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A NEW METHOD FOR FAST NEUTRON DOSIMETRY WITH A CIRCULATING LIQUID

D.L. SMITH

Technology Development Division, Argonne National Laboratory
Argonne, Illinois 60439, U.S.A.
E-mail: Donald.L.Smith@anl.gov

Y. IKEDA, Y. UNO and F. MAEKAWA

Department of Reactor Engineering, Japan Atomic Energy Research Institute
Tokai-mura, Ibaraki-ken 319-11, Japan

E-mail: Ikeda@fnshp.tokai.jaeri.go.jp, Uno@fnshp.tokai.jaeri.go.jp, Fujio@fnshp.tokai.jaeri.go.jp

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ABSTRACT

A new approach to fast-neutron dosimetry is described. It involves circulating a liquid with dissolved chemicals between a localized neutron field and a remote area where neutron-induced radioactivities are measured continuously. This approach employs reactions which generate relatively short-lived radioactive byproducts. Sensitivity to the neutron energy spectrum is achieved by choosing several reactions with differing thresholds. Neutron field strengths and their time variations can be derived from experimental data using the known radioactive-decay properties, cross sections, detector efficiencies, liquid flow rates and composition of the liquid. The concept has been demonstrated by measurements which utilized an aqueous solution of yttrium chloride hexahydrate, a 14-MeV neutron generator and Ge gamma-ray detectors. The $^{16}\text{O}(n,p)^{16}\text{N}$, $^{37}\text{Cl}(n,p)^{37}\text{S}$ and $^{89}\text{Y}(n,n')^{89\text{m}}\text{Y}$ activation reactions served as dosimeters.

1. Introduction

In active dosimetry, neutron spectrometers (e.g., proton-recoil or fission detectors) placed in the radiation environment generate spectra on-line.¹ Such real-time information is useful, but this approach has significant limitations in spectrum response and dynamic range of radiation intensity. In passive dosimetry, samples (e.g., activation foils or thermo-luminescent detectors) are placed in the neutron field, exposed for a well-defined time and then extracted for analysis.¹ This approach offers considerable spectrum sensitivity and the ability to handle a wide dynamic range of radiation intensities. A disadvantage is that dosimetry samples either have to be inserted before the irradiation begins and removed afterwards or they have to be transported mechanically (e.g., by a "rabbit" system) between the neutron field and a remote position for assay. The present concept offers desirable features from both the active and passive approaches, including real-time metrology, spectrum sensitivity and accommodation of a wide range of radiation intensities. A related technique has been exploited by Smith *et al.*² However, this earlier experiment was carried out to measure neutron activation cross sections, not to demonstrate a novel method of neutron metrology. Also, this investigation involved a collection of discrete cylindrical samples of solid material rather than a liquid.

This paper discusses the basic concept of the liquid dosimetry method, outlines the mathematical formalism used for data analysis and demonstrates the feasibility of this approach through measurements carried out with a simple experimental setup.

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2. Basic Concept

A homogeneous liquid, which consists of a solvent and dissolved chemicals in concentrations below the saturation level, is circulated in a closed system between a localized neutron field and detectors placed in a remote, low-background area. Water is a convenient solvent, with dissolved salts providing additional atomic species. The main components of this dosimetry system are: i) a known volume of the liquid, ii) uniform tubing to contain and transport this liquid, iii) a pump, iv) a flow meter, v) gamma-ray detectors, vi) a flow delay, vii) a probe and viii) a reservoir. This arrangement is shown schematically in Fig. 1.

Fig. 1. Schematic Diagram of a Circulating Liquid Dosimetry System

The liquid flows through a probe located in the neutron field. Position sensitivity is enhanced if the probe is small and the time spent there is maximized while that spent in neighboring locations is minimized. After leaving this probe, the liquid flows out of the neutron field, passes through an access port in the surrounding shielding and is observed by gamma-ray detectors. One detector is adequate, but greater flexibility is possible if there are two detectors with a flow delay in between them to aid in discriminating between various radioactivities with differing half lives that are present in the liquid after irradiation. Referring to Fig. 1, Detector 1 measures shorter-lived activities while Detector 2 measures longer-lived ones. After passing the detector station(s) the liquid enters a reservoir, where the radioactivities present can be expected to die away to negligible levels or to residual values low enough so that reliable corrections can be determined, before it is pumped back through the system. Such a dosimetry

system can be tailored for diverse applications by choice of: i) basic design and geometry, ii) liquid composition, iii) fluid flow rate and iv) detector type and size. Clearly, measurements with such a device can be continuous. When neutron fields and flow rates are steady, detector count rates for individual radioactivities approach equilibrium values with time constants that depend on the decay half lives. Fluctuations in the neutron field can be monitored by examining data for short-lived radioactivities. A detector will reproduce these fluctuations, but with a delay equal to the time taken by the fluid to flow from probe to detector. Data for longer-lived radioactivities reflect behavior of the neutron field averaged over longer time intervals.

