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SUMMARY OF THE SAVANNAH RIVER SITE CRITICALITY DOSIMETRY PROGRAM*

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ABSTRACT

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The mission of the Savannah River Site (SRS) includes working with fissionable materials. A program is in place, therefore, to assess neutron and gamma doses to individuals in the event of a criticality accident at SRS. The program consists of a method to quickly screen for potentially exposed personnel, a method to provide early but preliminary dose estimates, and a nuclear accident dosimeter and assay procedure to enable final dose estimates.

INTRODUCTION

The Savannah River Site (SRS) is a 300 square mile reservation operated for the United States Department of Energy by the Westinghouse Savannah River Company (WSRC). It is located in the State of South Carolina, in the southeastern United States, and adjacent to the Savannah River. It is about 25 miles from the city of Augusta, and about 125 miles from the city of Savannah.

SRS includes the facilities necessary to produce nuclear materials for defense purposes, such as plutonium and tritium. Major facilities include a fuel fabrication facility, heavy water reactors, chemical separations facilities, waste management facilities, and adjunct support and applied research facilities. A major new facility is being completed to vitrify highly radioactive liquid wastes generated over the past 40 years.

The mission of SRS includes handling and working with fissionable materials. While the probability of a criticality accident at SRS is kept small by engineered and administrative controls, a criticality dosimetry program has long been in place to enable the assessment of neutron and gamma doses to personnel in the event of a criticality accident at SRS. Over the years, the program has been improved and thoroughly tested.

The SRS Criticality Dosimetry Program consists of three components:

- (1) A field method for screening all potentially exposed personnel.
- (2) A method for quick approximations of dose.
- (3) A nuclear accident dosimeter capable of providing neutron doses in various energy ranges with acceptable accuracy.

This report summarizes the SRS Criticality Dosimetry Program.

DISCUSSION

Measurement of radiation doses resulting from a criticality accident cannot always be readily accomplished by evaluation of neutron and gamma dosimeters used for routine occupational dose measurement. Radiation levels from a criticality accident may well exceed the measurement capability of such dosimeters. Also, most neutron dosimeters used for routine occupational exposures do not provide response in different energy levels, their responses are energy dependent, and the calibration of such neutron dosimeters usually incorporates neutron Quality Factors for occupational exposures, which are not applicable for the prompt neutrons resulting from a criticality accident. Therefore, nuclear accident dosimeters are used in facilities containing fissionable materials.

The nuclear accident dosimeter in use at SRS is the Criticality Neutron Dosimeter (CND). The CND was originally developed at SRS and described by J. E. Hoy in 1960.¹ The CND and the SRS Criticality Dosimetry program have been substantially refined over the years, and are described in detail in a 1990 technical report by C. N. Wright.² A procedure

is maintained for use in evaluation of doses received in the event of a criticality accident at SRS.³

Screening

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At SRS, all personnel who enter a facility which requires installed alarming criticality monitors (at SRS they are called Nuclear Incident Monitors, or NIMs) are required to wear a CND on their front torso, in addition to routine dosimetry. Fixed CNDs are also mounted on walls in such facilities, especially in personnel egress pathways. In the event of a NIM alarm, personnel are trained to quickly evacuate the facility to a designated rally point. To quickly identify personnel who may have received doses of concern, field screening methods are used at the rally point. These methods are proceduralized for field health physics staff use.

One screening method used is to survey the SRS security badge worn by all personnel on SRS. This badge contains a small strip of indium foil. Neutron activation of this indium foil is measured with a GM survey instrument to identify exposed personnel.

Another screening method used is to have individuals bend over a GM survey instrument held against the abdomen. Activation of sodium in the body may be detected, but results can be adversely affected by variation in body size, geometry, and by interference from short-lived ³⁸Cl.

Persons determined to have been exposed by these screening methods are checked for contamination, decontaminated (unless badly injured), and placed under medical care. Information about these individuals, their positions and locations during the criticality are collected, along with all available dosimetry (routine gamma and neutron dosimeters, CNDs, security badges), and are transported to the health physics laboratory for analysis.

