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#### APPLICATIONS OF SOLID STATE TRACK RECORDER NEUTRON DOSIMETRY FOR FUEL DEBRIS LOCATION IN THE THREE MILE ISLAND UNIT 2 MAKEUP AND PURIFICATION DEMINERALIZERS

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# ABSTRACT

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 and auxilliary systems of the reactor. The presence of fuel may be traced by using the neutron activity which is associated with the burn-in of higher actinides (about 300 neutrons/sec/kgU). Solid state track recorder (SSTR) neutron dosimetry is the most sensitive technique for measuring low neutron fluxes. Hence, neutron dosimetry is being performed at TMI-2 to locate fuel debris and subsequently aid the reactor recovery effort. Herein, the results of a scoping measurement on the fuel content of TMI-2 Makeup Demineralizer A are reported along with relevant calibration measurements. The total amount of fuel estimated in Demineraizer A, 1.7 kg, corresponds to a total neutron source of about 500 neutrons/sec. At the detector positions, data were obtained with neutron fluxes as low as  $10^{-3}$  n/sec/cm<sup>2</sup>, demonstrating the extreme sensitivity of the SSTR method.

#### KEYWORDS

Solid State Track Recorders, Mica, Reactor Dosimetry, Neutron Dosimetry, Fuel Tracing.

#### INTRODUCTION

As a part of the TMI-2 reactor recovery program, the quantities and locations of fuel debris that has been dispersed into the primary coolant and auxilliary systems of the reactor must be determined. 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 Westinghouse Hanford Company TMI-2 Demineralizer Resin Removal Program, a measure of the amount of fuel debris in Demineralizer A was sought so that other phases of the ion exchange resin removal could proceed.

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

#### TMI-2 MEASUREMENTS

TMI-2 fuel has an estimaged average neutron specific activity of about 300 n/sec/kg (Vinjamuri et al., 1981; Gold et al., 1983) which originates mainly from Pu buildup. Assuming that Pu is a good 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 has been described previously (Gold et al., 1983). Briefly, enriched  $^{235}$ U foils are placed in firm contact with mica SSTRs and the neutron-induced fission fragments from  $^{235}$ U register a tracks in the mica. The track density in the mica can be used to deduce the fission density in the adjacent uranium foil and, with

appropriate calibration data, this fission rate can be used to determine the neutron fluence. The neutron fluence and duration of the exposure of the SSTRs can then be used to ascertain the amount of fuel present.

SSTR neutron dosimeters were constructed as shown in Figure 1. 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 (Gold et al., 1983). The total SSTR area of this neutron dosimeter is approximately 85 cm<sup>2</sup>.



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

These dosimeters were assembled at TMI-2 immediately prior to the exposure (Figure 2) to reduce background due to cosmic ray neutron induced fission and from spontaneous fission of the  $^{238}$ U in the uranium. Also circular (1" diameter) CR-39 SSTRs were attached to the outer surface of each dosimeter (see Figure 2) to measure the high energy (E >  $^{0.5}$  MeV) neutron flux. (The CR-39 SSTRs have not yet been analyzed.)

The arrangement of the TMI-2 Demineralizer A and B Cubicles is shown in Figure 3. 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 pene-tration #891 shown in Figure 3.

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 Figure 4. The dosimeters were inserted at 11:55 p.m. on September 14, 1982, and occupied the positions shown in Figure 5. 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. After exposure, the SSTRs were 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  $^{235}$ U track densities for this exposure are given in Table 1.



Fig. 2. SSTR Neutron Dosimeters During Assembly. Mica and <sup>235</sup>U foils are wrapped in polyethylene bags in foreground. Fully assembled dosimeters are in background.





## RESULTS

The background corrected SSTR  $^{235}$ U track densities from Table 1 are plotted in Figure 6. Shown for comparison is a curve resulting from gamma scanning (McNeece et al., 1983) of the fission product  $^{144}$ Ce activity present in the demineralizer tank. Within experimental limitations, the positions of the two peaks are the same, indicating the presence of fuel near the 309' elevation. The assumptions are made here that  $^{144}$ Ce and Pu are both 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/cm<sup>2</sup> due to cosmic radiation, the baseline for the measurements in the demineralizer cubicle is about 10 tracks/cm<sup>2</sup>. The 5 tracks/cm<sup>2</sup> difference is due to room return neutrons resulting from thermalization of source neutrons in the walls of the cubicle. The use of this room return signal to quantify the total neutron source (fuel) in the cubicle is described below.

## SSTR ROOM RETURN NEUTRON 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



\* \* \*

Fig. 4. Emplacement of SSTR Neutron Dosimeters--Measurement of Reference Point for Stringer Location.



Fig. 5. Locations of SSTR Neutron Dosimeters During Exposure.

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.

