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SOLID STATE TRACK RECORDER NEUTRON DOSIMETRY MEASUREMENTS FOR FUEL DEBRIS ASSESSMENT OF TMI-2 DEMINERALIZER-A

FH Ruddy JH Roberts R Gold CC Preston JA Ulseth

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Frank H. Ruddy, James H. Roberts, Raymond Gold, Christopher C. Preston, and James A. Ulseth

December, 1982

Hanford Engineering Development Laboratory Richland, Washington

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SOLID STATE TRACK RECORDER NEUTRON DOSIMETRY MEASUREMENTS FOR FUEL DEBRIS ASSESSMENT OF TMI-2 DEMINERALIZER-A

Frank H. Ruddy, James H. Roberts, Raymond Gold, Christopher C. Preston, and James H. Ulseth

ABSTRACT

Solid State Track Recorder (SSTR) neutron dosimetry measurements have been made in TMI-2 makeup Demineralizer A Cubicle in order to assess the amount of fuel debris present by means of the specific neutron activity of TMI-2 fuel. Based on recent calibration data and the results of the TMI-2 SSTR neutron dosimetry, the amount of fuel present is estimated to be 1.7 ± 0.6 kg. This value is in excellent agreement with a value determined independently by Compton recoil gamma-ray spectrometry. Sources of uncertainty in and proposed refinements of the present SSTR measurements are discussed.

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SOLID STATE TRACK RECORDER NEUTRON DOSIMETRY MEASUREMENTS FOR FUEL DEBRIS ASSESSMENT OF TMI-2 DEMINERALIZER-A

Frank H. Ruddy, James H. Roberts, Raymond Gold, Christopher C. Preston, and James H. Ulseth

1.0 Introduction

As a result of the Three Mile Island Unit 2 (TMI-2) accident on March 28, 1979, fuel debris was dispersed into the primary coolant system of the reactor. Two makeup and purification demineralizers, A and B, which maintain coolant water purity, were in operation at the time of the accident. Due to the high gamma ray intensities in the location of these demineralizers, fuel was presumed to be located in the demineralizers. As part of the TMI-2 recovery task and WHC TMI-2 Demineralizer Resin Removal Program, the amount of fuel debris in these demineralizers required quantification before other phases of the ion exchange resin removal program could proceed.

The presence of fuel may be traced by using the radiation emitted by the fuel or fuel products. TMI-2 fuel emits two easily detectable forms of radiation which are (1) gamma rays from fission products, and (2) neutrons from actinide buildup in the fuel.

A companion paper⁽¹⁾ describes the use of Compton recoil electron gamma ray spectrometry to detect ¹⁴⁴Ce (which is correlated with the fuel),¹³⁷Cs, and ¹³⁴Cs absolute gamma ray fluxes. This report describes neutron detection using solid state track recorders (SSTRs). TMI-2 fuel has an estimated average neutron specific activity of about 300 n/sec/kg,^(2,3) which results mainly from Pu buildup. Assuming that Pu is a good chemical tracer for uranium, SSTR neutron dosimetry can be used to assess the location and quantity of fuel present.

The use of SSTRs for fuel detection applications has been described previously.⁽³⁾ Briefly, enriched ²³⁵U foils are placed in firm contact with mica SSTRs, and the neutron induced fission fragments from ²³⁵U register as regions of damage (tracks) in the mica. These tracks are chemically developed to a size that is visible with a microscope. The number of tracks per unit area in the mica is proportional to the number of fissions per unit area in the adjacent uranium foil. Using appropriate calibration data, this fission rate can be used to deduce the neutron fluence. The neutron fluence and duration of exposure of the SSTRs can then be used to ascertain the amount of fuel present.

This report describes the SSTR neutron dosimetry measurements that were made in the TMI-2 Demineralizer A Cubicle and the relevant calibration measurements that were made at Hanford Engineering Development Laboratory (HEDL). On the basis of these measurements and calibrations, the amount of fuel present in TMI-2 Demineralizer A is estimated.

