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# Standard Practice for Conducting Irradiations at Accelerator-Based Neutron Sources<sup>1</sup>

This standard is issued under the fixed designation E798; the number immediately following the designation indicates the year of original adoption or, in the case of revision, the year of last revision. A number in parentheses indicates the year of last reapproval. A superscript epsilon ( $\varepsilon$ ) indicates an editorial change since the last revision or reapproval.

## 1. Scope

1.1 This practice covers procedures for irradiations at accelerator-based neutron sources. The discussion focuses on two types of sources, namely nearly monoenergetic 14-MeV neutrons from the deuterium-tritium T(d,n) interaction, and broad spectrum neutrons from stopping deuterium beams in thick beryllium or lithium targets. However, most of the recommendations also apply to other types of accelerator-based sources, including spallation neutron sources (1).<sup>2</sup> Interest in spallation sources has increased recently due to their proposed use for transmutation of fission reactor waste (2).

1.2 Many of the experiments conducted using such neutron sources are intended to simulate irradiation in another neutron spectrum, for example, that from a DT fusion reaction. The word simulation is used here in a broad sense to imply an approximation of the relevant neutron irradiation environment. The degree of conformity can range from poor to nearly exact. In general, the intent of these simulations is to establish the fundamental relationships between irradiation or material parameters and the material response. The extrapolation of data from such experiments requires that the differences in neutron spectra be considered. area to standards/sist/1780d311

1.3 The procedures to be considered include methods for characterizing the accelerator beam and target, the irradiated sample, and the neutron flux and spectrum, as well as procedures for recording and reporting irradiation data.

1.4 Other experimental problems, such as temperature control, are not included.

1.5 The values stated in SI units are to be regarded as standard. No other units of measurement are included in this standard.

1.6 This standard does not purport to address all of the safety concerns, if any, associated with its use. It is the

responsibility of the user of this standard to establish appropriate safety and health practices and determine the applicability of regulatory limitations prior to use.

# 2. Referenced Documents

- 2.1 ASTM Standards:<sup>3</sup>
- C859 Terminology Relating to Nuclear Materials
- E170 Terminology Relating to Radiation Measurements and Dosimetry
- E181 Test Methods for Detector Calibration and Analysis of Radionuclides
- E261 Practice for Determining Neutron Fluence, Fluence Rate, and Spectra by Radioactivation Techniques
- E263 Test Method for Measuring Fast-Neutron Reaction Rates by Radioactivation of Iron
- E264 Test Method for Measuring Fast-Neutron Reaction Rates by Radioactivation of Nickel
- E265 Test Method for Measuring Reaction Rates and Fast-Neutron Fluences by Radioactivation of Sulfur-32
- E266 Test Method for Measuring Fast-Neutron Reaction Rates by Radioactivation of Aluminum
- E393 Test Method for Measuring Reaction Rates by Analysis of Barium-140 From Fission Dosimeters
- E854 Test Method for Application and Analysis of Solid State Track Recorder (SSTR) Monitors for Reactor Surveillance, E706(IIIB)
- E910 Test Method for Application and Analysis of Helium Accumulation Fluence Monitors for Reactor Vessel Surveillance, E706 (IIIC)

## 3. Terminology

3.1 Descriptions of relevant terms are found in Terminology C859 and Terminology E170.

# 4. Summary of Existing and Proposed Facilities

4.1 T(d,n) Sources:

4.1.1 Neutrons are produced by the highly exoergic reaction  $d + t \rightarrow n + \alpha$ . The total nuclear energy released is 17.589

<sup>&</sup>lt;sup>1</sup>This practice is under the jurisdiction of ASTM Committee E10 on Nuclear Technology and Applicationsand is the direct responsibility of Subcommittee E10.08 on Procedures for Neutron Radiation Damage Simulation.

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 $<sup>^{2}</sup>$  The boldface numbers in parentheses refer to a list of references at the end of this practice.

<sup>&</sup>lt;sup>3</sup> For referenced ASTM standards, visit the ASTM website, www.astm.org, or contact ASTM Customer Service at service@astm.org. For *Annual Book of ASTM Standards* volume information, refer to the standard's Document Summary page on the ASTM website.

