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**NEUTRON DOSIMETRY (14 MeV) FOR FOIL SAMPLES
IRRADIATED WITH THE LAWRENCE LIVERMORE LABORATORY
ROTATING TARGET NEUTRON SOURCE**

Richard A. Van Konynenburg

April 17, 1973

Prepared for U.S. Atomic Energy Commission under contract No. W-7405-Eng-48

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NEUTRON DOSIMETRY (14 MeV) FOR FOIL SAMPLES IRRADIATED WITH THE LAWRENCE LIVERMORE LABORATORY ROTATING TARGET NEUTRON SOURCE

Abstract

The Lawrence Livermore Laboratory Rotating Target Neutron Source is being used to irradiate small metal and insulator samples to fluences of about 10^{17} 14-MeV neutrons/cm² as part of radiation-damage studies in support of the Controlled Thermonuclear Research materials effort.

Because of the characteristics of the neutron source, foil activation must be used for accurate dosimetry. Niobium is the material of choice for dosimetry. Present techniques yield an absolute accuracy of 7.5% in fluence for small samples.

Introduction

To obtain quantitative information about the effects of radiation on materials, one must devise methods of accurately determining the dose delivered to samples during experiments. Therefore, in support of materials studies for the Controlled Thermonuclear Reactor (CTR) project at Lawrence Livermore Laboratory, we developed a dosimetry technique for evaluating the fluence of (nominally) 14-MeV neutrons on metal foils and disc-

shaped insulator samples bombarded at the LLL Rotating Target Neutron Source (RTNS).

First, I will describe the neutron source itself, to provide a background for discussing dosimetry. Following this, I will discuss the choice of dosimetry material, analysis of activation and decay, monitoring for dose rate and gamma ray counting. Finally, I will estimate the absolute accuracy of this technique.

The LLL Rotating Target Neutron Source (RTNS)

D-T FUSION REACTION

The RTNS machine produces 14-MeV neutrons by fusion of deuterium and tritium¹:

When neutrons are the accelerated species, as is the case in the LLL-RTNS, the cross section for the reaction peaks at a value of about 5 barns, at a deuteron energy of about 110 keV.² At deuteron energies below 400 keV, the reaction is

isotropic in the center of mass system² and therefore somewhat anisotropic in the laboratory system. In particular, when a 200-keV deuteron interacts with a triton, the relative neutron intensity in the 180° lab direction is about 86% of that in the 0° direction.²

Although the energy of the neutron is nominally 14 MeV for low deuteron energies, the exact value depends on the deuteron energy and the angle between the path of the incoming deuteron and the path of the emitted neutron. In the RTNS the deuterons are accelerated to an energy of 490 keV and impinge on a target that is thicker than the deuteron range. Their energy is dissipated primarily by ionization and excitation of electrons. As they are being slowed by these processes, about 0.001% of the deuterons interact with tritons in the target, producing neutrons. The energy that a deuteron possesses when it undergoes fusion may therefore range from as much as 400 keV to as little as 15 keV or less. The most probable value of this energy depends on the bombarding energy, the energy-dependence of the $T(d,n)^4\text{He}$ cross section, the rate of energy loss in the target, and the tritium loading as a function of target depth. The most probable deuteron energy for a given accelerator can be determined by comparing the neutron energy as a function of angle with theoretically calculated values. This has been done for the LLL-RTNS, and the data are consistent with a most-probable reacting deuteron energy of about 200 keV.³ For a reacting deuteron energy of 200 keV, the calculated neutron energy ranges from 13.2 MeV at a laboratory angle of 180° to 15.1 MeV at an angle of 0°.⁴

DESCRIPTION OF MACHINE

The LLL Rotating Target Neutron Source has been described in detail elsewhere.⁵ However, the following rough outline is offered as a basis for understanding the need for dosimetry: An Insulated Core Transformer (ICT) accelerator produces a 5-10 mA beam of 400 keV deuterons. This beam passes through a collimator and impinges upon a rotating target composed of a thin layer of titanium deposited on a copper backing. The titanium contains tritium at an average concentration of 1.2 tritium atoms per titanium atom. The target is mounted on a cylinder that has a rotating seal,⁶ and it revolves at 1100 rpm. The target is cooled by water flowing over the copper backing through a thin stainless steel water spreader. The entire target assembly is surrounded by a thin water-catch cage.