2.0 TMI-2 Measurements

SSTR neutron dosimeters were constructed as shown in Figures 1 and 2. Two 3" x 1" sheets of 0.004" thick 93% enriched 235 U were sandwiched between two pieces of mica and pressed in firm contact against an aluminum support plate between two 0.25" thick pieces of lucite. The lucite was used to enhance the neutron signal via the albedo effect which has been reported previously.⁽³⁾ The total SSTR area of this neutron dosimeter is approximately 85 cm². These dosimeters were assembled at TMI-2 immediately prior to the exposure (Figures 3 and 4) to



FIGURE 1. SSTR Neutron Dosimetry Holder for TMI-2 Demineralizer A Measurements.

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FIGURE 2. Photograph of Disassembled SSTR Neutron Dosimetry Holder. Parts are Assembled as Shown in Figure 1.



FIGURE 3. SSTR Neutron Dosimeters During Assembly. Mica and ²³⁵U foils are wrapped in polyethylene bags in foreground.



FIGURE 4. SSTR Neutron Dosimeters During Assembly. Fully assembled dosimeters are in background.

reduce background due to cosmic ray neutron induced fission and from spontaneous fission of the ²³⁸U in the uranium. Also circular (1" diameter) CR-39 SSTRs were attached to the outer surface of each dosimeter (see Figure 4) to measure the high energy (E > 0.5 MeV) neutron flux.

An overview of TMI-2 Demineralizer A and B Cubicles is shown in Figure 5. Due to the intense gamma ray fields present near the demineralizers, neutron dosimeters had to be placed remotely from outside the cubicle. The demineralizer A cubicle was accessible through penetration #891 shown in Figure 5.

A vertical stringer was prepared by fastening together SSTR dosimeters at measured intervals using fishing line. This stringer was enclosed in plastic tubing to protect the dosimeters from contamination inside the demineralizer cubicle. A horizontal set of dosimeters was prepared by attaching the dosimeters to a pipe which was then enclosed in plastic tubing. Both the horizontal and vertical stringers were inserted into the cubicle through penetration #891 as shown in Figures 6, 7 and 8. The dosimeters were inserted at 11:55 p.m. on September 14, 1982, and occupied the positions shown in Figure 9. The location of the vertical stringer was confirmed during a robot entry of the cubicle. The dosimeters were left in place for twenty-nine days and removed on October 13, 1982, at 6:20 p.m. Some difficulties were encountered during removal, resulting in a maximum uncertainty of 3% in the vertical stringer exposure time. In order to measure the detector background, control dosimeters were assembled at the same time and exposed in a demineralizer cubicle (D) where fuel was not present. Assembly and disassembly of all of the dosimeters required a few hours so that background cosmic ray neutron exposure is approximately the same for all detectors. A summary of the detector positions and labels is given in Tables 1 and 2. After exposure, the SSTRs were

TMI-2 MAKE-UP & PURIFICATION DEMINERALIZERS



FIGURE 5. Isometric View of Makeup Demineralizer Cells A and B.

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FIGURE 6. Emplacement of SSTR Neutron Dosimeters--Attaching Vertical Stringer to Support Pole.



FIGURE 7. Emplacement of SSTR Neutron Dosimeters--Measurement of Reference



FIGURE 8. Emplacement of SSTR Neutron Dosimeters--Final Insertion Through Penetration #891.



FIGURE 9. Locations of SSTR Neutron Dosimeters During Exposure.

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	TABL	.Ε	1.	SUMMARY	OF	DETECTOR	LABELS	AND	LOCATIONS	FOR	VERTICAL	STRINGER
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		Demineralizer	•
Holder Number	SSTR Labels	Location	Elevation
<u>e</u> 1	TMI-253-253'*	A	317'0"
· - .	-254,254'		
2	TMI-255,255'	A	313'6"
	-256,256'		
3	. TMI-257,257'	· A	313'0"
	-258,258'		
4	TMI-259,259'	Α	312'6"
	-260,260'		
5	TMI-261,261'	Α	312'0"
	-262,262'		
6	TMI-263,263'	Α	311'0"
	-264,264'		
. 7	TMI-265,265'	Α	310'0"
	-266,266'		
8	TMI-267,267'	Α	309'0"
	-268,268'		
9	TMI-269,269'	- A	308'0"
	-270,270'		
10	TMI-271,271'	А	307'6"
	-272,272'	· · · · ·	
11	TMI-273,273'	Α	307 '0"
	-274,274		
12	TMI-275,275'	Α	306'0"
•	-276,276'		
13	TMI-277,277'	A ·	305'9"
·· ··	-278,278'		
21	TMI-293,293'	D	307 '6"
	-294,294'		
23	TMI-297,297'	D	311'6"

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* Mica SSTRs were numbered in sequence. Unprimed numbers correspond to SSTRs nearest to lucite. Primed numbers correspond to SSTRs nearest to aluminum.

