MASTER

Suggestions for Fast Neutron Dosimeter Techniques

by

P. R. Moran and E. B. Podgoršak
 Department of Physics
 University of Wisconsin
 Madison, Wisconsin

I. General Background

The interest in practical applications of fast neutrons is growing because of their possible use in radiotherapy (treatment of cancer), radiobiology and nuclear medicine. Neutrons are available from many different types of sources with energies from 10^{-4} to 10^3 MeV. The most important fast neutron sources at present are: (d,t) generators which produce 14 MeV neutrons, small cyclotrons with (d,n) neutrons of about 8 MeV, and nuclear reactors with fission spectrum neutrons. The production of neutrons is almost inevitably accompanied by X- and γ -radiation, which increases the radiation hazard of occupational personnel and complicates the absorbed dose measurements. The various neutron sources are capable of production of neutrons in a wide energy range and some of them can be used for production of monoenergetic neutrons. The dosimetry of neutrons with unknown energies is very complicated since the types of interaction of neutrons with matter vary with energy.

The neutrons are classified on energy intervals where certain interactions predominate. The lowest energy neutrons are called thermal neutrons and at "room temperature" their most probable energy is 0.025 eV. The dosimetry of this type of neutron is more developed than the dosimetry of other types of neutrons since the most important interaction of thermal neutrons with matter is capture which usually is accompanied

LEGAL NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

by the emission of γ -radiation and often the production of a radioactive nucleus. Neutrons in the energy range from about 0.5 eV to 10 KeV are called epithermal or intermediate neutrons. They are obtained by the slowing down of fast neutrons by elastic collisions with the atoms of the moderator. The intermediate neutrons interact with matter in various ways. Their absorption spectrum shows resonant peaks which means that at certain energies the probability of absorption is very high. The dosimetry of this type of neutron is of relatively low importance since they are rapidly slowed to thermal energies.

Fast neutrons have energies above 10 KeV and their most important interaction with matter is elastic scattering at lower energies and elastic and inelastic scattering and nuclear reactions at high energies. Fast neutrons with energies greater than 10 MeV are sometimes called relativistic neutrons. Since their energy exceeds the binding energy of nucleons their most important interactions with matter are nuclear reactions. The dosimetry of fast and relativistic neutrons is very complicated and relatively unsatisfactory (1).

There are two quantities which are most often used in neutron dosimetry: the absorbed dose and the neutron flux density from which the absorbed dose can be calculated. The absorbed dose is expressed in rads (100 ergs/gm) if the energy spectrum of the neutrons is known. This is a convenient unit since it is used for all types of ionizing radiations. The fact is that equal absorbed doses delivered by different kinds of ionizing radiations do not produce the same biological effect; consequently the concept of rem, which is defined as (rad) x (quality factor) was introduced. The quality factor (Q.F.) makes a qualitative correction for the relative biological effect. The main reason for different biological effect due to the same absorbed doses of radia-

tions of different types is in the energy deposited per unit length of the path of the ionizing particle. This is usually called the linear energy transfer (LET) of the particle. A Q.F. of 1 is proposed for all LET values up to $3.5 \text{ KeV}/\mu$. In the range from 3.5 to $175 \text{ KeV}/\mu$ the Q.F. is assumed to increase linearly from 1 to 20. For LET's greater than $175 \text{ KeV}/\mu$ the Q.F. is assumed to be 20.

The other commonly used quantity in neutron dosimetry is the neutron flux density, defined as the number of neutrons passing an area of 1 cm^2 in 1 second.

The accurate measurement of energy absorbed in biological materials by exposing them to fast neutrons is a difficult problem and depends on various factors:

- 1) energy spectrum and flux density of neutrons,
- 2) angular distribution of neutrons on the whole surface of the irradiated object,
- 3) additional undesired radiation accompanying neutrons,
- 4) physical state and chemical composition of the irradiated object (2).