Preliminary Dose Estimates

The SRS Medical Department will obtain a 50-mL sample of blood from each person determined to have been exposed. This must be done before any additional sodium is administered, either through food or medical treatment. Ammonium oxalate will be added to the sample to prevent coagulation. Following this, the sample will be centrifuged and 10-mL samples of the plasma will be forwarded to a designated counting facility.

The plasma samples are counted on a high purity germanium detector to determine the ²⁴Na content in μ Ci/mL. This figure is then decay-corrected back to the time of the incident (T₀). The sample activity must also be corrected for sodium concentration if it is different from 3.2 mg/mL because the conversion factors in the dose calculation equation are keyed to the average concentration of sodium in human plasma⁴.

The first estimate of personnel dose from blood sodium activity requires a preliminary classification of the type spectrum involved from knowledge of the incident. The following guidelines are used:

• <u>Fast spectrum</u>. Includes all bare metal assemblies regardless of size or shape. Principal criterion is the absence of moderating materials such as water and organic solvents or thick concrete shields.

Neutron dose (rad) = (μ Ci/mL ²⁴Na at T₀) x 1.2E+05

• <u>Semi-moderated spectrum</u>. Includes process tanks or small vessels filled with either light water or organic solvents.

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Neutron dose (rad) = (μ Ci/mL ²⁴Na at T₀) x 1.0E+05

<u>Moderated spectrum</u>. Includes reactors, basins, or large tanks containing considerable volumes of moderator.

Neutron dose (rad) = (μ Ci/mL ²⁴Na at T₀) x 0.8E+05

Corrected sodium activity, multiplied by the appropriate conversion factor for the incident type, yields a very accurate estimate of the dose received by the person. After the CND elements are processed, the dose conversion factor (rad/ μ Ci/mL) value may be further refined by use of another factor (T/F) derived from the sulfur and indium activities in the dosimeter elements. This factor adjusts the sodium activity to dose relationship for the fastness of the exposing spectrum.

The ability of this sodium activation technique to determine personnel dose has been tested in series of exposures to the Health Physics Research Reactor (HPRR) at the Oak Ridge National Laboratory.⁵ The HPRR is a bare critical assembly that can be pulsed to yield a very short, high intensity burst of neutrons and accompanying gamma radiation. In these tests, phantoms are fitted with all types of dosimeters and are filled with a salt solution having the same sodium concentration as human blood. The dosimeters are positioned on the front, side, and back of the phantoms, as well as in air. The phantoms may be exposed to the bare reactor or shielded from it by a variety of shielding materials. The doses received in these exposures are well documented by the operators of the reactor and have been confirmed by practically every major organization in the world that has an interest in this type of dosimetry⁶. Table 1 shows the results of this testing.

Gamma dose measurements may be obtained from three sources:

- (1) A 0.4-mm x 12.7-mm disk of LiF-Teflon laminated in every security badge issued at SRS
- (2) The two LiF chips in the CND

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(3) The routine personnel monitoring TLD dosimeter.

Preliminary estimates of total dose may be made from either measured neutron or gamma dose using the following neutron-to-gamma ratios that are based on the estimate of incident geometry as discussed previously. The ratios used are:

Spectrum	Ga	mma/Neutron Ratio
Fast fission spectrum		0.278
Semi-moderated neutron spectrum		1.00
Moderated neutron spectrum	, · ·	2.00

Final Dose Estimates

The CND is shown in Figure 1. It contains cadmium-shield indium foil and unshielded indium foil, copper foil, sodium fluoride powder, sulfur powder, and TLD chips. Its contents are listed in Table II. The CND is commercially available.