TABLE 2. SUMMARY OF DETECTOR LABELS AND LOCATIONS FOR HORIZONTAL STRINGERS

Holder Number	SSTR Labels	Demineralizer A Location
14	TMI-279,279'	-2',0"
	-280,280'	
15	TMI-281,281'	-1',4"
	-282,282'	
16	TMI-283,283'	-0',8"
	-284,284'	
17	TMI-285,285'	0
	-286,286'	
18	TMI-287,287'	+0',8"
	-288,288'	
19	TMI-289,289'	+1',4"
	-2,90,290'	
20	TMI-291,291'	+2',0"
	-292,292'	

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۰ . transported to HEDL where they were processed by etching with 49% HF at room temperature for 90 minutes. The developed tracks from selected dosimeters were manually counted with the aid of a microscope. The measured ²³⁵U track densities for this exposure are given in Table 3.

3.0 Analysis and Results

The SSTR ²³⁵U track densities from Table 3 are plotted in Figure 10. Shown for comparison is a curve resulting from gamma scanning⁽¹⁾ of the ^{1,4+}Ce activity present in the demineralizer tank. Within experimental limitations, the positions of the two peaks are the same, indicating the presence of fuel at the 309' elevation. The assumptions are made here that ¹⁺⁴Ce and Pu both are closely associated with the fuel, resulting in fuel traceable gamma rays and neutrons, respectively.

Although the background measurements give a track density of about 5 tracks/cm² due to cosmic radiation, the baseline for the measurements in the demineralizer cubicle is about 10 tracks cm/². The 5 tracks/cm² difference is due to room return neutrons resulting from thermalization of source neutrons in the walls of the cubicle. This effect, which will be discussed in detail in Section 4.2, has been observed before in this type of measurement. Figures 11 and 12, which are taken from Reference 3, show radial and axial fission rate distributions in the vicinity of a spent fuel subassembly housed in a hot cell. The isotopes with threshold neutron response (238 U, 237 Np, 232 Th) respond to fast neutrons from the source and show the expected spatial distributions.* However, the 235 U

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^{*}CR-39 SSTRs were included with the TMI-2 SSTR neutron dosimeters to provide a measure of the high energy neutron intensity in Demineralizer A Cubicle. The CR-39 SSTR has a higher sensitivity than the ²³⁸U, ²³⁷Np, and ²³²Th. The CR-39 SSTRs have not as yet been analyzed.

TABLE 3.	MEASURED SS	STR TRACK DENSITIE RAVERSE (DATA TAKE	S AT SELEC	TED LOCATIONS ALONG THE DEMINERALIZER A CUBICLE)
				<u></u>
TMI#	Elevation (Ft.)	Meas Track Obs. 1	ured s/cm ² Obs. 2	Measured Tracks/cm² Avg.* - B. G.
253	317	11.1		6.1 ± 2.0
261	312	26.1		21.1 ± 2.8
263	311	25.2	27.0	20.2 ± 2.7
265	310	30.1	30.6	25.3 ± 3.2
267	309	36.8	36.1	31.5 ± 4.2
269	308	13.3	12.4	7.9 ± 1.7
273	307	10.2		5.2 ± 1.9
Deminera	lizer D	-		.

Measured Background (B. G.)

4.8 5.3

(B. G. Average = $5.0 \pm 1.1^*$)

* Average of Observers 1 and 2.





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FIGURE 11. Fission Rates as a Function of Radial Distance from a Reactor (PWR) Spent Fuel Assembly.



FIGURE 12. Axial Fission Rate Distributions Measured Along the Face of a Reactor (PWR) Spent Fuel Assembly.

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fission rates tend to be constant at all positions, since the ²³⁵U responds mainly to room return thermal neutrons. The room return neutron intensity, which is proportional to the total source neutron intensity, can be used to determine the amount of fuel present as will be shown in Section 4.2.