There are several detection methods currently available for integrated fast neutron dosimetry. They all have serious limitations, but the most successful method has been the detection of radiation damage induced in a solid by fast neutron bombardment (e.g. tracks in film or etched tracks in insulators). Various techniques for detection of damage sites will be discussed in the next section. Some of the interactions of fast neutrons with matter have been extensively studied but not from a dosimetry viewpoint. The radiation damage of fast neutrons in solids and biological materials is very complex and although a large amount of research time has already been spent, the generalizations from simple examples

to general cases were not very successful.

Since the most important interaction of fast neutrons with matter is the elastic scattering from nuclei, and the amount of energy which can be transferred by a single elastic scattering event is inversely proportional to the mass of the scatterer, the hydrogen atom which has the lowest atomic mass and a large cross-section arising from a nearly bound singlet deuteron state, will be the most efficient scatterer for fast neutrons. From this fact it is obvious that the material used in fast neutron radiation damage studies should contain a lot of hydrogen atoms. This requirement agrees very well with the requirement for tissue equivalency of dosimeters. Since the human body has a large percentage of hydrogen in its composition, the material used in absorbed dose studies ought to have similar properties. Hence, a high hydrogen content dosimetric material has a double advantage; it is an efficient fast neutron scatterer and is suitably tissue equivalent.

The goal of fast neutron radiation damage studies is to find the defect sites produced by the interaction of neutrons with matter, identify them, count them and from these data obtain some information about the absorbed dose in the sample. The identification of defect sites is of interest in solid state physics for studies of interaction of radiation with matter, and the information about the absorbed dose has a great practical importance in radiation dosimetry.

On the average about half of the energy of the fast neutron is imparted to the recoil proton in the elastic scattering event. The binding energies of electron to proton and of the hydrogen atom in the molecule can be neglected in comparison with the energy of the fast neutron. In order to get a feeling for the number of damage sites pro-

duced in the exposed material, a simple calculation is quite revealing. Assume that a typical hydrocarbon sample weighing 1 gram is exposed to fast neutrons and that 1 rad of energy is absorbed in the sample.

$$1 \text{ rad} = 100 \text{ erg/gm} = 10^2 \text{ eV} / 1.6 \times 10^{-12} \text{ gm} = 0.5 \times 10^{14} \text{ eV/gm}$$

Since the minimum threshold for production of one damage site must be of the order of 10 eV, the absorbed dose of 1 rad can produce no more than 5×10^{12} damage sites per gram of material. On the other hand, since one gram of typical hydrocarbons contains about 10^{22} molecules, we can see that the concentration of damage sites following 1 rad is very low in comparison with the number of molecules in the sample, i.e., about one damage site per 10^{10} molecules. This is an upper bound calculation and the real relative concentration of damage sites is undoubtedly considerably smaller. That the calculation was made for the absorbed dose of 1 rad, which is a relatively high dose, means that the detection of even lower relative concentrations of damage sites is necessary if we want the proposed methods to be useful in personnel radiation dosimetry. For real systems, to measure an absorbed dose of 1 mrad, techniques would be required to detect at least 10^7 damage sites per gram with a background of about 10^{22} molecules per gram of the sample. We see that this means a detection of a relative concentration of 1 damage site per 10^{15} molecules which is by no means a simple task. Actually, of course 10^7 damage sites per gram is hardly a hopelessly small number; the problem is the masking effect of the abundant undamaged background, which is 10^{15} times as large in number. The key problem then, for fast neutron dosimetry, is to discover techniques which enable one to see only the damage sites with minimal interference of the background.