GEOMETRY OF SOURCE

The actual localized source of neutrons consists of a disc-shaped region of the target, a few millimeters in diameter and of negligible thickness. The exact size, location, and variation of neutron yield across this spot are somewhat uncertain, and efforts are underway to characterize it more accurately.^{7,8} However, an approximate description can be obtained from a consideration of the collimation geometry and previous experiments that included film exposures,⁷ foil activation,⁹ and damage to lucite sample holders. The deuteron collimator in use at the time of writing has a 16-mm-diam aperture. The deuteron beam itself may have a

diameter as small as 3 mm or less; its variation in intensity with radius is not known, but probably could be approximately described by a Gaussian curve. The beam-deflecting magnets are tuned for maximum neutron count rate on a tightly collimated, pencil-shaped scintillation detector located outside the water-catch cage.⁷ During a long irradiation, the beam moves within the aperture and must be periodically retuned.⁸ The effective beam diameter is therefore probably larger than the instantaneous beam diameter but less than the diameter of the collimator aperture.

The distance between the plane of the target spot and the outside of the water-catch cage varies between 2.5 and 4 mm,⁹ depending on the alignment of the target and water spreader and on the force pressing the sample holder against the water-catch cage. A flexible sample-mounting assembly has been devised that will conform to irregularities in motion of the target backing while maintaining this distance as small as practicable.⁷

The uniformity of neutron yield across the target spot depends on the variation of current density across the deuteron beam and the variation of tritium loading in the target. Because the target is rotated and scanned radially, the latter factor should not be important, and the time-averaged neutron yield should correspond only to the beam-current density. As pointed out above, the beam also moves within its aperture, so that the effect of nonuniformities in beam-current density should also be somewhat smoothed out over the course of a long irradiation.

VARIATION OF FLUX WITH POSITION

Because of the small size of the neutron spot source, the flux decreases rapidly with distance away from it. The variation of flux with position relative to the source can be estimated by representing it as a uniform, isotropically emitting disc of radius a and zero thickness.^{9,10} The justification for assuming uniformity is given in the "Geometry of Source" section. The assumption of isotropy is valid enough for our calculations for small angular ranges near 0°; for larger angles, this assumption breaks down, both because of the intrinsic anisotropy (in the laboratory system) of the reaction itself and because of variation in attenuation by the target backing with change in projected length in the material. This varies with angle and becomes quite significant at 90°. Because of these approximations, and others to be discussed below, the absolute values indicated by these calculations should be regarded as somewhat approximate, but the relative trends should fit the actual situation.

For a point located on the axis of the deuteron beam at a distance Z downstream from the tritiated target, the flux is given by integration over the target spot as

$$\phi = \frac{Y}{4\pi a^2} \ln \left[1 + \left(\frac{a}{Z} \right)^2 \right], \quad (1)$$

where Y is the total neutron emission rate over the entire 4π solid angle in neutrons/sec. It can be seen that, at large distances, where $Z \gg a$, this expression reduces to $\phi = \frac{Y}{4\pi Z^2}$, as for a point source.

For a point located off the axis with coordinates (r, θ, Z) relative to the center of the target spot, the flux is given by

$$\phi = \frac{Y}{4\pi a^2} \ln \left[\frac{\sqrt{(a^2 + Z^2 - r^2)^2 + 4Z^2 r^2} + a^2 + Z^2 - r^2}{2Z^2} \right] \quad (2)$$

It can be seen that Eq. (2) reduces to Eq. (1) for $r = 0$.

