome that has 0.003-inch-thick cadmium. The entire dome is covered with an outer protective shell of 20 gauge stainless steel. A stainless steel back plate holds the unit together

* The information contained in this article was developed during the course of work under Contract AT (07-2)-1 with the U.S. Atomic Energy Commission.

and provides the means of attachment to a belt. A small 1/8-inch-diameter hole in the rim of the badge is provided for locking the unit together to discourage disassembly by field personnel.

The badge, when mounted against a body, becomes a dosimeter. The purpose of the belt is to maintain a close fit between the badge and the body. An air space of one-fourth inch reduces the response by 50%. All calibration studies have included the thickness of the belt in the derivation of response information by attaching the dosimeter to the midsection of a six gallon jug filled with water. Normal calibration is for a front exposure with distance being measured from the source center to body front. Directional sensitivity is poor but about the same as observed for NTA film.

The lower limit of sensitivity of the badge is dependent on TLD material, annealing procedures, and associated gamma activity during the neutron exposure. In the presence of 1000 mR of gamma activity, 10 mrem can be detected with a $\pm 160\%$ precision at 1σ . Without significant gamma, 5 mrem can be detected with 1σ of $\pm 4\%$.

The highest dose equivalent measured was 46,000 rem and was within 10% or the estimated dose equivalent by Hankins⁽²⁾ at LASL. No rate dependence was observed up to 10^7 rem/sec.

DERIVATION OF THE DOSE EQUIVALENT

The determination of energy response of the TLND requires the exposure of the TLND to neutron spectra of different energy distributions. The response must be calibrated against a reproducible standard and interpreted in units related to "risk" of radiation damage to the employee.

The following equation gives the best fit to experimental data from neutron spectra of different energy distributions.

$$DE = (B_6 R - B_7) - (S_6 R - S_7) \quad CF \times (\ln T / -1.52) = \text{mrem}$$

where: DE = dose equivalent at the surface of the body (eye lens)

B_6 = TLD-600 against body

B_7 = TLD-700 against body

S_6 = TLD-600 near source

S_7 = TLD-700 near source

R = TLD-700/TLD-600 gamma sensitivity ratio

CF = empirically determined from some known source for your TLD material

$(\ln T/-1.52)$ = an energy correction and limited to positive values between 0.05 to 1.0. If outside these limits set = 1.0.

$$T = \frac{(S_6 R - S_7)}{(B_6 R - B_7)} = \text{ratio of } S_6/B_6 \text{ after gamma subtraction}$$

Actual values of the constants are dependent on reader sensitivity and TLD batch characteristics. It has been helpful to normalize reader response and TLD material response to that of the TLD-700 ribbon in the following manner: To determine the relative sensitivity of the TLD-600 and TLD-700 material to gamma radiation, let R equal the ratio of TLD-700/TLD-600 for the material when exposed to gamma radiation, and set the reader to give one unit of output (coulombs, counts, etc.) per mR of gamma on the TLD-700 material.

At the Savannah River Plant, we have chosen to calibrate the response of the TLND to a $^{238}\text{PuF}_4$ spectrum which is "scattered" slightly by the presence of its paraffin-filled storage drum and the walls and floor of the shielded storage room. The spectrum shape is shown in Figure 5 for energies above 0.25 Mev. Other measurements were made to determine the thermal and low energy components below 0.25 Mev and are included in the table at the top of Figure 5.

The derivation of DE from this $^{238}\text{PuF}_4$ spectrum is based on fluence-to-DE factors at the surface of an infinite slab 30-cm-thick. This implies the lens of the eye as a critical organ and is generally the maximum DE with respect to other tissue depths. Figure 6 compares the infinite slab interpretation with the older first collision concept. Little difference is seen at energies greater than 0.1 Mev, but large differences exist at lower energy regions. The best estimate of DE from the PuF_4 source is shown in Figure 5 to be 13.3 mrem/hr at 32 inches.

In the equation, the CF is derived from an exposure by substituting the known value of DE and TLD readout results and solving for CF. The relative response of the badge to all other sources discussed is based on this calibration.

EXPERIMENTAL DATA

The determination of neutron dose equivalent from a suitable number of test spectra of different energy distributions is one of the major problems in developing and testing personnel dosimeters. Since all instruments are energy dependent and no dose equivalent standards exist, the most accurate technique, at present, is to derive the dose equivalent from fluence spectral measurements. The derivation of dose equivalent as shown for the $^{238}\text{PuF}_4$ source in Figure 5 was carried out in a similar manner for most of the other test exposures discussed in the following paragraphs. These test exposures are included in a more detailed report⁽³⁾ to be issued in the near future. Fluence-to dose equivalent conversion factors were based on interpretation at the surface of an infinite slab 30-cm-thick and is shown as the topmost curve in Figure 6.

The response of the $^{238}\text{PuF}_4$ standard to a large number of neutron spectra of widely different energy distribution has been measured. These data are summarized according to laboratory where the TLND was exposed or according to the degree of scattering present during the exposure. The precision of measuring the DE from any one spectrum is $\pm 8\%$ at one standard error.

SRP EXPOSURES

A movable, 10 Ci PuBe source was contained in a stainless steel drum filled with D_2O . The source could be positioned against the inside of the front face (0 mm position) or at other depths in the tank. Our calibrated positions were at 0 mm D_2O , 150 mm D_2O , and 330 mm D_2O . The relative response of the dosimeter for each of these settings is shown in Figure 7. In general, the results fall within $\pm 20\%$ of the known DE's. The responses of other type neutron survey meters are shown in Figure 8.

ORNL EXPOSURES

In Figure 9, results are shown for two consecutive years for three spectra. The first is the unmoderated fission spectrum in air at 3 meters; the second represents the degradation of the fission spectrum by a 12-cm-thick Lucite* shield; and the last is a spectrum degraded by a 13-cm-thick iron shield. All responses are higher than the DE based on threshold detector units exposed at the same time on the 3-meter arc in air. For personnel monitoring, an over response of this magnitude is acceptable.

LOS ALAMOS EXPOSURES

The results of the Los Alamos exposures (Figure 10) were reported in units of absorbed dose (rads) and were converted to surface DE by using spectra published by Hankins.⁽²⁾

The Parka⁽²⁾ spectrum was used to interpret DE from Honeycomb and Pee Wee exposures. The procedure was to calculate the fluence from each spectrum that would be required to yield the reported dose, and then use this fluence and spectrum to derive the DE. Because the leakage spectra are theoretically calculated, there is some error introduced by applying this spectrum to exposures at distance greater than 3 meters where scatter and air moderation could affect the distribution.

The result for the 510-cm position away from the flattop assembly was based on inverse square from the 110 cm position with the assumption that the spectrum distribution was unchanged. The result probably represents a maximum over response of the dosimeter at that position.