3. Mathematical Formalism

The following assumptions lead to a simple but useful data-analysis formalism for this dosimetry approach: i) invariant neutron spectrum across the probe, ii) stable neutron field and liquid flow, iii) uniform fluid composition, iv) laminar flow (no turbulence) and v) use of a single detector. An equilibrium between production and decay of radioactivity will then exist during the measurements. Under these conditions, the absolute (normalized) detector count rate R_i associated with decay of the i^{th} radioactive species in the liquid is given by

$$R_i = (\lambda_i B_i \epsilon_i G_d) \exp(-\lambda_i t) [1 - \exp(-\lambda_i T)]^{-1} (M_i \langle \sigma_i \rangle G_p) \Phi_0, \quad (1)$$

where $i = 1$ to n ($n \geq 1$), λ is the decay constant ($\lambda = \ln 2/t_{1/2}$), B is the gamma-ray branching factor, ϵ is the efficiency for detecting the observed gamma ray, G_d is a parameter that depends on the volume of liquid observed, decay constant, detector geometry, flow rate and radiation absorption, t is the transit time of liquid from probe to detector, T is the time needed for liquid to make a complete circuit through the system, M is the number of target atoms involved in the activation reaction per unit volume of liquid, $\langle \sigma \rangle$ is the spectrum-averaged activation cross section for neutrons near the probe, G_p is a parameter that depends on the probe geometry, decay constant and flow rate, and Φ_0 is a constant that measures the absolute neutron-field intensity near the probe. The factor $(M \langle \sigma \rangle G_p) \Phi_0$ determines the formation rate for radioactive atoms in the liquid. The term $(\lambda B \epsilon G_d)$ accounts for properties of the gamma-ray detector and radioactivity decay. The factor $\exp(-\lambda t)$ adjusts for the decay of radioactivity in the liquid during transit from probe to detector. The factor $[1 - \exp(-\lambda T)]^{-1}$ compensates for residual radioactivity in the liquid due to preceding cycles. This factor is close to unity for short half lives ($\lambda T \gg 1$).

G_p and G_d are both rather insensitive to reaction and radioactivity decay properties. It is useful to consider the reaction-rate ratio ρ_{ij} (defined as R_i/R_j) since these parameters "G" tend to cancel along with the absolute neutron intensity Φ_0 . This leads to the formula

$$\rho_{ij} \approx \{F_i \exp(-\lambda_i t) [1 - \exp(-\lambda_i T)]^{-1}\} / \{F_j \exp(-\lambda_j t) [1 - \exp(-\lambda_j T)]^{-1}\} \quad (2)$$

for calculating detector count-rate ratios that can be compared to measured ones. It is obvious from Eq. (1) that F equals $(\lambda B \epsilon)(M \langle \sigma \rangle)$. If f is the flow rate (volume per unit time) and V_0 is the total volume of the liquid, then the cycle time T for liquid passing through the system is V_0/f . Similarly, if fluid flows from probe to detector through tubing with an internal diameter

d and length L , then the volume V_L of liquid in this segment is $\pi(d/2)^2L$ and the corresponding transit time t is V_L/f . For the stable measurement conditions described above, the full-energy-peak count ratio C_i/C_j for gamma-ray lines observed in recorded spectra is comparable to ρ_{ij} , as defined by Eq. (2). If the decay data, cross sections and various system parameters are accurately known, then differences observed between measured and calculated ratios ρ_{ij} that exceed estimated errors can be fully attributed to a discrepancy between the assumed (*a priori*) and actual neutron-energy spectrum shapes. This information is therefore quite valuable for spectrum-adjustment purposes.^{3,4}

4. Experimental Investigation

A simple experiment has been performed to demonstrate the present dosimetry concept. The apparatus, measurements and results obtained from this work are described in this section.