Equation (1) is plotted in Fig. 1 (see Table 1 for detailed data) for the values $Y = 2 \times 10^{12}$ neutrons/sec and $a = 5$ mm. It should be noted that Y actually decreases with time as described in Ref. 5. The value chosen here is characteristic of a well-behaved, fresh target. Also, as noted above, the radius of the spot is not yet well-defined. The value $a = 5$ mm is chosen as lying between the probable instantaneous beam radius and the radius of the collimator aperture.

Table 1. Data for Fig. 1.

Z , mm	Flux, neutrons/(cm ² -sec)
2	1.26×10^{12}
3	8.47×10^{11}
4	5.99×10^{11}
5	4.42×10^{11}
6	3.36×10^{11}
7	2.63×10^{11}
8	2.10×10^{11}
9	1.71×10^{11}
10	1.42×10^{11}
11	1.20×10^{11}
12	1.02×10^{11}
16	5.94×10^{10}
20	3.86×10^{10}
30	1.74×10^{10}
40	9.88×10^9
51	6.09×10^9

Table 2. Data for Fig. 2.

r , mm	Flux, neutrons (cm ² -sec)		
	$Z = 4$ mm	$Z = 16$ mm	$Z = 51$ mm
0	5.99×10^{11}	5.94×10^{10}	6.09×10^9
1	5.90×10^{11}		
2	5.62×10^{11}	5.86×10^{10}	6.08×10^9
3	5.17×10^{11}		
4	4.57×10^{11}	5.63×10^{10}	6.06×10^9
5	3.91×10^{11}		
6	3.25×10^{11}	5.28×10^{10}	6.01×10^9
7	2.66×10^{11}		
8	2.17×10^{11}	4.86×10^{10}	5.95×10^9
9	1.79×10^{11}		
10	1.49×10^{11}	4.40×10^{10}	5.87×10^9
11			
12	1.06×10^{11}	3.94×10^{10}	5.78×10^9
13			
14	7.91×10^{10}	3.51×10^{10}	5.67×10^9

Figure 1 shows that the rate of decrease in flux is quite rapid in the range of closest possible approach to the source. This necessitates the use of a thin sample, to obtain a uniform dose. The importance of placing the sample as close as possible to the source to maximize the dose is also apparent. Finally, the uncertainty in the source-sample separation distance Z leads to a large uncertainty in the calculated dose that the sample receives. This has led us to adopt the technique discussed below for measuring the dose directly.

Figure 2 (see Table 2 for detailed data) shows a plot of Eq. (2) for representative values of Z and r , using the same values for Y and a as in Fig. 1. The successive values of Z are chosen so that the flux for $r = 0$ decreases in increments of about one order of magnitude from one Z value to the next. This plot shows that, using the distance of closest possible approach

Fig. 1. Calculated variation of flux with distance on-axis ($r = 0$).

(as is customary, to achieve the highest dose), one finds that the lateral uniformity is much decreased, compared to that at greater distances. For example, a 10-mm-diam sample located at $Z = 4$ mm receives a dose about 35% lower at the edge than at the center. This plot also shows that coaxial sample location becomes more critical at close source-sample separations.

In summary, these curves show that, if one desires to achieve the highest pos-

sible sample dose, still uniform across the sample and well-known in absolute value, one must use thin, disc-shaped samples with diameters less than or equal to about 10 mm. They must be placed as closely as possible to the back of the target with accurate axial alignment. Foil activation appears to be the best choice for dosimetry, preferably by using the activation of the sample itself to eliminate the necessity for geometrical and neutron-flux attenuation corrections. If this is

Fig. 2. Calculated variation of flux with distance off axis of source for three values of separation along axis. The multipliers of the values along the ordinate are shown on the curves.

not possible because of unfavorable half-life or cross section, a sandwich assembly with thin dosimetry foils in front and in back of the sample appears to be the best approach.