Similarly, the 13 meter position was behind a wall where the spectrum distribution was unknown, but highly scattered. This position was reported by Hankins.⁽²⁾ Normalizing the reported absorbed dose to the response of neutron detectors, the following DE's were obtained for this exposure:

<u>Dosimeter</u>	<u>Measured DE, rem</u>
TLND	391
Multisphere	84
TDU (above 200 keV only)	88
10 inch sphere	121
NTA film	22
Hurst Proportional	12

* Tradename of duPont Company

To derive the relative response of the TLND, we used the 84 rem from the multisphere. The dosimeter possibly responds high to this particular spectral distribution, even though the response to the Water Boiler (a very thermalized spectrum) was low.

All other exposures to the various types of assemblies at Los Alamos were reasonably well known and were satisfactory from a personnel monitoring viewpoint.

UNSCATTERED SPECTRA IN AIR

Exposures at SRP, LASL, and National Accelerator Laboratory confirmed that energetic neutron sources (PuBe or ^{252}Cf) suspended above the floor and away from walls will produce a low response in the dosimeter at distances of less than 3 meters. The albedo badge not only requires a body behind the badge but is also sensitive to room scatter for its response to energetic spectra. This is reasonable because there is insufficient moderation in the badge to thermalize a large fraction of the fast neutron fluence.

The effect of scatter was confirmed in an additional experiment where a PuBe source was suspended 1 meter off the floor and held at a constant distance of 100 cm from the dosimeter-jug phantom. As the entire source-phantom combination was shifted toward a 12-inch-thick concrete wall, the response increased smoothly until, with the source against the wall, the response was 10% high.

EXPOSURES AT MOUND LABORATORY

Another demonstration of the effects of scatter was observed during an exposure of various dosimeters to a bare SNAP-19 thermoelectric generator (^{238}Pu) at Mound Laboratory. The experiment consisted of a circle of water-filled jugs with dosimeters raised 10 feet above the nearest scattering material which was the floor. The distance from the source to phantom front was 100 cm. After one hour exposure time, at least two of the jugs were removed and the exposure continued for an additional six hours. There was sufficient scatter from associated phantoms during the first hours to result in a

satisfactory response. The longer exposure was more scatter-free and resulted in an under response which agrees with the ^{252}Cf and PuBe exposures in a scatter-free geometry. The $^{238}\text{PuO}_2$ calibration standard used at Mound agreed with our $^{238}\text{PuF}_4$ calibration which confirmed that both interpretations of DE were similar.

EXPOSURES AT THE NATIONAL ACCELERATOR LABORATORY

This series of exposures were made to determine the response, if any, to energetic neutrons resulting from high energy acceleration operation. The badge and a 10-inch-sphere response were compared to the DE derived from multisphere data. The results were somewhat erratic but better than was expected when the high average spectral energies of these exposures were considered.

MONOENERGETIC RESPONSE

Tests of response to monoenergetic neutrons were made at BNWL and WPAFB. The results indicate a high response of a little less than a 100% between 200 to 400 KeV. Above 1 MeV the response drops to about 0.25 and remains relatively constant to the 9 MeV data points. No information was available for energies below 125 KeV except for the thermal result from the Water Boiler exposure at LASL. It is not difficult to see from this curve how scattered energetic neutrons rapidly increase the response of the dosimeter by the build-up of fluence in the peak region of over response.

DIRECTIONAL RESPONSE

The directional response of the TLND shows some energy dependence but is almost equal to the directional sensitivity of NTA film type dosimeter, that is, front = 1.0, side = 0.6, and rear = 0.2.

CONCLUSIONS

The albedo dosimeter described in this report is not the final answer to all personnel neutron dosimetry problems. Like all other devices it has limitations in certain energy regions of the spectrum. At installations where neutron sources are contained or sufficient scatter is present, the dosimeter provides a reasonably accurate method of integrating personnel exposures from unknown spectra.

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LOCATION	FILM mR	NTA FILM mrem	MBLE CaF ₂ mR	MBLE LiF mR	U PITS 1.9mrem/tr mrem	ALB (7) Badge γ mR	ALB (6) Badge Neut mrem	12" BONNER SPHERE DE mrem
1. Tgt. Desk	150	60	73	195	135	12	142	45
2. SEB Cross walk	215	2770	159	550	1830	135	690	820
3. 922 Irma	50	200	34	-	214	0	138	-
4. P. S. East Hyperon	205	1130	96	425	595	53	690	285
5. Collins- Argo	20	33	24	46	15	0	15	11
6. Col-Christ	75	380	55	136	95	24	173	91
7. Rutgers- Test	80	310	54	-	173	15	142	97
8. C-8 Col- Brdge.	295	920	151	420	940	87	1250	580
9. HO" Bin Shield	170	270	68	-	376	50	490	210
10. Prince- Fitch	80	435	50	213	210	11	123	82
11. Muon Trl.	50	10	37	-	57	97	23	9

Consider the expected pit response to 1/E spectrum

$$(P)_{1/E} = \left[\int (\phi_i \sigma_i)_{1/E} dE \right] (N \text{ eff}) \phi_{1/E}$$

$$\text{where } \int \phi_i dE = 1 \text{ N/cm}^2$$

$$\phi_{1/E} = \text{Field Integ. flux}$$

$$(N \text{ eff}) = \text{Target sized counting eff.}$$

Both (N eff) and $\phi_{1/E}$ unknown

$$(P)_{\text{AmBe}} = \left[\int (\phi_i \sigma_i)_{\text{AmBe}} dE \right] (N \text{ eff}) \phi_{\text{AmBe}}$$

$$(N \text{ eff}) = \frac{(P)_{\text{AmBe}}}{\left[\int (\phi_i \sigma_i)_{\text{AmBe}} dE \right] \phi_{\text{AmBe}}}$$

substituting

$$\phi_{1/E} = \frac{(P)_{1/E} \left[\int (\phi_i \sigma_i)_{\text{AmBe}} dE \right] \phi_{\text{AmBe}}}{\left[\int (\phi_i \sigma_i)_{1/E} dE \right] (P)_{\text{AmBe}}}$$

or

$$\phi_{1/E} = (P)_{1/E} \left[\frac{\int (\phi_i \sigma_i)_{\text{AmBe}} dE}{\int (\phi_i \sigma_i)_{1/E} dE} \right] \left[\frac{\phi_{\text{AmBe}}}{(P)_{\text{AmBe}}} \right]$$

$$DE = \phi_{1/E} \left(\frac{DE_{1/E}}{\text{unit flux}} \right)$$

where

$$DE_{1/E} = \int (\phi_i DE_i)_{1/E} dE$$

$$DE = (P)_{1/E} \left[\frac{\int (\phi_i \sigma_i)_{AmBe} dE}{\int (\phi_i \sigma_i)_{1/E} dE} \right] \left[\frac{\phi_{AmBe}}{(P)_{AmBe}} \right] \left[\int (\phi_i DE_i)_{1/E} dE \right]$$