Theory

The CND is based on activation analysis techniques. The basic idea is to incorporate, in the dosimeter, elements that have an activation cross-section in the desired energy range. If these elements are exposed long enough to reach equilibrium, the decay rate of the daughter will be equal to the rate at which neutrons of that energy were captured by the original element. If equilibrium exposure is not reached, suitable corrections are made.

Another important feature of the personnel CND is that it is automatically retrieved from the accident area as its wearer exits upon an alarm. Many criticality dosimetry systems rely on fixed dosimeters, wall-mounted, or otherwise placed in areas where a possibility of a criticality exists. While much useful data about potential damage to equipment of facilities where a criticality has occurred may by derived from fixed dosimeters, it is very difficult to use them to determine exposure to personnel. To do so, data from the fixed dosimeters must first be used to derive isodose data. Any shielding materials which are present will make this a very difficult task. Additionally, the exit path and time in various dose rate areas must be combined with the dose rates in these areas to determine the dose to a person. This is a time-consuming task which incorporates high potential for uncertainty. The personnel CND automatically integrates the dose received by its wearer and furnishes information on the direction from which the majority of the dose was received. For these reasons, primary reliance for dose estimates at SRS is placed on the personnel CND and/or blood sodium activation. Fixed CNDs, mounted on walls in facilities where the potential for a criticality exists, are intended for backup evaluation or accident analysis should a criticality occur in an unoccupied facility.

The activation materials used in the CND and their physical characteristics are discussed in the following paragraphs.

Energy Intervals of 0.0 eV to 0.5 eV and 0.5 eV to 2 eV

Bare and cadmium-shielded indium foils are used to determine neutron fluence in these energy intervals. Indium has two stable isotopes: ¹¹³In (4.28% abundance) and ¹¹⁵In (95.72% abundance). ¹¹⁵In (n,gamma) ¹¹⁶In is the reaction of interest. ¹¹⁶In has three isomers, two of which have very short half-lives (2 seconds and 14 seconds); the third isomer, with a 54-minute half-life, is the principal source of radioactivity for a few hours after neutron irradiation. ¹¹⁴In, resulting from the ¹¹³In (n,gamma) ¹¹⁴In reaction, has an isomer with a 50-day half-life, which is useful when a considerable delay occurs before counting.

The cross-section of indium approximates a 1/v relationship from 0.025 eV to 0.3 eV; that is, the cross-section decreases as neutron velocity increases. Above 0.3 eV, resonances occur, with the principal peak at 1.4 eV.

Cadmium of the thickness used in the dosimeter (0.003 inch) absorbs essentially all neutrons with energies less than 0.5 eV, the cadmium cutoff. Therefore, the difference between the activities of the bare and the cadmium-shielded indium foils is caused by neutrons in the energy interval from 0.0 eV to 0.5 eV. A factor of 1.15 is use to correct for the resonant absorption of neutrons by the cadmium shield⁸. The effective activation cross-section for this interval is 145 barns. Activation of cadmium-shielded indium is used to

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measure the fluence in the energy interval from 0.5 eV to 2 eV. A resonance integral (corrected for flux depression) of 650 barns is assigned to this interval.

Energy Interval of 2 eV to 1 MeV

The reaction of interest for this energy interval is 63 Cu (n, gamma) 64 Cu. 64 Cu has a 12.8hour half-life. A competing activity resulting from 66 Cu (5-minute half-life) can be minimized by counting the copper no sooner than 1 hour after irradiation. The average activation cross-section for 63 Cu in this energy range is 0.300 barn. Above 1 MeV the inelastic neutron scattering cross-section becomes dominant. The copper is shielded by indium and cadmium to minimize the effect of neutrons below 2 eV.

Energy Interval of 1 MeV to 2.9 MeV

The inelastic scattering reaction of fast neutrons with ¹¹⁵In is used to determine fluence above 1 MeV. While the threshold for this reaction is about 450 keV, its cross-section becomes significant at 1 MeV. The average cross-section above 1 MeV is 0.180 barn. The ¹¹⁵In isomer has a 4.5-hour half-life. Gamma spectrometric analysis is used to distinguish the activation due to the inelastic reaction from the activation by the epithermal neutrons. The cadmium-shielded indium foil is used for this measurement.