A detailed analysis of the room phenomenon has been given by White, 1966, 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

$$\phi = C \cdot S_n / R^2 \quad , \tag{1}$$

Elevation*	Measured Tracks/cm <sup>2</sup>		Measured Tracks/cm <sup>2</sup>
(Ft.)	Obs. 1	Obs. 2	Avg.**- B. G.
317	11.1		6.1 <u>+</u> 2.0
312	26.1		21.1 <u>+</u> 2.8
311	25.2	27.0	21.1 + 2.7
310	30.1	30.6	25.3 <u>+</u> 3.2
309	36.8	36.1	31.5 <u>+</u> 4.2
308	13.3	12.4	7.9 <u>+</u> 1.7
307	10.2		5.2 + 1.9

 TABLE 1
 Measured SSTR Track Densities at selected Locations Along The

 Vertical Traverse (Data Taken in TMI-2 Demineralizer a Cubicle)

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Demineralizer D Measured Background (8. G.) 4.8 5.3 (B. G. Average =  $5.0 \pm 1... **$ )

\*The elevation corresponding to the bottom of the demineralizer tank is 307 ft.

\*\*Average of Observers 1 and 2.



Fig. 6. Track Density as a Function of Axial Location for SSTR Neutron Dosimeters Exposed in TMI-2 Demineralizer A Cubicle in Comparison with <sup>144</sup>Ce Gamma Activity Obtained from Continuous Gamma Ray Spectrometry (See McNeece et al., 1983)

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

$$R^{-2} = 1/6 \sum_{i=1}^{6} D_i^{-2} , \qquad (2)$$

where  $D_i$ ,  $i = 1, \ldots 6$  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 (McNeece et al., 1983). 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 the basis, one finds a value of approximately 4.27' for R.

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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 a  $^{252}$ Cf source suspended at a height of 2' in this cubicle, the corresponding value of R is 3.59' and with the source at 3.5', the corresponding value of R is 4.33'. These two heights bracket the effective radius value for the TMI-2 Demineralizer A Cubicle neutron source.

The radial response results for source heights of 2' and 3.5' are shown in Figures 7 and 8. Here,  $^{2350}$  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 3.5' source heights in Figure 9. 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.



Fig. 7. Response as a Function of Radial Location for SSTR Neutron Dosimeters Exposed to a <sup>252</sup>Cf 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'.



Fig. 8. As in Figure 7, with a Source Height of 3.5'.





X Source Height 2' O Source Height 3.5'

In a separate measurement, the intensity of the  $^{252}$ Cf source was calibrated and found to be (3.08 + 0.42) x 10<sup>8</sup> n/sec by measuring the induced fission rate in a  $^{238}$ U-SSTR dosimeter. Since  $^{238}$ U(n,f) is a threshold reaction, the fission rate will closely follow an inverse square law with distance from the source. The response to this source of an albedo-type dosimeter identical to that used at TMI-2 Demineralizer A is 74.8 ± 7.5 tr;cks/cm<sup>2</sup>min.

In an exposure of slightly less than 29 days, a room return response of  $5.2 \pm 1.9$  tracks/cm<sup>2</sup> was obtained or  $1.26 \times 10^{-4}$  tracks/cm<sup>2</sup>/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 hydrogenous media exist in the laboratory, as is the case for the Demineralizer Cubicle. Neutron absorption decreases the number of neutrons which attain thermal equilibrium in the cubicle. On the other hand, moderation by a hydrogenous 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  $^{252}$ Cf source suspended in the cubicle described previously 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 10. The response is less by a factor of about two from that shown in Figures 7 and 8, 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 11. As in the corresponding axial distributions of Figure 9, no albedo devices were used. Below the level of the water, 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 12. The shape of this albedo response is qualitatively similar to the axial distribution of Figure 6 obtained for Demineralizer A.

The peak to room return ratio obtained in the demineralizer cubicle is much higher than the ratio obtained in Figure 12, indicating that the HEDL calibration experiment 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



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Fig. 10. Response as a Eunction of Radial Location for SSTR Neutron Dosimeters Exposed to a  $^{252}$ Cf Source Suspended at the Center of the Surface of a 2' High by 4' Diameter Tank of Water.



Fig. 11. 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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Fig. 12. As in Figure 11 but with Albedo Dosimeters of the Design Shown in Figure 1.

the basis of the calibration data obtained, the room return neutron estimate of the amount of fuel debris in IMI-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.

#### SUMMARY

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The SSTR intensity distribution in Figure 6 is asymmetric, decreasing less rapidly above the 309' elevation than below. The 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.

Independent Compton Recoil Gamma Ray Spectrometry measurements (McNeece et al., 1983) resulted in a fuel estimate of  $1.3 \pm 0.6$  kg. This result is in excellent agreement with the SSTR estimate. Whereas SSTR evidence that the demineralizer tank is dry above the 309' level provided guidance for reduction of the Compton-recoil spectrometer data, source distribution data from these gamma ray measurements were used to guide the direction and planning of the SSTR calibration measurement. Thus, the two methods provided independent but complimentary data.

The track densities observed in the SSTR neutron dosimeters correspond to extremely low neutron fluxes. The total neutron emission rate in the Demineralizer A Cubicle is about 500 neutrons per second, corresponding to a flux of less than 3 x  $10^{-3}$  n/cm<sup>2</sup>/sec at the position of the SSTR neutron dosimeter with the maximum signal. To our knowledge, SSTR neutron dosimetry is the only known method that could be successfully applied in such fuel debris quantification experiments. Our conclusion that the Demineralizer A tank was dry has been substantiated over six months later by gathering of samples from the demineralizer tank which showed that the tank was dry.

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