$$DE = (P)_{1/E} \frac{2.09 \times \frac{5.78 \times 10^6 \text{ N/cm}^2}{18.6 \text{ pit/cm}^2} \times 1.29 \times 10^{-5} \frac{\text{mrem}}{\text{N/cm}^2}}$$

$$\frac{DE}{(P)_{1/E}} = \frac{8.41 \text{ mrem}}{\text{pit/cm}^2}$$

Sparker Area 4.46 cm^2

$$\frac{DE}{(P)_{1/E}} = \frac{8.41}{4.46} = \frac{1.9 \text{ mrem}}{\text{pit/cm}^2}$$

$$\frac{\int_{10^2 \text{ MeV}}^{.5 \text{ MeV}} (\phi_i^{DE_i})_{1/E} dE}{10^{-8}} = .848$$

$$\int_{10^2} (\phi_i^{DE_i})_{1/2} dE$$

$$\text{Calc} \quad \frac{(P)_{\text{AmBe}} \text{Th}^{232}}{(P)_{\text{AmBe}} \text{U}^{238}} = 0.286$$

$$\text{Exp.} \quad 0.35 \pm 20\%$$

$$\text{Sens. (AmBe)} \quad {}^{238}\text{U} \quad \frac{2.3 \pm 0.2 \text{ mrem}}{\text{pit}/4.46 \text{ cm}^2}$$

$${}^{232}\text{Th} \quad \frac{6.9 \pm 0.6 \text{ mrem}}{\text{pit}/4.46 \text{ cm}^2}$$

TLD 600 Thermal Neut. Sens.

$$\frac{\phi_{1/E} \int_{10^{-8}}^{10^{-2}} (\phi_i^{DE_i})_{1/E} dE}{\int_{4 \times 10^{-6}}^{10^{-8}} (\phi_i)_{1/E} dE} \approx \frac{1}{3.5 \times 10^4}$$

where TLD 600 N_{th} sens is $\frac{7.75 \times 10^3 N_{th}}{cm^2 mR}$

$$DE_{1/E} = \frac{7.75 \times 10^3}{3.5 \times 10^4} = 0.22 \frac{mrem}{mR}$$

Fig. 1

Fig. 2

Fig. 3

Fig 4. SAVANNAH RIVER LABORATORY TLD NEUTRON DOSIMETER

SKETCH OF BADGE COMPONENTS

FIG. 5 SPECTRUM OF PuF_4 SOURCE IN AIR AT 32 IN.

Flux Distribution, n/cm^2-sec :

Thermal - 8.14

0.4 eV to 0.2 Mev - 43.2

>0.2 Mev - 91.8

Estimated Dose Equivalent: 13.3 mrem/hr

FIG. 6 =COMPARISON OF FIRST COLLISION AND INFINITE-SLAB
DE AS FUNCTION OF NEUTRON ENERGY

THERMOLUMINESCENT NEUTRON DOSIMETER
(TLND) RESPONSE COMPARED TO NUCLEAR
EMULSION NTA AND OTHER NEUTRON SURVEY INSTRUMENTS
(TLND response normalized to PuF_4 in air)

Fig. 8

Fig. 9

EFFECT OF SCATTER ON RESPONSE OF TLND
($^{239}\text{PuBe}$ SOURCE TO PHANTOM DISTANCE = 100cm)

Fig. 10

"UNSCATTERED" MONOENERGETIC NEUTRON RESPONSE

FAST NEUTRON RADIATION DAMAGE EFFECTS IN
CRYSTALLINE ANTHRACENE AND EVALUATION AS
A DOSIMETRIC TECHNIQUE

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Some background information might be helpful to give a perspective to the work discussed here. The University of Wisconsin research program in luminescence dosimetry began in about 1960 under the direction of Professor John R. Cameron. This program involved both fundamental studies and applied developmental work on thermoluminescence dosimetry for x-ray and gamma radiation. Within the past few years, common interests in certain classes of materials led to some collaborative experiments joining Professor Cameron's radiological physics group and my solid state physics project in investigating defects and impurities in solids. During the past year this program has evolved somewhat from the gamma dosimetry area and has begun to stress investigations in fast neutron radiation damage. The damage phenomena induced in solids by fast neutron irradiation and evaluation for development of possible dosimetric techniques now involves full cooperation and coinvestigation by the radiological and solid state research groups.

We have completed some preliminary studies in a variety of solids, we are continuing some of the promising ones, and we are attempting to initiate some new ones. I shall be most happy to describe our efforts in detail during informal discussion; for the immediate presentation, however, I will limit myself to one area, a study of radiation damage effects on the optical properties of metastable excitons in anthracene. To preface this discussion, I would like to review briefly some general considerations of fast neutron solid state dosimeter materials and techniques from the viewpoint of a solid state physicist.

The first problem in low level dosimetry is the extremely rare concentration of damage sites produced in a material. We can estimate upper

bounds on the order of 10^{12} defect centers per cm^3 induced by a dose of one rad. For typical tissue density materials this corresponds to one part in 10^{10} relative to undisturbed atoms; by a solid state physicist's standards this constitutes a totally insignificant degree of imperfection of the sample material. Only accidentally might certain associated phenomena occur by which such damage levels could be measured.

In order to achieve satisfactory sensitivity for detecting low level damage, a simple consideration of intrinsic properties of materials and of detectors indicates that measurements should be based upon physical phenomena whose characteristic energies are at least on the order of an electron volt. Such phenomena include optical activity (e.g., luminescence), chemical interactions (e.g., photographic development and etched tracks), and induced electrical conduction effects (e.g., TSEE and photoconductivity). In addition, radiation damage effects should be measured above the smallest possible background; that is, the measurement technique should be a non-subtractive one like thermoluminescent emission rather than a subtractive technique like optical attenuation.

A second problem relevant to dosimetry materials for fast neutrons is to discover phenomena which favorably discriminate (or, at least, do not unfavorably discriminate) on the higher LET characteristic of proton recoils or (n, α) reactions. To discriminate according to local damage distributions, either the damage mechanisms themselves or the measured outputs which reveal the damage (or both) must involve what can generally be called non-linear phenomena. For example, non-linearities in the yield of dicentric chromosome aberrations in living cells appears to discriminate in favor of high LET radiation, whereas photoexcited fluorescence from radiation damage centers is often quenched if the local distribution of active sites rises above some critical density; the latter would produce, on a rad basis, very unfavorable neutron to gamma discrimination factors.

With this background in mind, I would like to outline some basic electronic properties of solid anthracene; much of this is applicable to the other cyclic hydrocarbon solids as well. The three lowest lying electronic states of the molecule are illustrated in the following diagram.