MeV, resulting in about a 14.8-MeV neutron and a 2.8-MeV alpha particle at low deuterium beam energies (3). The deuteron energy (generally 150 to 400 keV) is chosen to maximize the neutron yield (for a particular target configuration) from the resonance in the d-t cross section near 100 keV. The number of neutrons emitted as a function of angle ( $\theta$ ) between the neutron direction and the incident deuteron beam is very nearly isotropic in the center-of-mass system. At a deuteron energy of 400 keV in the laboratory system, the neutron flux in the forward direction is about 14 % greater than in the backward direction, while the corresponding neutron energy decreases from 15.6 to 13.8 MeV (4). In practice, the neutron field also depends on the gradual loss of the target material and the tritium deposition profile. Detailed calculations should then be made for a specific facility.

4.1.2 The flux seen at a point (r,  $\theta$ , z) in cylindrical coordinates from a uniform T(d,n) source of diameter *a* is given by the following (5):

$$\varphi(r,\,\theta,\,z) = \frac{Y}{4\pi a^{2}} \ln\left\{\frac{\left(k^{4} + 4r^{2}z^{2}\right)^{1/2} + k^{2}}{2z^{2}}\right\}$$
(1)

where:

 $k^2 = a^2 + z^2 - r^2$ , and Y = the total source strength.

For z >> a and r = 0 (on beam axis) this reduces to  $Y/4\pi z^2$ , as expected for a point source. The available irradiation volume at maximum flux is usually small. For a sample placed close to the target, the flux will decrease very rapidly with increasing radial distance off the beam axis. However, since the neutron energy is nearly constant, this drop in flux is relatively easy to measure by foil activation techniques.

4.1.3 Other existing sources, such as Cockroft-Walton type accelerators, are similar in nature although the available neutron source strengths are much lower. 4.1.4 Rotating Target Neutron Source (RTNS) I and II (5-7)—RTNS I and II, which formerly were operated at the Lawrence Livermore National Laboratory, provided 14 MeV neutron source strengths of about  $6 \times 10^{12}$  and  $4 \times 10^{13}$  neutrons/s, respectively. Although these facilities have been shut down, they were the most intense sources of 14 MeV neutrons built to date for research purposes. They are discussed here because of their relevance to any future neutron sources. Their characteristics are summarized in Table 1. A discussion of similar sources can be found in Ref (8). The deuteron beam energy was 400 keV and the target was a copper-zirconium alloy (or copper with dispersed alumina) vapor-plated with tritium-occluded titanium. The beam spot size was about 10 mm in diameter. In addition to being rotated, the target also was rocked every few hours and the deuteron beam current was increased slowly in an attempt to maintain a constant flux in spite of tritium burn-up in the target. Samples could be placed as close as 2.5 to 4.0 mm from the region of maximum d-t interaction resulting in a typical flux of  $10^{13}$  n/cm<sup>2</sup>·s over a small sample. The neutron fields were well characterized by a variety of methods and the absolute fluence could be routinely determined to  $\pm 7$  %. Calculated neutron flux contours for RTNS-II are shown in Fig. 1.

4.2 *Be or Li(d,n) Sources* (9):

4.2.1 When a high-energy (typically 30- to 40-MeV) deuteron beam is stopped in a beryllium (or lithium) target, a continuous spectrum of neutrons is produced extending from thermal energies to about 4 MeV (15 MeV for lithium) above the incident deuteron energy (see Figs. 2-4). In existing facilities, cyclotrons with deuteron beam intensities of 20 to 40  $\mu$ A provide neutron source strengths in the range of 10<sup>13</sup> n/s, using solid beryllium targets with water cooling. A more intense source (>10<sup>16</sup> n/s) is now being designed employing liquid lithium targets. In the remainder of this document the term Be(d,n) source is meant as a generic term including Li(d,n) sources, whether solid or liquid targets.

4.2.2 Neutrons are produced by several competing nuclear reaction mechanisms. The most important one for radiation damage studies is the direct, stripping reaction since it produces almost all of the high-energy neutrons. When the incident deuteron passes close to a target nucleus, the proton is captured and the neutron tends to continue on in a forward direction. The high energy neutrons are thus preferentially emitted in the direction of the incident deuteron beam. However, as the deuterons slow down in the target, lower energy neutrons will be produced with angular distributions that are much less forward peaked. Furthermore, when the residual nucleus is left in an excited state, the angular effects are also much less pronounced. These latter two effects tend to decrease the average neutron energy at angles other than  $0^{\circ}$  in the direction of the beam.