ENERGY SPECTRUM CLOSE TO SOURCE

Because samples are placed on the opposite side of the target from the im-

pinging deuteron beam, the maximum neutron energy occurs at an angle of 0° and is equal to 15.1 MeV. However, at distances Z that are small compared to the source diameter $2 \cdot a$, the neutron angle is not sharply defined, so that the sample is bombarded by neutrons having a spectrum of energies. In the case of a 10-mm-diam sample placed 4 mm from the source, the energy ranges from about

14.5 MeV to 15.1 MeV. For this reason, it is desirable that the cross section of the nucleus used for dosimetry possess little energy-dependence in this range; otherwise, it is necessary to calculate the spectrum from geometrical considerations (which are not well-defined) and integrate over the energy range.

PRACTICALLY ATTAINABLE FLUENCE AND TIME REQUIRED

To simulate CTR conditions, it is desirable to attain the highest possible neutron fluence. However, this must be balanced against the cost and time required. For the present accelerator with a 6-in. rotating target, we have somewhat arbitrarily concluded that a maximum practically attainable fluence is of the order of 10^{17} neutrons/cm². This can be achieved on small foil samples in

times of less than 2 weeks, operating with one 8-hr shift per working day. With around-the-clock operation, this fluence has been achieved in 3 days. Improvements are being made to increase the average flux, including increases in beam current and target diameter, and longer irradiation times could be contemplated if required. Under present conditions, however, the irradiations last at most a few days.

An activation product used for dosimetry should have a half-life of a few days. If it is much longer, the resulting activity will be reduced. If it is much shorter, scheduling of counting becomes critical. It should be noted that the saturation effect is not important in this work, because of the additional information supplied by the proton recoil telescope, as discussed below.

Choice of Dosimetry Material

In 1965, at a conference held in Texas,¹¹ a standardized dosimetry technique was adopted for use with 14-MeV neutron generators. This technique,¹² known as the "Texas convention," includes the use of copper discs and the reaction ${}^{63}_{29}\text{Cu}(n, 2n){}^{62}_{29}\text{Cu}$, which has a cross section of about 500 mbar at 14 MeV. The copper discs are bombarded for 1 min. The ${}^{62}_{29}\text{Cu}$ that is produced decays by positron-emission and has a half-life of 10 min. The 0.511-MeV gamma rays resulting from positron-annihilation are counted and the fluence is calculated, correcting for the duration of irradiation and the time of decay.

This convention is particularly useful for neutron generators in which the flux is low and the irradiation time must be short, but it has the disadvantages of a cross section that is rather energy-dependent¹³ and a short half-life that necessitates counting inconveniently soon after the irradiation. Hence, we have chosen to base our dosimetry on the activation of another nuclear species. After considering other candidates, we selected niobium and are using the reaction

The ${}^{92}_{41}\text{Nb}$ decays primarily by electron

capture¹⁴ to $^{92}_{40}\text{Zr}$, emitting gamma rays, which are counted after the irradiation. There are several advantages to this choice:

- The half-life of $^{92}_{41}\text{Nb}$ is 10.16 $\pm$ 0.03 days,¹⁵ which is in the optimum range for convenience and high induced counting rates.
- The cross section is relatively large and fairly energy-independent in the range of interest. Also, it recently was measured¹⁶ to an estimated absolute accuracy of 5%, based upon the recent evaluation¹⁷ of the $^{27}_{13}\text{Al}(n, \alpha)^{24}_{11}\text{Na}$ cross section to an estimated absolute accuracy of better than 1%. The value of the niobium cross section ranges from 498

$\pm$ 25 mbar at 14.10 MeV to 487 ± 24 mbar at 14.80 MeV.