The first excited singlet state, $1S$, is strongly coupled optically to the singlet ($1A_g$) ground state and produces the characteristic strong blue fluorescence of anthracene. The triplet state, $1T$, is metastable; the electron spin selection rules make its optical coupling to the singlet states very weak so that even in the purest materials the normal triplet lifetimes are governed by molecular vibration non-radiative processes. In the solid state a single molecular excitation is mobile and, during its lifetime, is associated with many molecular sites; such an unlocalized excitation state is called an exciton. An exceptional non-linear property of the triplet excitons is that two of them may collide and annihilate to produce one singlet exciton which promptly decays via the blue fluorescence emission; this is called delayed fluorescence or triplet annihilation fluorescence and becomes the dominant decay mode at triplet concentrations above about $5 \times 10^{12} \text{ cm}^{-3}$. The triplet exciton properties are known to be extremely sensitive to sample quality and impurity content although specific mechanisms are not well understood; the normally weak red phosphorescence can be greatly enhanced by some kinds of trapping sites, the non-radiative decay rate can be greatly influenced, and the triplet-triplet annihilation probability substantially modified. Anthracene can be zone refined for purification and grown in large single crystals of good optical quality for experimental measurements.

Anthracene thus seemed to us to be a promising material for some fundamental studies at room temperature relating to fast neutron dosimetry methods.

It is hydrogen rich to give desirable tissue equivalency properties, it has optical emission properties involving long-lived excitonic states and strong non-linear phenomena, high purity optical quality samples can be grown, and it is known to be very sensitive to defects and impurities.

In the following, I will present only a genral outline of our results and reserve the experimental details for later discussion, if appropriate. We began preliminary rough experiments just about one year ago and first observed that 14 MeV neutron irradiation suppressed the blue annihilation fluorescence yield when triplet excitons were directly created by strong incident red light. We also noted that these neutron irradiation effects were at least as strong on a rad basis as those induced by x-rays. Our first attempts at neutron dosimetry to calibrate the anthracene dose level gave us, in fact, about a 6 to 1 discrimination factor favoring the fast neutron rad; this now appears, after more careful calibrations using copper activation analysis, to be incorrect, unfortunately. Within the experimental variations observed, it seems that the fluorescence suppression effects follows basically a simple rad dependence and does not vary grossly with the quality of the incident radiation. I would like to emphasize here that annihilation fluorescence suppression, although easily measured, is basically a subtractive technique and suffers the associated sensitivity limitations.

During the past year we have extended the anthracene studies to investigate the following questions.

1. Which parameters describing the excitron emission properties are sensitive to radiation damage?
2. Do the observed suppression effects show any fading properties?
3. What are the sample dependences between different batches obtained from the same supplier or between samples from different suppliers?
4. Are there observable neutron incident energy dependences?
5. Are there any spectral or yield changes associated with the red phosphorescence triplet direct optical emission?

The parameters governing the measured triplet exciton properties are the optical cross-section for creating the excitations, the linear decay rate ($\beta = (1/\tau_{\text{rad}}) + (1/\tau_{\text{nonrad}})$), and the bilinear decay rate or triplet annihilation cross-section. In a series of experiments we measure both the relative annihilation blue fluorescence total yield under steady state red light excitation and the delayed fluorescence decay curve after pulsed red light illumination. Analysis of the results shows that only the linear decay rate appears to be sensitive to radiation damage: $\beta = \beta_0 + \underline{a}D$, where D is the dose in rads and $\underline{a} \approx 0.4 \text{ sec}^{-1} \text{ rad}^{-1}$.

At room temperature we have observed no measurable fading of these effects over a period of many months. We have looked briefly at annealing properties which give erratic results; post-irradiation annealing seems temporarily to change the degree of suppression (we have observed both positive and negative changes), but after a few days the suppression seems to stabilize returning to its pre-annealing value.

We do observe substantial sample dependences both between batches of one supplier and from different suppliers. It appears, unfortunately, that both the pre-irradiation lifetime, β_0 , as well as the dose sensitivity factor, $\underline{a}$, can vary perhaps as much as a factor of two in extreme cases. We have acquired high purity anthracene powder, but have not determined if its optical properties are satisfactory for such studies.

Experiments using 2.5 MeV D-D neutrons have been compared with the 14 MeV D-T neutron and x-ray irradiation experiments. Within the accuracy of our exposure measurements and observed inter-sample variations the fluorescence suppression seems to depend only upon deposited energy.

Finally, we have not yet been able to instrument the experiment to study the red phosphorescence emission. This is of great practical concern because if even a small part of the enhanced triplet decay rate is associated with a direct red emission decay mode one can avoid the intrinsically subtractive annihilation fluorescence suppression measurement. We would then expect greatly increased dose sensitivity and hopefully elimination of some of the sample dependences.

We feel that the presently observed damage effects in anthracene do offer some potential for selected dosimetric applications, but even when the sample dependences can be controlled a sensitivity limit of 2-5 rads is about the best expected. The lack of any apparent discrimination against dense neutron damage is good, and the fading and stability properties appear excellent. We hope to pursue the red emission studies as quickly as possible to determine if this might provide far more sensitive methods for detecting the radiation damage effects.

THE USE OF TRACK REGISTRATION FROM FISSION FRAGMENTS
FOR DOSIMETRY AT THE 20 GEV STANFORD LINEAR ELECTRON ACCELERATOR (SLAC)

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INTRODUCTION

Only a very small number of people at SLAC are exposed to any significant radiation dose, more than 90% of it being due to photon radiation. In fact the monthly film badges indicated only 5 significant neutron exposures during the entire year of 1970, the highest of which was 300 mrem. From the first of July 1971, the film badge system was abandoned and ^7LiF and ^6LiF are now being used. These dosimeters will be issued on a yearly basis for most employees and only about 20% of the people will have a TLD badge issued and evaluated on a quarterly basis.

The success of this system is strongly dependant on a frequent area monitoring system involving fission track detectors. The technique was used frequently during the past two years in order to resolve specific neutron problems around the accelerator and its application is described in SLAC-TN-70-33 by R. Gay and G. Svensson⁽¹⁾. The fissile materials to be discussed are thick foils of Thorium and Uranium with different degrees of depletion.

Of the different counting methods described, we have chosen to use an electric spark counter⁽²⁾ for low track densities ($<10^3$ tracks/cm²) and a binocular microscope for 10^3 to 10^6 tracks/cm².