The SSTR intensity distribution in Figure 10 is asymmetric, decreasing less rapidly above the 309' elevation than below. A probable explanation is that the tank is dry above the 309' elevation and contains degraded resin and possibly boronated water below this level. The resin attenuates fuel neutrons below 309', whereas the slow falloff in intensity above 309' is due to the increase in distance between the SSTR neutron dosimeters and the neutron source.

The track densities observed in the SSTR neutron dosimeters correspond to extremely low neutron fluxes. Using calibrations described in Section 4.0, the total neutron emission rate in the demineralizer A Cubicle is found to be about 500 neutrons per second, corresponding to a flux of less than 3×10^{-3} n/cm²/sec at the position of the SSTR neutron dosimeter with the maximum signal. The present SSTR neutron dosimetry is the only known method capable of detecting such a low neutron flux.

During unloading of the dosimeters, the lucite albedo blocks were observed to have been discolored by radiation damage. The intensity of the discoloration appears to be proportional to the intensity of the gamma ray dose at the location of each dosimeter, as can be seen in Figure 13. Quantitative light absorption or transmission measurements should be made on these lucite blocks to determine if usable gamma ray dose information can be derived.



FIGURE 13. Lucite Blocks from SSTR Neutron Dosimeters that were Exposed in Demineralizer Cubicle A. Discoloration is the Result of Accumulated Radiation Damage from Gamma Ray Exposure. The Maximum Discoloration Corresponds to the Location of Highest Gamma Ray Dose.

4.0 <u>Calibration Studies</u>

4.1 SSTR Neutron Dosimeter Response

In order to convert observed fission track density into neutron fluence, calibration data on the response of the SSTR neutron dosimeter is required. This response will depend on the energy spectrum as well as the intensity of the incident neutrons. Neutrons from the source (fuel in the demineralizer) will be moderated, absorbed, and reflected by the surroundings, and the resultant neutron spectrum will be sensitive to the presence of moderators such as water or materials with high neutron absorption cross sections. Calibration data had been obtained previously⁽³⁾ using a ²⁵²Cf source in an experimental configuration designed to "mock-up" the demineralizer and its surroundings. Unfortunately, at the time these calibrations were carried out it was believed that the demineralizer was filled with 1100 ppm borated water.

On the basis of the present work and the related gamma ray spectrometry measurements,⁽¹⁾ the demineralizer can be assumed to be dry above the 309⁺ elevation. Below this elevation, degraded resin is present containing unknown amounts of water and boron. Nevertheless, the previous dosimeter calibrations⁽³⁾ provide insight into the response of the ²³⁵U fission rate as a function of moderation and absorption of source neutrons. Figure 14 shows the response of SSTR dosimeters to a ²⁵²Cf source immersed in borated water at various depths. Note that the SSTR response peaks directly above the source but drops off rapidly for other "positions as a result of the attenuation (absorption) of source neutrons by the borated water. This effect is also shown in Figure 15 where the peak intensity is plotted as a function of water depth. This response is the basis of the argument presented in Section 3.0 that the tank is dry above the 309' level. If water were present, the response curve plotted in Figure 10 would decrease much more rapidly above the 309' level.

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FIGURE 14. SSTR Neutron Dosimeter Response Along the Surface of a Tank of 1100 ppm Borated Water Containing a ²⁵²Cf Source Located at Various Depths.



The dosimeter design shown in Figure 1 differs from the design used for previous calibrations⁽³⁾ in that two layers of 0.004" uranium are used to increase the detector surface area and lucite albedo blocks are used to increase the neutron response of the ²³⁵U by contributing further moderation of the neutron spectrum. In order to calculate the relative response of the two SSTR neutron dosimeters, additional ²⁵²Cf exposures were carried out. For reasons detailed in Section 4.2, the ²⁵²Cf calibrations were carried out in a concrete walled cubicle. Both types of dosimeters were exposed to the source which was suspended at heights of 2' and 3.5' in the center of the room. The room has overall dimensions of 11.5' x 8' and a height of 8'. Axial response distributions at a radial distance of 4' from the source. Measurements were made both with no water present in "the room and above a 4' diameter tank filled to a depth of 2' with water: Since these results were dominated by room return effects, they are summarized in the following section.