4.2.3 Neutrons can also be produced by compound nuclear reactions in which the entire deuteron is captured by the target nucleus and neutrons are subsequently evaporated. Neutrons are preferentially emitted with energies less than a few MeV and the angular distribution approaches isotropy at neutron energies below 1 MeV. Neutrons also are produced by deuteron

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Facility	Availability	Beam	Target	Source Strength, n/s	Maximum Flux at Sample, n/cm <sup>2</sup> ·s	Experimental Volume for Maximum Flux, cm <sup>3</sup>
RTNS I	No longer available	400 keV d	t	6 × 10 <sup>12</sup>	>10 <sup>12</sup>	0.2
RTNS II	No longer available	400 keV d	t	4 × 10 <sup>13</sup>	>10 <sup>13</sup>	0.2
					>1012	5.0
Existing Be or Li(d,n)	U.C. Davis Cyclotron <sup>A</sup>	30–40 MeV d	Solid Be or Li	~10 <sup>13</sup>	>1012	~1.0
Proposed Li(d,n)	Conceptual design (9)	30–40 MeV d	Liquid Li	3 × 10 <sup>16</sup>	>10 <sup>15</sup>	10.0
			-		>10 <sup>14</sup>	600.0

<sup>A</sup> This is the only existing facility that has been well characterized and is readily available, although other facilities can be used.



NOTE 1—Flux contours assume a symmetric, Gaussian beam profile. Figure from Ref. (5). FIG. 1 Flux Contours for RTNS II



NOTE 1—Neutron spectra as a function of energy and angle for  ${}^{9}Be(d,n)$  source at ORNL,  $E_d = 40$  MeV. (Data from Ref (8).)



break-up, in which the deuteron simply breaks apart in the Coulomb field of the nucleus, although this effect is very small for low-Z materials.

4.2.4 The neutron spectrum thus depends very strongly on the angle from the incident deuteron direction, and the flux is very sharply peaked in the forward direction (see Fig. 2). Materials studies for which the maximum total neutron fluence is desired are usually conducted close to the target and may subtend a large range of forward angles (for example, 0 to  $60^{\circ}$ ). This practice primarily will be concerned with this closegeometry situation since it is the most difficult to handle properly. 4.2.5 Other factors can also influence the neutron field during a particular irradiation, especially beam and target characteristics, as well as the perturbing influence of surrounding materials. At present, these facilities have not been completely characterized for routine use. In particular, some uncertainties exist, especially at low (<2 MeV) and high (<30 MeV) neutron energies, since these regions are either difficult to measure with existing techniques, or the required nuclear data are insufficient. In these cases, neutron dosimetry data should be reported directly to allow reanalysis as procedures and nuclear data improve in the future.

4.2.6 Existing Sources:

4.2.6.1 Whereas virtually any deuteron accelerator with reasonable energy and intensity can be used as a neutron source, only two facilities have been used routinely for materials effects irradiations, namely the cyclotrons at the University of California at Davis (10) and at Oak Ridge National Laboratory (11,12). Typical flux-spectra obtained are shown in Figs. 2-4 (9,11,13), and typical characteristics are listed in Table 1.

4.2.6.2 Since the neutron flux and spectral gradients are so steep, experimenters are faced with the problem of nonuniform irradiations over their samples unless specimen sizes are severely limited. Alternatively, the field gradients may be moderated by deliberately moving or enlarging the beam spot on the target. This technique will result in a lower total fluence as well as a lower average neutron energy for a small-size sample on the beam axis, although larger samples will not be so severely affected and may in fact show an overall improvement in average fluence and neutron energy.

4.2.6.3 At present, the neutron field can be determined reasonably well at existing facilities. The flux-spectrum can be measured to within  $\pm 10$  to 30 % in the 2- to 30-MeV energy region where about 90 % of neutron damage is initiated (assuming  $E_d = 30$  to 40 MeV). Highly accurate ( $\pm 10$  %) time-of-flight spectrometry has been used to study the field far from the source, except for the energy region below a few MeV (11). However, close geometry irradiations must rely on passive dosimetry with larger errors due to uncertainties in the nuclear cross sections, especially above 30 MeV (12).