Two types of plastic have been used as detector material. 1 mm thick Lexan in connection with microscope counting and 10 μm thick film of polycarbonate (KIMFOL or MAKROFOL) when electric spark counting is preferred. The RC-circuit used with the electric spark counter is shown in Figure 1. The actual counter has been described by W. G. Cross.⁽²⁾

A major problem for this area monitoring system when applied at SLAC is the photon component, which has a wide energy distribution. To evaluate

the importance of such a photon field when using Thorium and Uranium foils together with plastic detectors, an exposure was made in a 13 GeV bremsstrahlung beam at SLAC. A more detailed study of photofission has been going on for the last six months, using SLAC's monoenergetic photon beam. Preliminary results of the cross sections are available.

Basic Equation: It is easily shown that the measured track density T (cm^{-2}) relates to the particle fluence through:

$$\begin{aligned} {}^{232}\text{Th} \quad T &= \epsilon \cdot 1.33 \cdot 10^{-5} \int_{E_1}^{E_2} \phi(E) \sigma(E) dE \\ &= \epsilon \cdot 1.33 \cdot 10^{-5} \sigma_{\text{eff}} \int_{E_1}^{E_2} \phi(E) dE \end{aligned}$$

$$\begin{aligned} {}^{238}\text{U} \quad T &= \epsilon \cdot 1.29 \cdot 10^{-5} \int_{E_1}^{E_2} \phi(E) \sigma(E) dE \\ &= \epsilon \cdot 1.29 \cdot 10^{-5} \sigma_{\text{eff}} \int_{E_1}^{E_2} \phi(E) dE \end{aligned}$$

where

$\phi(E)$ = differential particle fluence [cm^{-2} , MeV^{-1}].

$\sigma(E)$ = fission cross section per nucleus [barn] at energy E .

ϵ = efficiency of fission fragment registration and counting.

Experimental Measurement of ϵ :

To measure ϵ a 10 Ci Pu-Be neutron source was used. For ${}^{232}\text{Th}$ and the Pu-Be spectrum σ_{eff} has been determined to be: $\sigma_{\text{eff}} = 0.14$ barn. Thin (10 μm) polycarbonate insulating film detectors (area = 1.27 cm^2) coupled with ${}^{232}\text{Th}$ were placed on a device such that they rotated about the 10 Ci Pu-Be source at a radius of 40 cm. The yield of the source is given

as 2.05×10^7 n/sec. With this knowledge $\int \phi(E) dE$ is easily calculated for a given run time. Experimental results follow.

Theoretical No. of Tracks per cm^2 ($= 1.33 \times 10^{-5} \sigma_{\text{eff}} \int \phi(E) dE$)	No. of Counts per $\text{cm}^2 (=T)$	$\epsilon = \frac{T}{1.33 \times 10^{-5} \sigma_{\text{eff}} \int \phi(E) dE}$
41	17	0.423
72	37	0.434
113	49	0.434
291	124	0.425
643	266	0.414
1094	336	0.308
2547	794	0.312

These results are plotted in Figure 2. It is seen that ϵ remains essentially constant over a range below 650 counts/cm^2 . It is quite convenient that this is the range over which this method works best since microscopic counting is applicable from the top of this range up to about 10^6 counts/cm^2 . With a larger area detector ($\sim 10 \text{ cm}^2$) it appears feasible to count track densities as low as 0.5 tracks/cm^2 .

Neutron Cross Section for ^{232}Th (Figure 3)

There are no published neutron fission cross sections for ^{232}Th above about 50 MeV. Proton fission cross sections have therefore been used. This is justified under the assumption that the Coulomb barrier of the nucleus is the major contributor to differences between neutron and proton cross sections. This Coulomb barrier is negligible at high energies.

In order to determine σ_{eff} , the energy distribution of the neutrons has to be known. Only a few actual measurements using nuclear emulsions or neutron spectra have been performed at SLAC. One measurement was performed in the accelerator tunnel, 30 meters away from a Cu dump (23 cm thick) in which a 9.9 GeV electron beam was absorbed. The spectrum showed a $1/E$ dependence on neutron energy ($\phi(E) dE = k E^{-1} dE$) between 2 - 10 MeV.

Another measurement⁽³⁾ using the same technique in a more shielded situation showed an E^{-2} dependence below 6 MeV. This resembles the shape of a giant resonance neutron spectrum. Above 6 MeV the spectrum seemed to approach an E^{-1} dependence. Depending upon electron beam operation and shielding configuration, the energy distribution of the neutrons may change drastically and σ_{eff} for these energy distributions and for a Pu-Be spectrum have been calculated, using Figure 3 and the assumptions mentioned above.

$$1. \quad \phi(E) dE = \phi_0 E^{-2} dE \quad \text{when } 1.5 \text{ MeV} < E < 6 \text{ MeV}$$

$$\text{and } \phi(E) dE = \phi_1 E^{-1} dE \quad \text{when } 6 \text{ MeV} < E < 10^3 \text{ MeV}$$

$$\text{This gives } \sigma_{\text{eff}} = 0.40 \text{ barn}$$

$$2. \quad \phi(E) dE = \phi_2 E^{-1} dE \quad \text{when } E > 1.5 \text{ MeV}$$

$$\text{This gives } \sigma_{\text{eff}} = 0.43 \text{ barn}$$

$$3. \quad \text{Pu-Be spectrum}$$

$$\sigma_{\text{eff}} = 0.14 \text{ barn}$$

Minimum Detectable Neutron Fluence ($\int \phi(E) dE$) for Personnel Dosimetry.

The three cross sections discussed above will be used. The quantity of interest can be solved from the basic equation.

$$\int \phi(E) dE = \frac{T}{1.3 \cdot 10^{-5} \epsilon \sigma_{\text{eff}}} \quad \text{neutrons/cm}^2$$

The detector system will consist of 10 cm^2 of ^{232}Th together with $10 \mu\text{m}$ thick Kimfol. The experiments indicate that the number of background tracks in one month's exposure is less than 1 track per 10 cm^2 of ^{232}Th . Because of this it is possible to interpret as low as 5 tracks. For a 10 cm^2 ^{232}Th foil this corresponds to 0.5 tracks/cm^2 .

High energy: ($\sigma_{\text{eff}} = 0.40$ barn):

$$\int_{1.5}^{10^3} \phi(E) dE = \frac{0.5}{1.3 \cdot 10^{-5} \times 0.42 \times 0.4} = 2.3 \cdot 10^5 \text{ n/cm}^2$$

If, for example, the absorbed dose to fluence conversion for 100 MeV neutrons is chosen as $1.9 \cdot 10^{-8} \text{ rad cm}^2$, the minimum detectable neutron fluence corresponds to $4 \cdot 10^{-3} \text{ rad} = 4 \text{ mrad}$.