The cross-section for this reaction remains significant to values above 5 MeV. Therefore, the fluence of neutrons above 2.9 MeV, as determined from the activation of sulfur, must be subtracted from the fluence calculated for indium activation. The difference represents the fluence in the range from 1 MeV to 2.9 MeV.

Energy Interval Above 2.9 MeV

The threshold reaction, ${}^{32}S(n,p) {}^{32}P$, is used to measure fluence above 2.9 MeV. Sulfur has a relatively high and constant cross-section, and ${}^{32}P$ has a conveniently long half-life.

Processing

For processing, the dosimeter elements are removed from the cylindrical housing, and the foils and powders are placed on 2-inch diameter planchets for counting. The foil elements are laid flat on the planchet surface, and the powder elements are spread evenly over the surface of the planchet in a level layer. All elements are counted in a 2.25-inch diameter gas flow proportional counter that is calibrated with a ¹³⁷Cs standard source. The cadmium-shielded indium foil is also counted on a high purity germanium detector.

In practice, each element is counted in two separate counters and the decay-corrected results compared, to detect any potential counting error. This is necessary, not only for accuracy, but because the short half-lives involved dictate that any error be discovered while enough activity remains to allow the element to be measured. Results are recorded as disintegrations per minute (d/m) referenced to the standard calibration.

Neutron fluence (neutrons/cm²) in five energy intervals is then determined. The effective d/m is calculated by correcting the observed d/m for decay to the time of the incident, for non-equilibrium exposure, and for the abundance of the daughter product being counted. The neutron fluence is then derived based on the neutron cross-section for the production of the daughter being counted, and the experimentally derived relation between effective d/m and neutron fluence.

Orientation of Dosimeter

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Since the dosimeter is normally worn on the front of the person and its activation is affected by the body's shielding and moderation, it is critically important to know the direction from which the person was exposed. If a correction for direction of exposure is not made, the dose estimate may be low by a factor of as much as two in the case where the exposure was received from the back of the wearer. The SRS dosimeter incorporates a unique technique for determining the direction from which the majority of the exposure was received. By comparing the activity of the ²⁴Na in the subject's blood with the activity of the ²⁴Na produced in the sodium fluoride element in the dosimeter, the direction of exposure can be correctly predicted in almost every case of exposure from the back. Occasionally, exposures from the side or front may be incorrectly predicted, but the error in these cases is small. The method is as follows:

 $Ratio = \frac{activity per gram of sodium fluoride}{activity per mL of blood}$

This ratio is compared to the predicted ratio that is a function of the energy of the exposing spectrum. The predicted ratio is determined as follows:

Predicted ratio = Y = 0.068 x [Thermal Fluence/Fast Fluence]^{0.622}

where thermal and fast fluence are the values determined by the dosimeter (without directional correction)

If the ratio is less than 0.53Y, the majority of the exposure was received from the back of the wearer and the following correction factors must be applied when calculating the neutron dose:

СТ	=	Thermal correction	=	0.64
CET	=	Epithermal correction	=	1.6
CCU	=	Resonance correction	=	2.0
CM	=	Medium energy correction	=	5.4
CS	=	Fast correction	=	6.7

If the ratio is greater than 1.3Y, the majority of the exposure was received from the front of the wearer and the following correction factors must be applied when calculating the neutron dose:

CT	=	Thermal correction	=	0.23
CET	=	Epithermal correction	=	0.35
CCU	=	Resonance correction	=	0.45
СМ	=	Medium energy correction	=	0.79
CS	=	Fast correction	=	0.72

Determination of Neutron Dose

The neutron dose equivalent is the product of the absorbed dose (rad), quality factor, dose distribution factor, and any other modifying factors. The quality factor to be used for neutrons in a criticality accident has not been defined. Since the rad dose value is a physical quality that can be measured, all results from the CND are given in rad.