- The gamma rays emitted¹⁴ by $^{92}_{41}\text{Nb}$ have energies of 0.934 and 0.912 MeV, making them relatively easy to detect and count.
- Niobium occurs naturally as a single isotope,¹⁴ and the cross sections and half-lives of competing reactions are such that there are no interferences.
- Niobium is relatively inexpensive (i.e., it is not a precious metal).
- Niobium is a metal of interest to the CTR materials research program, so that, when it is the object of study, it can serve as its own dosimeter.

Analysis of Activation and Decay

As is well known, the net rate of production of activated nuclei is given by

$$\frac{dN^*}{dt} = N\phi\sigma - \lambda N^*, \quad (3)$$

where N^* is the number of activated nuclei, N is the number of stable nuclei (assumed constant), ϕ is the flux in neutrons/(cm²-sec), σ is the cross section in square centimeters, and λ is the decay constant of the activated species in sec⁻¹, which in turn is equal to $0.693/t_{1/2}$, where $t_{1/2}$ is the half-life in seconds.

In our case, ϕ varies as a function of time over the duration of the irradiation, because of accelerator shutdowns, tritium depletion of the target, and variations in accelerator tuning. However, the use of a proton recoil telescope as described in the next section enables one to obtain a record of the relative neutron output as a

function of time. Assuming that the source-sample geometry and counter response remain constant, the flux at the sample is given by

$$\phi(t) = KR(t), \quad (4)$$

where $R(t)$ is the telescope output and K is a constant to be determined for each irradiation, because the geometry may vary from one to the next.

It is desired to obtain the total fluence, which is given by

$$\Phi = \int_0^{t_f} \phi(t) dt = K \int_0^{t_f} R(t) dt, \quad (5)$$

where t_f is the duration of the irradiation.

Equation (3) is integrated¹⁸ by substituting Eq. (4) for ϕ and setting N^* equal to $X(t) e^{-\lambda t}$, where $X(t)$ is an integrating device. The constant K is then found to be given by

$$K = \frac{N^*(t_f)}{N\sigma \int_0^{t_f} R(t) e^{\lambda(t-t_f)} dt} \quad (6)$$

The fluence can be expressed [from Eqs. (5) and (6)] as

$$\Phi = \frac{N^*(t_f) \int_0^{t_f} R(t) dt}{N\sigma \int_0^{t_f} R(t) e^{\lambda(t-t_f)} dt} \quad (7)$$

The data produced by gamma ray counting are in terms of dN^*/dt at the time of counting, t_c . Since

$$\left(\frac{dN^*}{dt}\right)_{t_c} = -\lambda N^*(t_c) \quad (8)$$

and

$$N^*(t_c) = N^*(t_f) e^{-\lambda(t_c-t_f)}, \quad (9)$$

we find that

$$N^*(t_f) = -\frac{1}{\lambda} \left(\frac{dN^*}{dt}\right)_{t_c} e^{\lambda(t_c-t_f)}, \quad (10)$$

so that the final expression for the fluence is

$$\Phi = -\frac{\left(\frac{dN^*}{dt}\right)_{t_c} e^{\lambda(t_c-t_f)} \int_0^{t_f} R(t) dt}{\lambda N\sigma \int_0^{t_f} R(t) e^{\lambda(t-t_f)} dt} \quad (11)$$

The integral in the numerator is equal to the total accumulated count from the proton recoil counter. The integral in the denominator is evaluated by summing the contributions of several time segments; within each of them, R is assumed constant. This has proven to be a good approximation over time intervals of 8 hr. The quantity $(dN^*/dt)_{t_c}$ is evaluated by gamma ray counting, as discussed below. The constant λ is taken¹⁵ as $2.843 \times 10^{-3} \text{ hr}^{-1}$. The number N is evaluated by weighing the foil before irradiation and using the relation

$$N = \frac{6.0248 \times 10^{23} m}{A}, \quad (12)$$

where m is the mass in grams and A is the atomic mass of Nb ($= 92.906$). The cross section σ is taken as 480 mbarn; this is obtained by extrapolating the data from Ref. 15 to 15.11 MeV and taking an average value for the cross section between 14.5 and 15.11 MeV, weighted toward the higher energies. Although this is admittedly an approximation, it is believed to be valid in view of the fact that the measured cross section varies only from 498 to 487 mbarn over the energy range from 14.10 to 14.80 MeV.