4.2 SSTR Room Return Response

Whenever neutron dosimetry is conducted in a laboratory bounded by walls containing moderator materials (hydrogeneous concrete in the present case), the source neutrons will be transported, scattered, and absorbed throughout the environment and particularly in the walls if the dimensions of the laboratory are small. Room return neutrons are the last vestiges of neutrons originally emitted by the source, and, indeed, these neutrons have been scattered so often that they have attained thermal equilibrium with their environment. They pervade the entire laboratory space like a uniform homogeneous mist or fog. They retain no knowledge of their origin with the exception of their intensity, which is proportional to the total emission rate of the source. This phenomenon is apparent in the data referred to previously in Section 3.0 and shown in Figures 10, 11, and 12.

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A detailed analysis of the room return phenomenon has been given by White⁽⁴⁾ who used diffusion theory to describe experimental data obtained in a laboratory bounded by concrete walls. According to this analysis, the thermal neutron flux can be expressed as

$$\phi = C \cdot S_n / R^2 , \qquad [1]$$

where S_n is the neutron emission of the source in neutrons/sec, C is a constant and R is the effective radius of the laboratory given by

$$R^{-2} = 1/6 \sum_{i=1}^{6} D_{i}^{-2}$$
, [2]

where D_i , i = 1, . . . 6 are the six distances from the source to each wall of the laboratory. A value of R that is pertinent to the TMI-2 Demineralizer A Cubicle can be derived from the known dimensions of the cubicle and the approximate location of the fuel as derived from the Compton gamma ray spectrometer data.⁽¹⁾ These gamma-ray results show that the source can be approximately represented by a point source which lies two feet from the bottom of the demineralizer tank and very close to the north side of the tank. On this basis, one finds a value of approximately 4.27' for R.

In order to experimentally determine the room return response of the SSTR dosimeters, calibrations were carried out in a cubicle where similar values of R would be obtained. With the 252 Cf source at 2' as described in Section 4.1, the corresponding value of R is 3.59' and with the source at 3.5', the corresponding value of R is 4.33'.

The radial response results for source heights of 2' and 3.5' are shown in Figures 16 and 17. Here, ²³⁵U dosimeters with no albedo devices were used, and the response is seen to be roughly constant for both cases with the exception of the 2' radial measurement with the source at 2' which corresponds to the smallest source to dosimeter distance and the highest response. Axial response distributions at a radial distance of 4' are shown for the 2' and 4' source heights in Figure 18. In both cases, the response is flat and constant for both source heights. Apparently, equation [1] is not valid for the small room dimensions addressed here, and the constant response of the two calibration cases must be applied to the demineralizer data.

In a separate measurement, the intensity of the 252 Cf source was calibrated and found to be(3.08 ± 0.42)x 10⁸ n/sec by measuring the induced fission rate in a 238 U-SSTR dosimeter. The response to this source of an albedo-type dosimeter identical to that used at TMI-2 Demineralizer A is 74.8 ± 7.5 tracks/cm²/min.

In an exposure of slightly less than 29 days, a room return response of 5.2 \pm 1.9 tracks/cm² was obtained or 1.26 x 10⁻⁴ tracks/cm²/min. This response corresponds to a total neutron source in the demineralizer A Cubicle of 517 neutrons/ second. If an average neutron specific activity of 300 n/sec/kg is assumed for the TMI-2 fuel, an SSTR neutron source intensity derived value of fuel debris of 1.72 \pm 0.63 kg is obtained.

This SSTR estimate is subject to error from a number of sources. The most important effects that have not been taken into account are the absorption and moderation of neutrons in the laboratory. These effects are particularly significant when homogeneous media exists in the laboratory as in the case for the Demineralizer

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FIGURE 16. Response as a Function of Radial Location for SSTR Neutron Dosimeters Exposed to a ²⁵²Cf Source Suspended at 2' from the Floor in the Center of a Concrete Cubicle with Overall Dimensions 11.5' x 8' and a Height of 8'.



FIGURE 17. As in Figure 16, with a Source Height of 3.5'.

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FIGURE 18. SSTR Neutron Dosimeter Axial Responses at a Radial Distance of 4' from a 252 Cf Source.

X Source Height 2' O Source Height 3.5'

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A cubicle. Neutron absorption decreases the number of neutrons which attain thermal equilibrium in the cubicle. On the other hand, moderation by a hydrogeneous medium produces a softer neutron spectrum incident upon the laboratory walls, thereby increasing neutron reflection and the thermal neutron room return flux. Consequently, these two effects work in opposite directions and tend to cancel each other. Obviously, the specific laboratory environment and geometry will play a dominant role in the relative weighting of these effects.