Pu-Be: In the case of a lower energy spectrum, the effective cross section is smaller and the minimum detectable neutron fluence correspondingly higher,

$$\int_{1.5}^{10.6} \phi(E) dE = 6.5 \cdot 10^5 \text{ n/cm}^2$$

The absorbed dose to fluence conversion for 4.5 MeV neutrons is $4.5 \cdot 10^{-9} \text{ rad cm}^2$, which gives

$$6.5 \cdot 10^5 \cdot 4.5 \cdot 10^{-9} = 2.9 \cdot 10^{-3} \text{ rad} = 2.9 \text{ mrad}$$

Photofission

A major component of a radiation field at an electron accelerator is photons, created by radiative processes when the electrons interact with targets or other absorbing materials. The energy distribution and intensity of the photons around SLAC varies strongly depending on the shielding material and configuration. Many of the electron beam absorbing materials consist of Cu or Fe. A strong contribution to the photon component is, therefore, seen in the 8 MeV region, corresponding to the minimum of the attenuation coefficient for Cu and Fe.

For U and Th, photofission is a competing process. Even for the fairly low energy of 8 MeV (see above), the cross section for photofission is significant. Measurements of the U cross section in the energy range of

5.4 to 7.7 MeV for mono-energetic photons has been done by Manfredini et al,⁽⁴⁾ and at 7.7 MeV the cross section is about 9 mb. No data seems to be available for Th at this low energy, but between 50 MeV and 200 MeV the ratio between U and Th photofission cross sections stay constant and is approximately equal to 3 (see Figure 4).

Using this ratio gives a photofission cross section for Th equal to 3 mb, for 8 MeV photons. The importance of this competing process in a neutron measurement can now be evaluated as above:

$$\int \phi_{\gamma}^{-}(E) dE = \frac{0.5}{0.43 \times 1.3 \cdot 10^{-5} \times 0.003} = 3 \cdot 10^7 \text{ quanta/cm}^2$$

This corresponds to an energy fluence of $24 \cdot 10^7 \text{ MeV cm}^{-2}$. The energy absorption is now

$$24 \cdot 10^7 [\text{MeV cm}^{-2}] \mu_{\text{en}} [\text{cm}^2 \text{ g}^{-1}]$$

$$1.6 \cdot 10^{-6} [\text{erg MeV}^{-1}] \cdot 10^{-2} [\text{rad g erg}^{-1}]$$

For 8 MeV photons and water as absorbing medium, $\mu_{\text{en}} = 0.018 \text{ cm}^2 \text{ g}^{-1}$.

The absorbed dose from 8 MeV photons corresponding to the minimum detectable track density is

$$24 \cdot 0.018 \cdot 1.6 \cdot 10^{7-8} = 0.69 \cdot 10^{-1} \text{ rad} = 69 \text{ mrad}$$

In many areas the photon spectrum may be very high energy and as mentioned above, experimental work has been performed to evaluate high energy photofission. Figure 4 shows data from other authors plus one point from an experiment performed at SLAC.

Using 0.28 barn as cross section for ^{232}Th at 13 GeV bremsstrahlung, the minimum detectable fluence of photons (equivalent quanta = Q) is

$$Q = \frac{0.5}{0.43 \times 1.3 \cdot 10^{-5} \times 0.28} = 3.2 \cdot 10^5 \text{ quanta/cm}^2$$

The energy fluence is

$$QE_0 = 3.2 \cdot 10^5 \times 13 \cdot 10^3 = 41 \cdot 10^8 \text{ MeV/cm}^2$$

The dose to fluence conversion for this high energy is about $4 \cdot 10^9 \text{ MeV/cm}^2$, rad. Therefore the minimum detectable photon dose will be as high as 1 rad before it seriously interferes with the neutron monitoring.

CONCLUSION

In using the fission fragment technique for area monitoring of neutrons at SLAC we need to simultaneously measure absorbed dose of photons and understand the energy distribution of photons at the different locations.

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Fig. 1 - The electrical circuitry used in this study.

Fig. 2 - The fission fragment registration efficiency (ϵ) versus track density.

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Fig. 3

Fig. 4

SPARK COUNTING SURVEY SUMMARY

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I. REGISTRATION PLASTIC

Makrofol is very difficult to obtain in the United States. Most people are using 10 micron Kimfol. LLL uses 6 micron Kimfol currently, although Oak Ridge has indicated in the past that they had experienced difficulty with reproducibility using foil that thin. The thinner material is of advantage only for low counting situations when the increased efficiency is valuable.

II. EXPOSURE CONDITIONS

Radiators used include ^{239}Pu , ^{237}Np , ^{238}U (both thick and electro-deposited), thick ^{232}Th , thick ^{235}U , and ^{252}Cf . The plastic is exposed with or without a support, but usually etched in an embroidery hoop arrangement using two small rings. The exposed areas ranged from 1 to over 4 cm². The separation between foil and radiator has ranged from contact to 0.040".

III. ETCHING CONDITIONS

Everyone surveyed used KOH, most at 28%. The etchant temperature reported ranged from 40°C to 70°C, with associated etch time from 3 hours down to 40 min. The trade off seems to be easier control of etching conditions as opposed to shorter waiting times. A similar degree of success has apparently been experienced over this range of etching temperatures.

Most people reporting use stirring and gentle agitation would seem to provide more uniform etching.

The heat source does not seem critical. Los Alamos does as well with a hot plate as others who use constant temperature baths.

Only one laboratory cleans the material before use and the success of those who don't indicate it is probably not necessary.

The laboratories that do not use an acid rinse have reported reasonable success. However, intuitively, an acid rinse should provide more positive elimination of the etchant.

IV. COUNTING CONDITIONS

Most people feel it is necessary to use pressure on the foil during counting. The pressures indicated range from 100-500 gms/cm². In some cases a layer of rubber padding is used on the weight to apply more uniform pressure to the foil during sparking.

Electrodes of nickel, brass, or stainless steel were reported.

The most popular capacitance is about 100 pf. Although successful counting can be done with higher capacitance, it probably reduced the upper limit of linear countable track density.

The prespark voltages reported range from 600 to 1500 volts. It is clear that tracks which do not penetrate the film or enter at steep angles should be "cleared" at a voltage higher than that used for the actual count. Again success was achieved over the range of values reported.

The record counts are uniformly made at 500 V, although the number of counts required to determine a measurement varies.

Both positive and negative polarities were reported for the metal electrode.

We see wide variation in the criterion used to terminate a particular count. At high track densities, counts will be registered for many minutes after counting is begun.

Repositioning and cleaning of foils between counts is again a matter of wide variation. It is well known that debris builds up on the foil and electrode after repetitive counts, and that should be kept in mind when establishing the counting procedure.

V. SYSTEM PERFORMANCE

Spark counting systems can have almost statistical reproducibility for track densities up to 3000 cm². This upper limit will be reduced when larger capacitance is used.

The problem of a "stuck count" or a foil that won't count is probably, as suggested by Bill Cross, a short either by debris or an improperly cleaned and dried foil.