After many years of intercomparison testing at HPRR as discussed above, the neutron dose values obtained from the CND appear to closely match the "element 57" dose reported by DOSAR as the referee's value⁶. See Table III.

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The total neutron dose in rads is determined by multiplying the fluences in the five energy ranges, as determined from counting the CND materials, by the respective dose conversion factors for that energy interval (from Table III), then summing the five doses. If the exposure was determined to be from the front or rear, the five doses are corrected by the factors from the previous page. Side exposures require no correction.

Manual calculation of the dose is lengthy and involved. Computer programs are available that accept counting data, decay, exposure times and weights of activated materials and calculates the orientation of the dosimeter relative to the wearer, fluence and dose in the five energy intervals, and the total dose.

SUMMARY

The SRS Criticality Dosimetry Program consists of quick field screening methods to identify potentially exposed personnel, a quick method for preliminary dose estimates based on blood sodium analysis, and a Criticality Neutron Dosimeter with procedures for analysis which allow final dose estimates with acceptable accuracy.

Testing results have shown that both the preliminary dose estimates and the dose estimates based on the CND average bareeing within about 20 percent of reference dosimetry. Continued testing of the CND is important for the continued readiness to provide neutron and gamma doses in the event of a criticality accident at SRS. Table 1

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Analysis of CND Test Results (Phantom Measurements)

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	Blood Sodium Dosimetry		CND
	Using "Standard" Factor	Using "T/F" Factor	Result
	26 Tests	<u>1970-1985</u>	
Average Error Maximum Error Errors >30% Errors >25%	±14.9 +35.0 1 6		±17.1 -39.7 4 7
	<u>10 "Bare"</u>	Exposures	
Average Error Maximum Error Errors >30% Errors >25%		± 14.1 +32.6 1 2	±18.7 -37.0 2 3
	5 "Steel"	Exposures	
Average Error Maximum Error Errors >30% Errors >25%	±10.1 +19.8 0 0		±20.4 -39.7 2 2
	5 "Lucite	e Exposures	
Average Error Maximum Error Errors >30% Errors >25%	±11.4 +25.6 0 1		±11.1 -26.3 0 1
	6 "Concre	te" Exposures	
Average Error Maximum Error Errors >30% Errors >25%	±16.4 +35.0 1 1	±16.5 +27.0 0 1	±16.8 +31.0 1 1

Note:

"Bare" refers to the unshielded HPRR assembly
"Steel" refers to 13-cm steel shield between reactor and dosimetry stations
"Lucite" refers to 12-cm Lucite shield between reactor and dosimetry stations.
"Concrete" refers to 20-cm concrete shield between reactor and dosimetry stations.

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Table IIDosimeterComponents

Material		Size or Weight	
Cadmium	2 pieces	3/8" diameter x 1/32"	
	1 piece	1" x 5/8" x 1/32"	
Indium	1 piece	15/16" x 5/8" x 0.005"	
	1 piece	1.5" x 5/8" x 0.005"	
copper		15/16" x 5/8" x 0.005"	
Sodium fluoride		1.5 gram	
Sulfur		1.0 gram	
TLD 700 chips		2 each	
Plastic Tube		3.5-4" long	
(with tamper-resistant end caps)		$0.5" \pm 0.05"$ inside diameter	
	-	0.02" - 0.03" wall thickness	
Vials (with caps)	4 piece	5/8" - 1" long	
· • •	-	3/8" minimum inside diameter	
		1/2" maximum outside diameter	

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Table III Average Dose Conversion Factors

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Energy Interval	<u>rad/(n)(cm²)</u>
thermal to 0.5 eV	0.32E-10
0.5 eV to 2.0 eV	0.22E-09
2.0 eV to 1 MeV	1.25E-09
1 MeV to 3.0 MeV	2.87E-09
3.0 MeV and above	4.80E-09

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