4.1 Apparatus and Pertinent Nuclear Data

The apparatus used resembled that shown in Fig. 1. The FNS accelerator at Japan Atomic Energy Research Institute (JAERI), Tokai, Japan, provided 14-MeV neutrons.⁵ The output from $^3\text{H}(d,n)^4\text{He}$, with 20 mA of 350-keV deuterons impinging on a rotating Ti-T target, was $\leq 3 \times 10^{12}$ n/s.⁵ The probe consisted of 275 cm stainless-steel tubing (0.41-cm ID) coiled into a hemisphere with ≈ 11 cm diameter. It was placed ≈ 5 cm from the point neutron source. The probe, water pump, reservoir and flow meter were connected by 0.96-cm ID plastic tubing. The flow delay between Detector 1 and Detector 2 was a coiled section of tubing. The liquid consisted of 264.9 grams of yttrium chloride hexahydrate ($\text{YCl}_3 \cdot 6\text{H}_2\text{O}$) in 16.094 liters of water. This corresponds to 16.45 g/l $\text{YCl}_3 \cdot 6\text{H}_2\text{O}$ in water, far less than the room-temperature saturation level of 2170 g/l.⁶ Some measurements were also performed using pure water. The flow rates were in the range 2.8-11 l/m. This corresponds to the following liquid transit times: in the probe (0.2-0.8 s), probe to Detector 1 (12.6-49.6 s) and complete cycle (87.8-344.9 s). The tubing was viewed by two shielded Ge gamma-ray detectors. They were located in a remote area of the laboratory. The distance from probe to Detector 1 was measured directly along the tubing. It equalled 32 m. A direct measurement of distance along the tubing from Detector 1 to Detector 2 would have been inconvenient, so it was estimated by using a Geiger-Mueller (G-M) counter to measure radiation dose at the entrance and exit (at a flow rate of 5.2 l/m) and attributing it entirely to ^{16}N decay. This led indirectly to a length of ≈ 74 m. This approach is prone to underestimating the length significantly due to neglect of ^{37}S and $^{89\text{m}}\text{Y}$ activities as well as background radiation. This possibility appears to have been supported by results from the analysis indicated in Section 4.4. A shielded NaI(Tl) scintillation detector (Detector 3), placed near Detector 1, measured response to fluctuations in neutron output.

The $^{16}\text{O}(n,p)^{16}\text{N}$, $^{89}\text{Y}(n,n')^{89\text{m}}\text{Y}$ and $^{37}\text{Cl}(n,p)^{37}\text{S}$ reactions were used as dosimeters. Nuclear data required for these reactions appear in Table 1.⁷⁻¹¹ The spectrum near the FNS target was dominated by neutrons with energies near 14-MeV, so $\langle\sigma\rangle$ could be approximated by $\sigma(14 \text{ MeV})$. The 14-MeV cross sections in Table 1 were obtained from the literature.⁷⁻⁹ $^{16}\text{O}(n,p)^{16}\text{N}$ differential cross-sections are reasonably well known but better accuracy is likely to be necessary for the other two reactions in practical neutron metrology applications.

Table 1. Nuclear Data for the Dosimetry Reactions*

"i"	Reaction	I (%)	E _t (MeV)	λ (s ⁻¹)	E _{γ} (MeV)	B (%)	σ (mb)
1	¹⁶ O(n,p) ¹⁶ N	99.762	10.245	0.09722	6.129	68.8	42.8
2	³⁷ Cl(n,p) ³⁷ S	24.23	4.194	0.002288	3.104	94.0	29.2
3	⁸⁹ Y(n,n') ^{89m} Y	100	0.919	0.04316	0.909	99.14	478

* I (isotopic abundance), E_t (threshold energy), E _{γ} (energy of measured gamma ray), σ (14-MeV cross section).

4.3 Response to Fluctuations in Neutron Intensity

The system response to fluctuations in neutron intensity was tested using pure water as the liquid. Neutron intensity was monitored with a fission detector located a few meters from the FNS target while the gamma-rays from ¹⁶N decay were measured with Detector 3. These detector signals were recorded with multi-channel scalars (dwell time of 10 s per channel). The results for flow rates f equal to 3 and 11 l/m are shown in Fig. 2. The dosimetry system clearly responds to the neutron output with a time delay consistent with the liquid flow rate. This occurs because the ¹⁶N half life is so short that any residual activity from preceding cycles of the liquid through the system is negligible.