VI. GENERAL COMMENTS

See individual summaries, particularly that from Los Alamos.

SPARK COUNTING SURVEY

Laboratory: Stanford Linear Accelerator

Representative: Goran Svensson

Registration Plastic

Trade Name: Kimfol

Thickness: 10 microns

Exposure Conditions

Radiators: Thick radiators. Primarily Th

Exposed Area: Usually 1 cm².

Plastic Support: Two concentric plastic rings.

Plastic-Radiator Separation: Contact.

Etching Conditions

Etchant: KOH Temperature: 60°C. Concentration: 5 N Etch Time: 60 min.

Stirring Used? Yes. Etchant Volume: 400 ml.

Etchant Heat Source: Constant temperature water bath.

Pretreatment Used: Foils tested at 1500 volts for pinholes before use.

Rinse Conditions: Water and low concentration weak acid.

Counting Conditions

Pressure Applied: 1 lb, 14 oz lucite lid

Electrode Material: Stainless steel. Capacitance: 2000 pf Prespark Voltage: 1500 V

Number of Prespark Counts: 1

Electrode Polarity: Negative.

Counting Voltage: 485 V

Number of Record Counts: ≤ 5 .

How Are Individual Counts Terminated?

Is the Foil Repositioned Between Counts? No.

Is the Foil Cleaned Between Counts? No.

System Performance

Counting Reproducibility: $\sim 5\%$.

Maximum Linear Density: $< 900 \text{ tracks/cm}^2$

Problem with foil that will not count without repositioning? Yes.

Explanation: If the passive R-C circuitry is OK, I always get holes in the Al Mylar. However, sometimes the scaler does not record without repositioning. I believe that the pulse train may be too fast under certain circumstances.

Alpha Spark Counting

Experience: No.

Plastic Used:

Source and Energy of Alphas:

Degree of Success:

General Comments on Spark Counting Experience:

SPARK COUNTING SURVEY

Laboratory: Chalk River, Canada

Representative: George Cowper, Bill Cross

Registration Plastic

Trade Name: Makrofol KG

Thickness: 10 microns

Exposure Conditions

Radiators: ^{232}Th , $^{237}\text{Np-Al}$

Exposed Area: 5 cm^2

Plastic Support: Contact.

Plastic-Radiator Separation:

Etching Conditions

Etchant: KOH Temperature: 40°C Concentration: 28% Etch Time: 3 hours

Stirring Used? Yes. Etchant Volume: 1 liter.

Etchant Heat Source: Constant Temperature Water Bath.

Pretreatment Used: None.

Rinse Conditions: Running, cold water ~ 10 minutes.

Counting Conditions

Pressure Applied: 100 to 500 g/cm^2 (not critical to reproducibility).

Electrode Material: Nickel or brass. Capacitance: 100 pf Prespark Voltage: 1500 V

Number of Prespark Counts: 2 or 3.

Electrode Polarity: Solid electrode + Mylar grounded. Counting Voltage: 500 V

Number of Record Counts: 2.

How Are Individual Counts Terminated?

Is the Foil Repositioned Between Counts? No.

Is the Foil Cleaned Between Counts? Only between prespark and record counts. Washed with dilute KOH, and rinsed in water.

System Performance

Counting Reproducibility: Repeated counts of same Makrofol - $\sigma \leq \pm 1\%$. Reproducibility of many Makrofol foils exposed to same neutron flux $\sigma \leq \pm 2\%$ after statistical variations (2%) subtracted in quadrature.

Maximum Linear Density: 3000/cm² for 10% counting loss.

Problem with foil that will not count without repositioning? Yes, occasionally.

Explanation: Short circuit through Makrofol from evaporated aluminum, so voltage can not build up. Does not occur when foil is cleaned as above, after precount sparking.

Alpha Spark Counting

Experience: Only on an experimental basis.

Plastic Used: Cellulose nitrate.

Source and Energy of Alphas: ²⁴¹Am + absorbers, to give 2-3 MeV.

Degree of Success: Less reproducible than fission counting.

General Comments on Spark Counting Experience:

SPARK COUNTING SURVEY

Laboratory: Los Alamos

Representative: Dale Hankins

Registration Plastic

Trade Name: Kimfol

Thickness: 10 microns

Exposure Conditions

Radiators: ^{239}Pu , ^{237}Np , ^{238}U

Exposed Area: 3 cm²

Plastic Support: Exposure: foil wrapped around radiator.

Etching: foil placed between concentric plastic rings.

Plastic-Radiator Separation: None except for radiator curvature.

Etching Conditions

Etchant: KOH Temperature: 70°C Concentration: 5 N Etch Time: 40 minutes

Stirring Used? Yes. Etchant Volume: 1 $\frac{1}{4}$ liters

Etchant Heat Source: Hot plate.

Pretreatment Used: None

Rinse Conditions: (See general comments.)

Counting Conditions

Pressure Applied: Thin rubber pad, with 2" x 4" dia. plastic

Electrode Material: Copper. Capacitance: 110 pf Prespark Voltage: 600 v

Number of Prespark Counts: 3 + one for each 500 sparks over 1500

Electrode Polarity: Negative Counting Voltage: 500 v

Number of Record Counts: 2 - 4.

How Are Individual Counts Terminated? Manually. Normally allowing 5 minutes, more if count has not stopped or is high (up to 8 minutes).

Is the Foil Repositioned Between Counts? Yes. Picked up, electrode brushed with horse hair brush, foil rotated slightly and replaced.

Is the Foil Cleaned Between Counts? No, except to brush off any foreign material with a horse hair brush.

System Performance

Counting Reproducibility: σ is 2% or maximum spread ± 4 to 5%, below 5000 counts.
(See below.)

Maximum Linear Density: 5000 sparks in a $3/4$ " diameter exposed area. At 5000 it is 4% low. Above 6000 count not reproducible.

Problem with foil that will not count without repositioning? Yes. After recent trip to Oak Ridge.

Explanation: Acetic acid was not used and I suspect traces of KOH may be causing part of this problem.

Alpha Spark Counting

Experience: No.