II. Detection Considerations

Concerned only with detectability, one wishes a detection system which minimizes randomly generated noise. Since room temperature thermal energies correspond to about 0.025 eV, this consideration implies detection phenomena of characteristic energies greater than tenths of electron volts; examples of techniques thus indicated are optical methods (such as those which are successful in various luminescence based dosimeters) and chemical reaction methods (such as those used in photographic development and etched track techniques). For best rare event sensitivity with any general method one should, if possible, seek non-subtractive measurement techniques in order to avoid the fluctuation noise characteristics inherent in any probe source. For example, suppose one performs an optical absorption experiment with a source of intensity I_0 having a fractional fluctuation Δf over the experimental measuring time. In an undamaged sample one would measure, therefore, a signal proportional to $[I_0 \times (1 \pm f)]$, and in a damaged sample where defect centers absorbed a small fraction, d , of the incident light one would measure a signal of $[(1-d) \times I_0 \times (1 \pm f)]$. The absorption signal is subtractive and sees the full incident intensity fluctuations; absolute signal is proportional to $(dI_0 \pm 2fI_0)$. Thus the signal to noise ratio is $(d/2f)$ so that, in order to detect the absorption due to defect centers one must stabilize the source so well that its fluctuations are smaller than the fractional absorption to be measured. In contrast, consider a fluorescence measurement where the fluorescence is photoexcited by an incident source of fluctuating intensity $[I_0(1 \pm f)]$. Suppose the same fractional absorption, d , occurs and, subsequent to each absorption event, a fluorescent event is measured. This fluorescence signal is non-

subtractive; fluorescence signal is proportional to $[dI_0 (1 + f)]$, and thus the signal to noise is $(1/f)$. For example, for a maximum defect center absorption cross-section in a typical solid state absorption band a few hundred Angstroms broad, even the one rad number of at most 10^{12} per cm^3 defect states in hydrocarbons would give a one cm absorption fraction of only about 2×10^{-4} . Consequently both the incident light source and sample quality would have to be stabilized to better than 10^{-4} in order barely to observe the absorption signal.

The following paragraphs outline some suggestions for radiation damage defect state detection based upon the considerations discussed above.

1. Photoexcitation Fluorescence

This phenomenon is utilized, for example, in silver activated metaphosphate (3) glass gamma radiation dosimeters where it is called radiophotoluminescence. Damage centers are frequently characterized by optical excitation and emission bands easily distinguished from those of the bulk material. For fast neutron dosimetry purposes, different classes of materials should be carefully surveyed for radiation induced excitation spectra, and fluorescence spectra. Some speculations on promising solids for this purpose are given in the final paragraphs of this section.

2. Metastable Exciton Properties

Many molecular organic solids exhibit long-lived mobile optical excitation states (4). Since these excitations may sample 10^{11} or 10^{12} different molecular sites during their lifetime, the probability that they would encounter a radiation damage site is relatively large. A number of interactions are possible; for example, the damage site may capture the exciton with subsequent luminescent emission characteristic of the

trapping site or the perturbation of the defect may relax selection rules enabling the exciton to decay optically in contrast to its usual radiationless decay mechanisms characteristic of the undamaged solid.

3. Photoconductivity

Damage centers may have optical excitations which produce free charge carriers in the normally insulating solid. Although most insulators have charge transport mobilities very small relative to the semiconductor materials, survey experiments for photoconductivity are easily performed as a routine along with other optical studies and should not be overlooked. Because of the solid state polarization problems characteristic of many insulators, one should use high frequency a.c. conductivity techniques and null departure detection methods combined with phase sensitive detection on optical modulation.

4. Raman Scattering

In the Raman effect, incident light is scattered with a frequency shift characteristic of the molecular vibrational properties of the scattering centers. With the availability of intense coherent laser incidence light sources, Raman scattering has recently been employed successfully in detailed impurity center studies (5) in solids. Raman scattering has many of the previously listed desirable detection features. First, damage centers will be characterized not only by distinguishable vibration frequencies, but also by the degree of electronic asymmetry upon which the Raman cross-section depends very sensitively. Second, it is an optical technique for which modern photomultipliers and counting methods give remarkable noise rejection. Third, it is non-subtractive because the incident probe light is at a frequency different from the detected

scattered light; for example, typical C-H or N-H bonds give Raman splitting of 2000 to 3000 cm^{-1} which guarantees ease of experimental discrimination against stray incident light. In addition, with coherent optical sources there are a variety of observable non-linear effects, e.g., stimulated Raman emission, and these could be used for direct investigation of LET characteristics of damage tracks.