Proton Recoil Telescope

A proton recoil telescope is used to provide a continuous count that is directly proportional to the total neutron output (design of such detectors has been described elsewhere¹⁹). The principle of operation is as follows: The 14-MeV neutrons encounter a polyethylene film

in the front of the telescope and elastically scatter protons out of it. Some of the protons pass through a proportional counter and into a scintillation detector, which are operated in coincidence. This ensures that counts due to scattered radiation and background do not appear

in the output. The efficiency can be calculated from knowledge of the telescope geometry, the composition and thickness of the film, and measured values of the differential n-p scattering cross section.

In the present installation, the recoil telescope is located at a laboratory angle of 120° from the deuteron beam, at a distance of 1.2 m from the target spot. At this distance, motion of the spot has a negligible effect on the counting rate. The geometry and efficiency are such that a total count of 1.66×10^8 represents a dose of approximately 10^{17} neutrons/cm² on a foil

placed at the distance of closest approach.

The accumulated count from the telescope is recorded every hour. The results are plotted and approximated by a series of straight line segments and are used in Eq. (11). Because of the information supplied by this telescope, saturation with respect to the formation of $^{92}_{41}\text{Nb}$ does not increase the uncertainty in the calculated fluence, even for irradiation times that are long compared to its half-life, as long as the counting efficiency remains stable. The long-term drift of the telescope and counting system efficiency is believed to be less than 5%.

Gamma Ray Counting

Gamma ray counting is done by personnel of the LLL Radiochemistry Division (see Acknowledgments), using a computer-controlled Ge(Li) detector. Corrections are made for self-absorption in the foils, and the geometry is chosen to minimize coincidence losses. Counting

periods are normally about 30 min, and time at which the count is performed, t_c in Eq. (11), is taken as the mid-time. The absolute accuracy is about 1%, including statistical uncertainty and uncertainties due to geometry and detector calibration.²⁰

Estimate of Absolute Accuracy

The two largest possible sources of error in this work are the use of the measured $^{93}_{41}\text{Nb}(n, 2n)^{92}_{41}\text{Nb}$ cross section, estimated to be accurate to 5%,¹⁶ and the drift in counting efficiency of the proton recoil telescope, estimated at less than 5%.⁷ In addition, the gamma ray counting accuracy²⁰ is about 1%, and errors due to weighing are about 0.5% for the smallest foils used. Uncertainty in the accuracy of the tabulated half-life of $^{92}_{41}\text{Nb}$ is 0.3%.¹⁵

Errors due to measurement of time and linearization of the accumulated count data are considered negligible in comparison. The absolute accuracy of the average sample dose is estimated to be 7.5%. It should be noted that variation in dose over the lateral dimension of a foil sample can exceed this, as shown in Fig. 2. To be a more accurate measure of dose at various lateral positions, a dosimetry foil could be cut and counted

in smaller segments; so far, it has been unnecessary to do this. When a stack of samples is used, it is necessary to correct for the shielding effect of the mate-

rial up-beam from the sample in question. This is done by means of the cross sections for effective removal of neutrons.¹²

Summary

It has been shown that foil dosimetry is necessary for accurate evaluation of the 14-MeV neutron fluence of samples irradiated close to the target of the LLL Rotating Target Neutron Source. It has also been shown that niobium foils offer

particular advantages for the fluences and irradiation times being used in radiation-damage studies for CTR materials research. The resulting procedure yields an absolute accuracy in fluence measurement of 7.5% for small samples.

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