4.2.7 Conceptual Design for Li(d,n) Source (9,14)—A conceptual design for a fusion materials irradiation facility was done at the Hanford Engineering Development Laboratory (HEDL). The design consisted of a high-current (100-mA) deuteron accelerator and a liquid lithium target. This was expected to produce a neutron source strength of about  $3 \times 10^{10}$  n/s (14). The designs called for a wide-area beam spot on the target (for example, 3 by 1 cm), thereby moderating the steep neutron field gradients in close geometry. Neutron fluxes up to  $10^{15}$  n/cm<sup>2</sup>·s could be produced over a volume of several cubic centimetres, allowing much larger samples than with present sources. This facility would thus have a higher flux of high-energy neutrons over a larger volume than any available accelerator source. A more recent design that takes advantage of improvements in accelerator technology is discussed in Ref (15).

4.3 Other Sources:



Note 1—The maximum occurs at about 40% of the deuteron energy. (Data from Ref (6).) FIG. 3 Li(d,n) Spectra at 0° as a Function of Deuteron Energy

4.3.1 There are many other accelerator-based neutron sources available, generally having lower neutron energy and flux. Most are used for medical or nuclear research applications. Van de Graaffs and cyclotrons have also been used with many other nuclear reactions such as  $d(d,n)^3$ He and  $^7Li(n, p)^7$ Be. Facilities with much higher charged particles such as the Intense Pulsed Neutron Source (IPNS) (16) and the Los Alamos Meson Physics Facility (LAMPF) (17,18) have also been used. For example, the IPNS neutron flux spectrum is shown in Fig. 5 (16). A new irradiation facility has been brought on-line at the LAMPF (18). The primary objective of this facility is to study the basic aspects of radiation effects as

produced by medium energy protons and neutrons that are born through spallation reactions as the protons interact with the target nuclei. Another objective is to study radiation damage to structural and detector materials used with accelerators. A description of the facility is given in Ref (19). The available neutron flux and spectrum are described by the results of calculations (20) and foil activation measurements (21). Radiation damage parameters for the facility have also been calculated (22). In the case of facilities such as LAMPF and IPNS, the dosimetry and damage analysis must take into account the presence of very high-energy neutrons (>40 MeV), as well as a small flux of charged particles.



Note 1—Forward (0°), thick target neutron yield above 2 MeV from the  $^9Be(d,n)$  reaction as a function of deuteron energy.



4.3.2 The procedures recommended in this work also apply to these other sources and should be used where applicable. However, the experimenter should always be aware of the possibility of additional problems due to peculiarities of individual sources.

#### 5. Characterization of Irradiation Environments

5.1 *Scope*—The methods used to define the flux, fluence, and spectra precisely in accelerator environments are significantly different from those used in reactor environments. The reason for this difference is that, whereas reactors generally produce stable fields with gentle gradients, accelerators tend to produce fields with very sharp spatial flux and spectral gradients, which may vary over short time intervals and may not scale linearly with beam current. For example, small changes in accelerator tuning can move the spatial location of the neutron source relative to the irradiated sample, thereby changing the flux and spectrum. Consequently, it is critically

important to follow well established and well calibrated procedures in order to measure adequately the irradiation exposure parameters. Otherwise, it will be impossible to correctly calculate damage parameters such as DPA or to correlate materials effects measured at different facilities.

5.2 System Parameters—In the following section it is important to distinguish between T(d,n) (14-MeV) sources and broad spectrum <sup>9</sup>Be(d,n) sources. Whereas both types of sources exhibit strong flux gradients, only the broad-spectrum sources exhibit significant spectral gradients. Consequently, in the following subsections it should be understood that references to flux measurement refer to both facilities, whereas references to spectral measurement refer only to the <sup>9</sup>Be(d,n) sources.

5.2.1 Beam Characterization—It is important to realize that virtually any change in the accelerator beam will produce some alteration of the neutron field. Two classes of instabilities can be defined according to whether they affect only the neutron flux or the neutron spectrum as well. Whereas the flux may vary independently of the spectrum, spectral changes always imply a change in flux. Flux changes are usually easy to measure and to account for in calculating total exposure or damage rates (see 5.3). However, spectral changes are much harder to measure or to account for in subsequent calculations. For example, if the spectrum changes significantly even once during a long run, then activated foils with short half-lives may indicate an average spectrum that is quite different from that indicated by foils with long half-lives. Furthermore, it may be impossible to account for this difference unless great care is exercised to record the pertinent beam information, namely beam current, beam energy, and spatial alignment.