In order to explore the effect of having hydrogeneous media present, SSTR calibration measurements were done with the ²⁵²Cf source suspended at a height of 2' at the surface of the water in a 2' high by 4' diameter tank. The radial dosimeter response data are shown in Figure 19. The response is less by a factor of about two from that shown in Figures 16 and 17, but it is still flat. The decrease in intensity is due to thermalization and absorption of source neutrons in the water which subtends about 50% of the geometry of the source. The axial response distribution at a distance of 4' is shown in Figure 20. As in the corresponding axial distributions of Figure 18, no albedo devices were used. Below the level of the water, the response is quite low due to neutron absorption in the water. From 2' to 5' the response is reasonably flat and above 5' increases slightly as the 8' ceiling of the cubicle is approached. This measurement was repeated using albedo dosimeters identical to those used in the measurements in the Demineralizer A cubicle, yielding the data plotted in Figure 21. The shape of this albedo response is qualitatively similar to the axial distribution of Figure 10 obtained for Demineralizer A.

The peak to room return ratio obtained in the demineralizer cubicle is much higher than the ratio obtained in Figure 21, indicating that the HEDL calibration experiment

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FIGURE 19. Response as a Function of Radial Location for SSTR Neutron Dosimeters Exposed to a ²⁵²Cf Source Suspended at the Center of the Surface of a 2' High by 4' Diameter Tank of Water.



FIGURE 20. SSTR Neutron Dosimeter Axial Response at a Radial Distance of 4' to a ²⁵²Cf Source Suspended at the Center of the Surface of a 2' High by 4' Diameter Tank of Water.





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only qualitatively mocks up the demineralizer measurements. A direct calibration of the peak dosimeter response is not attempted here because of uncertainties in the neutron spectrum and corresponding spectrum averaged cross section for the fission response of the dosimeter. On the basis of the calibration data obtained, the room return neutron estimate of the amount of fuel debris in TMI-2 Demineralizer A should be regarded as a lower limit, with an upper limit that does not exceed this value by more than a factor of two.

5.0 Summary

Based on the room return response of the SSTR neutron dosimeters the amount of fuel present in TMI-2 Demineralizer A is estimated to be 1.7 ± 0.6 kg. This estimate assumes that the demineralizer is dry above the 309' level, an assumption that is supported by the shape of the axial response data. The presence of unknown amounts of water and boron below the 309' level could increase this estimate by a maximum of a factor of two.

This estimate is consistent with the value 1.3 ± 0.6 kg obtained in independent Compton recoil gamma ray spectrometry measurements.⁽¹⁾ Source distribution data from these gamma ray measurements were used to guide the direction and planning of the SSTR calibration measurements. In turn, SSTR evidence that the demineralizer tank is dry above the 309' level provided guidance for reduction of the Comptonrecoil spectrometer data. Thus, the two methods provide independent but complementary data.

The present SSTR estimate of the fuel quantity could be refined by the following:

(1) Improved knowledge of the neutron specific activity of TMI=2 fuel. The 300 n/sec/kg estimate is based on calculations. Previous experience^(5,6)

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has shown that such estimates are lower than what is found experimentally. Also, the neutron specific activity will vary with as irradiated fuel reactor core location. Since an estimated average value has been used for this analysis, measurements of the neutron specific activity of representative samples of TMI-2 fuel should be made.

- (2) Further calibrations using experimental mockups containing moderators and absorbers that are more characteristic of the contents of the demineralizer tank.
- (3) Further scanning of the mica SSTR dosimeters to obtain more tracks and the concomitant higher statistical accuracy. Also, the CR-39 SSTR could be processed, scanned, and analyzed to obtain data on the high energy (E > 0.5 MeV) neutron flux.
- (4) Neutron transport calculations using a realistic model of the demineralizer cubicle, tank and contents. This could provide further insight into the actual contents in the demineralizer A below the 309' elevation.

The present fuel estimates resulted from neutron dosimetry measurements in a field where the neutron flux was less than 3×10^{-3} neutrons/cm²/sec. Thus, the high sensitivity and cost effectiveness of the SSTR method makes it an excellent candidate for further TMI-2 applications involving fuel debris tracing by neutron dosimetry.

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