Fig. 2. Response of the Fluid Dosimetry System to Fluctuations in Neutron Intensity

4.4 Gamma-Ray Yield Measurements

Gamma-ray spectra were recorded from Detectors 1 and 2 for flow rates of 2.8, 3.0, 4.0, 4.2, 5.8 and 5.9 l/m using the aqueous solution of $\text{YCl}_3 \cdot 6\text{H}_2\text{O}$ described in Section 4.1. Gamma-ray lines from ¹⁶N and ⁸⁹Y were visible in the Detector 1 spectra but the ³⁷S line was obscured

by the Compton distribution from ^{16}N gamma rays. A typical spectrum from Detector 2 is shown in Fig. 3. The prominent gamma-ray lines from ^{16}N , ^{37}S and ^{89}Y were all clearly visible in Detector 2 spectra. Thus, peak yields for the gamma-rays listed in Table 1 were determined for the spectra from this detector in spite of the flow delay uncertainty mentioned above.

Fig. 3. Germanium Detector Spectrum Recorded with an Aqueous Solution of Yttrium Chloride Hexahydrate

The gamma-ray yield measurements were performed after attaining saturation conditions for all the observed radioactivities. Adjusting for the delay time indicated above, saturation was found to occur within a few seconds after the start of an irradiation for both ^{16}N and $^{89\text{m}}\text{Y}$. Similar tests conducted for ^{37}S showed that ~ 15 minutes were needed for this activity to saturate, as indicated in Fig. 4. This can be attributed to the longer half life of ^{37}S . The factor $[1 - \exp(-\lambda_i T)]^{-1}$ in Eqs. (1) and (2) also reflects this effect. Values near unity were obtained for both ^{16}N and $^{89\text{m}}\text{Y}$. For ^{37}S it ranged from 1.83 to 3.20 for flow rates from 2.8 to 5.9 l/m, respectively. Ratios of calculated to experimental values (C/E) for ρ_{ij} were determined using available nuclear data and estimated system parameters. The agreement was very poor (e.g., C/E differed from unity by more than a factor of 10 in one case). A systematic error in the length of the flow-delay tubing was suspected as a possible cause for the discrepancies. Thus, ρ_{ij} was recalculated for various tubing lengths. Individual C/E values very close to unity could be obtained if tubing lengths in the range 94-110 m were assumed (rather than 74 m). The

average of these individual tubing lengths was 101 m. C/E values obtained using this "adjusted" common flow-delay tubing length for all the data appear in Table 2. They are reasonably consistent with unity, considering the relatively large uncertainties associated with the present work. While this approach suggests a plausible resolution for the observed discrepancies, it has not been rigorously confirmed. New measurements with better-determined system parameters are needed to settle the issue. However, the uncertainty associated with this issue does not in any way invalidate the intrinsic value of the liquid dosimetry concept.

Figure 4. Approach to Saturation for the ^{16}N , ^{37}S and ^{89}Y Radioactivities in the Neutron-irradiated Liquid

Table 2. Comparison of Calculated and Experimental Count-rate Ratios (C/E)*

Ratio	Run#1	Run#2	Run#3	Run#4	Run#5	Run#6	Run#7
ρ_{21}	-	-	-	1.02	1.16	1.14	0.95
ρ_{31}	-	-	-	1.50	1.31	1.30	1.21
ρ_{32}	0.70	1.28	1.11	1.47	1.13	1.14	1.28

*Run#1 (2.8 l/m), Run#2 (3.0 l/m), Run#3 (4.0 l/m), Run#4 (4.2 l/m), Run#5 & #6 (5.8 l/m), Run#7 (5.9 l/m)

5. Summary

This study demonstrates that a circulating liquid with dissolved chemicals can be used for on-line dosimetry. Reactions with relatively short-lived reaction products are used to minimize radioactivity buildup effects and enhance responsiveness to neutron intensity and spectrum variations. Measured yields of ^{16}N , ^{37}S and $^{89\text{m}}\text{Y}$ gamma-rays under saturation conditions can be used to provide information on the neutron intensity and spectrum shape, provided that accurate nuclear data are utilized and the parameters of the system are well determined.

Many applications are envisioned. For example, fluid dosimeters could be placed at various locations in a fusion reactor to provide real-time data on neutron intensities and spectra in the machine. Such a device could also be used for neutron monitoring purposes at high-fluence irradiation facilities, such as those planned for materials testing applications.

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