Plastic Used:

Source and Energy of Alphas:

Degree of Success:

General Comments on Spark Counting Experience:

1. The foils are rinsed by placing briefly in cold water and then into concentrated acetic acid solution (1 part glacial acetic, 4 parts water) and the beaker placed for 5 minutes into ultrasonic cleaner. Acid solution then added to 1 liter of H_2O and allowed to remain 5 to 10 minutes. Rinsed with tap water for 5 to 10 minutes and then in distilled water for 10 minutes.
2. The acetic acid quench is probably more useful as a cleaning step to remove traces of KOH from the foil. I recommend its use to avoid damage to the electrode by KOH and difficulty in getting the foils to count.
3. The sparking voltage is a function of elevation. At Oak Ridge I had to decrease my 600 to 500 voltage by 50 volts to get the same results.
4. I always etch 3 standard foils with each batch. The standard foils have ~ 1740 counts and are made by exposing a Pu^{239} foil in the thermal neutron beam. Gold foils are used to check the irradiation of the standard foils.
5. The KOH is only used once since the second etch results in a 4% drop in counts, the third to 6%.
6. The Kimfol has a shiny and a dull side. We find that with the dull side up during counting, there is a 10% increase in the number of counts obtained for our "standard" foils. These foils are placed in the hoop with the dull side out. The Pu foil is placed inside the hoop exposing the shiny side to the fission fragments. When the Kimfol is placed shiny side out and the dull side exposed to fission fragments, 10% less sparks are formed. This may be due to the exposure of the shiny vs dull side or to our etching technique where we use a stirrer (rotation of KOH solution past hoops).

SPARK COUNTING SURVEY

Laboratory: Oak Ridge National Laboratory
Representative: Klaus Becker

Registration Plastic

Trade Name: Makrofol, Kimfol
Thickness: 10 microns

Exposure Conditions

Radiators: Cr^{252} , Nat. U, Np^{237} , Th^{232} , Pu^{239} , U^{238}
Exposed Area: 3 cm^2
Plastic Support: Foil attached to plastic ring with rubber cement.
Plastic-Radiator Separation: Spacer ring or 2 micron polycarbonate.

Etching Conditions

Etchant: KOH Temperature: 60°C Concentration: 28% Etch Time: 2 hours
Stirring Used? Yes Etchant Volume: $\frac{1}{2}$ liter
Etchant Heat Source: Lab. oven
Pretreatment Used: Normally none.
Rinse Conditions: Lab. water

Counting Conditions

Pressure Applied: 500 gms., 150 gms/ cm^2
Electrode Material: Brass or nickel. Capacitance: 100 pf. Prespark Voltage: 600 v
Number of Prespark Counts: 1
Electrode Polarity: - Counting Voltage: 500 v
Number of Record Counts: 2 - 5
How Are Individual Counts Terminated?
Is the Foil Repositioned Between Counts? Sometimes.
Is the Foil Cleaned Between Counts? Yes.

System Performance

Counting Reproducibility: $\sim 3 - 5\%$ (σ).

Maximum Linear Density: 3000 sparks/cm²

Problem with foil that will not count without repositioning? Only once in Taiwan.

Explanation:

Alpha Spark Counting

Experience: Yes.

Plastic Used: Home made 15 micron cellulose nitrate foils (NRDL-1 formula),
annealed before use for 16 hours at 80°C.

Source and Energy of Alphas: Radon daughters, Pu²³⁹ source, thick U, etc.

Degree of Success: Up to 100% counting efficiency.

General Comments on Spark Counting Experience:

Not always as good as originally hoped, but still better than the other automatic techniques.

SPARK COUNTING SURVEY

Laboratory: Brookhaven National Laboratory

Representative: Carl Distenfeld

Registration Plastic

Trade Name: Kimfol

Thickness: 10 microns

Exposure Conditions

Radiators: Th²³², U²³⁸

Exposed Area: >4.46 cm²

Plastic Support: Concentric plastic rings

Plastic-Radiator Separation: Contact

Etching Conditions

Etchant: KOH Temperature: 65°C Concentration: 6N Etch Time: 75 min.

Stirring Used? No Etchant Volume: 400 cc

Etchant Heat Source: Water bath

Pretreatment Used: Water + detergent wash

Rinse Conditions: 5 min at room temperature, oven dry at 100 C

Counting Conditions

Pressure Applied: 100 grams/4.46 cm²

Electrode Material: Stainless steel Capacitance: 3000 pf Prespark Voltage: 1060 volts

Number of Prespark Counts: 1

Electrode Polarity: Positive

Counting Voltage: 560 volts

Number of Record Counts: 1

How Are Individual Counts Terminated? 5 seconds after last spark

Is the Foil Repositioned Between Counts? Yes

Is the Foil Cleaned Between Counts? No

System Performance

Counting Reproducibility: 10%

Maximum Linear Density: $>1000 \text{ sparks}/4.46 \text{ cm}^2$

Problem with foil that will not count without repositioning? None

Explanation:

Alpha Spark Counting

Experience: No

Plastic Used:

Source and Energy of Alphas:

Degree of Success:

General Comments on Spark Counting Experience:

Physical properties of spark counting are good for accelerator spectra.

SPARK COUNTING SURVEY

Laboratory: Lawrence Livermore Laboratory

Representative: Dick Griffith

Registration Plastic

Trade Name: Kimfol

Thickness: 6 micron, 10 micron

Exposure Conditions

Radiators: Cr^{252} , Thick U^{235}

Exposed Area: 3 cm^2

Plastic Support: Concentric rings during exposure and etching. Polycarbonate inner ring, stainless steel outer ring.

Plastic-Radiator Separation: Contact to 0.040"

Etching Conditions

Etchant: KOH Temperature: 40°C Concentration: 28% Etch Time: 2 hours

Stirring Used? No. Etchant Volume: $\frac{1}{2}$ liter.

Etchant Heat Source: Constant temperature oil bath (oil eliminates evaporation trouble).

Pretreatment Used: None.

Rinse Conditions: Hot water containing glass cleaner for about 3 minutes. Distilled water one hour. Oven dried - 80°C .

Counting Conditions

Pressure Applied: 370 gms/cm^2

Electrode Material: Gold plated brass. Capacitance: 100 pf Prespark Voltage: 800 v

Number of Prespark Counts: 3

Electrode Polarity: Positive

Counting Voltage: 500 v

Number of Record Counts: 5 to 10

How Are Individual Counts Terminated? 10 to 30 seconds preset time.

Is the Foil Repositioned Between Counts? No.

Is the Foil Cleaned Between Counts? No. Cleaning sometimes will re-entrain dirt in otherwise clear holes.

System Performance

Counting Reproducibility: $\sigma \pm 5\%$

Maximum Linear Density: 2000 tracks/cm²

Problem with foil that will not count without repositioning? Yes.

Explanation: Problem seems to be involved with debris shorting through the foil.

Alpha Spark Counting

Experience: Yes.

Plastic Used: Cellulose nitrate - NRDL formula.

Source and Energy of Alphas: Am²⁴¹ source, Li⁶(n, alpha) 2 Mev

Degree of Success: Poor, particularly for the n, alpha reactions. The background is quite high when the foil is etched long enough to detect the alphas.

General Comments on Spark Counting Experience:

There seems to be a certain degree of black art to spark counting, although work by Bill Cross, the Oak Ridge group and Dale Hankins has gone a long way toward shedding science on the problem. It is certainly a useful technique for measuring low track densities.

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