5. Rayleigh Scattering

High LET radiation damage tracks provide a local microscopic perturbation of the solid's electronic polarizability. The damage geometry is small compared with an optical wavelength on the track cross-section, but long compared with optical wavelengths along the track. There would, therefore, exist a variety of directionally dependent Rayleigh scattering effects which might serve usefully, in principle, for dosimetry and LET investigations. It is a frequently used technique in lecture demonstrations, where one wants to demonstrate the path of a laser beam through plastic models of lenses and prisms, to subject the plastic to high level, high LET radiation doses. After irradiation, the beam path is easily visible to the students through the Rayleigh scattering transverse to the incident beam. This phenomenon should be quantitatively examined to determine its dosimetry potential.

III. Some Suggested Materials

1. Ammonium Salts

Many ammonium salts can be easily grown into large optical quality crystals. The protons on the NH_4 ion make these materials very hydrogen-rich inorganic solids and therefore good candidates for large fast neutron

cross-sections. In addition, ammonium→ammonia conversions due to energetic ionizations are certainly very likely. Ammonium sulphate is particularly interesting because of the possibility of sulphur neutron activation dose measurements in the same sample. Ammonium dihydrogen phosphate is another obvious candidate because of the strong electric effects associated with polar hydrogen bonds. Photoexcitation fluorescence, photoluminescence, and Raman scattering are all good analysis methods.

2. Silver Activated Ammonium Salts

Silver sulphate and silver phosphate have crystal structures very similar to the corresponding ammonium materials and, therefore, mixed crystals could perhaps be easily grown. The ammonium ions provide desirable fast neutron cross-sections and the silver has striking radiolysis reduction properties (as are apparent in radiophotoluminescence dosimeters for gamma dosimetry) to give stable radiation damage centers.

3. Cyclic Hydrocarbons

Anthracene, for example, can be grown into optical quality crystals and possesses many striking excitonic optical properties sensitive to impurity states. Preliminary experiments at this laboratory show that even such relatively unsensitive techniques as triplet exciton lifetime and annihilation fluorescence measurements do exhibit observable radiation damage changes. These materials also admit substantial diffusion of interstitial oxygen and are susceptible to radiolytic formation of quinones. Anthraquinone, for example, has a strong optical absorption in the blue-green and presumably associated optical emission properties.

4. Simple Grouped Organics

Examples are durene (tetramethyl benzene) and diphenyl. Both materials crystallize into optical quality samples, are widely used as host materials for other molecules or molecular fragments, and should be susceptible to single-bond rupture to produce a variety of C-H fragments as radiation damage centers. Such fragments would have characteristic photofluorencence properties with excitations likely in the near U.V. and far U.V. It is also likely that C-H fragment Raman scattering would be strong. Stable bond rupture centers induced by radiation are known to exist in covalently bonded inorganics (6) and analogous effects should exist in these simple organic crystals.

This work was supported in part through USAEC contract AT(11-1)-1105.

REFERENCES

1. Baarli, J. in Solid State Dosimetry, Amelinckx, S., Batz, B. and Strumane, R., ed., pp. 345-374, Gordon and Breach, New York (1969).
2. Sigurbjornsson, B., Smith, H. H. and Gustafsson, A. Neutron Dosimetry in Biology, In Pile Dosimetry - IAEA - Vienna (1965).
3. Becker, K. in Solid State Dosimetry, Amelinckx, S., Batz, B. and Strumane, R., ed., pp. 153-192, Gordon and Breach, New York (1969).
4. Avakian, P. and Merrifield, R. E., Molecular Crystals, Vol. 5, p. 37 (1968).
5. Callender, R. and Pershan, P., Phys. Rev. Letters, 23: 947 (1969).
6. Mitchell, E. W. J. in Solid State Dosimetry, Amelinckx, S., Batz, B. and Strumane, R., ed., pp. 261-322, Gordon and Breach, New York (1969).