5.2.1.1 *Flux Instabilities*—The most important sources of flux instability are the beam current and target condition. If the beam is well collimated, stable in energy, and stable in spatial position, then the flux should be directly proportional to the beam current, neglecting target effects. At solid Be(d,n) sources, target effects are usually unimportant. However, at T(d,n) sources, time-dependent changes in the target are the dominant cause of flux instabilities (6). The beam current should be read using a Faraday cup or well insulated target assembly where possible. The current-sensing equipment should be checked for beam leakage, linearity, and long-term stability. The output should then be recorded at regular time intervals.

5.2.1.2 Flux and Spectral Instabilities—A change in the beam energy will alter both the flux and spectrum, although most accelerators have active means of keeping the beam energy constant within relatively small preset limits. It is worth mentioning that beam stability is often linked to beam current since beam control systems may use slits or aperatures which in turn limit the transmission through the machine. Hence, attempts to maximize the beam current may allow a wider range of particle trajectories, resulting in a larger energy spread as well as poorer spatial definition. The experimenter should be aware of these problems and check that the energy stability, beam current monitoring, and target integrity are adequate. The most important source of spectral instability at broad-spectrum sources is the movement of the beam on the target (at T(d,n)



NOTE 1—Neutron flux spectrum at the Intense Pulsed Neutron Source of ANL with 500 MeV protons and a depleted uranium target. The solid line is calculated and the dashed is an adjusted spectrum based on radiometric dosimetry. (Data from Ref (12).)

FIG. 5 Neutron Flux Spectrum at the Intense Pulsed Neutron Source

sources this will only significantly affect the flux). Collimation aperatures are generally used to define the beam size and location. It is again important to note that attempts to maximize the beam current and hence the flux may result in unacceptably large variations in beam spot size and location on the target. The collimation system should thus be analyzed to predict the maximum possible variations. This can be translated into flux/spectral information by examining measured angular distribution data. For example, at a deuteron beam energy of 30 MeV on a Be target, the total flux falls a factor of two as the angle from the beam axis changes from  $0^{\circ}$  to only  $10^{\circ}$  (23). At a close irradiation distance of about 0.5 cm, this would correspond to a change in the beam spot location of only 1 mm. Beam spatial alignment and stability are thus crucial to the characterization of an irradiation. Active and passive methods of measuring flux and spectral instabilities are covered in 5.3.2 and 5.3.3.

### 5.2.2 Target Characterization:

5.2.2.1 Physical characteristics of the target assembly are also vitally important in determining the neutron field. The design of the target will strongly influence the field produced and instabilities in the target can lead to large variations in the flux and spectra. In order to understand these effects, it is important to understand neutron production in the target. Well designed targets are thick enough to stop the deuteron beam. This can be checked with any standard range-energy table such as Refs (24,25). However, improper target design may cause the target to burn up during exposure, leading to drastic alterations of the neutron field. Such catastrophic failures are easily seen by remote sensing systems (see 5.2.6.2).

5.2.2.2 As the deuteron beam is stopped in the target, it interacts with the tritium or beryllium, as discussed previously.

For T(d,n) sources the primary cause of concern is the burn-out and boil-off of tritium and slow build-up of deuterium (see Fig. 6). The former causes a reduction in flux but no significant difference in the geometric source specification. The latter can lead to neutron production from the  $d(d,n)^3$ He reaction, although this contribution is generally negligible since massive exposures are required to build up significant deuterium in the target and the neutron production cross section is much smaller than from tritium. At the RTNS, these effects were well understood. Remote neutron detectors were used to continuously monitor the target condition and the target was then slowly rocked in position in an attempt to maintain a nearly constant neutron flux. The experimenter could thus obtain an accurate time history of the neutron exposure.

5.2.2.3 More complex target problems are encountered at Be(d,n) facilities. The amount of material that backs the active Be region as well as the surrounding support material will attenuate or scatter the neutrons, probably accounting for some differences in the low-energy neutron flux reported at different facilities. On the other hand, backing material cannot be too thin or high energy protons from (d,p) reactions may escape from the target and irradiate the specimen. The lifetime of a beryllium target is not well established, although experience at U. C. Davis indicates that they should be able to withstand deuteron exposures of at least 200 C/cm<sup>2</sup>. However, if target cooling is inadequate, the beryllium may evaporate or melt within a matter of minutes. Such failures are readily apparent by sudden changes in the neutron flux. A more serious concern is the slow erosion of the beryllium since this leads to a gradual change in the location of the source in the beryllium and may produce perturbations in the flux and spectrum at close geometries. Passive in situ dosimetry should be able to