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Dr. Michael T. Masnik
U.S. Nuclear Regulatory Commission
Nuclear Reactor Regulation
Mail Stop 11, Building 20
Washington, D.C. 20555

Dear Dr. Masnik:

I am enclosing the final version of our report on the measurements of neutron emissions from the TMI-2 fuel samples. Although our measurements did not agree closely with the results of the GPU study, we do not feel that the results indicate a need to revise the conclusions of any studies performed by the licensee.

If you have any questions on this study, please feel free to call us at the above number.

Sincerely,

Robert I. Scherpelz
Senior Research Scientist
Dosimetry Research Section
HEALTH PHYSICS DEPARTMENT

RIS/ag

Enclosure

cc: LH Thonus, NRC
R Harty, PNL

THE MEASUREMENT OF NEUTRON EMISSION RATES FROM TMI-2 FUEL DEBRIS

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L. L. Nichols

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BACKGROUND

As part of their responsibility in managing the damaged Three Mile Island Unit 2 Reactor, General Public Utilities (GPU) Nuclear (the licensee) has conducted detailed surveys of the special nuclear material (SNM) remaining at TMI-2. The results of these surveys will serve as documentation for the licensee's SNM accountability. The evaluation of quantities of SNM remaining in different locations of the plant also serve as the basis for safety studies, particularly those ensuring criticality safety.

The largest quantity of SNM remaining in the TMI-2 facility after an extensive defueling effort is in the Reactor Vessel (RV). The quantity of fuel in the RV was estimated in 1990 by a study that relied on a video inspection of the internal region of the RV, which at that time was filled with water. This quantity, 630 kg, was included in the Defueling Completion Report.

Since the video inspection method included large uncertainties, and the uncertainty could not be quantified, a second measurement of the fuel remaining in the RV was conducted as the RV water was drained down. This measurement relied on the measurement of neutrons emitted by transuranic isotopes in the fuel. This method, referred to as the Passive Neutron Measurement technique, yielded an initial value of 1322 kg of fuel. The measurements were then re-evaluated and the value was adjusted to 925 kg of fuel, which is now considered to be the estimate of record for the RV fuel contents.

Two basic types of reactions account for the neutrons emitted by the fuel. One reaction is spontaneous fission, occurring in several isotopes of uranium and plutonium. The other reaction is (α,n), where an alpha particle emitted by a transuranic isotope in the fuel strikes either an oxygen or a boron atom and ejects a neutron. While the rate of spontaneous fission reactions are very predictable if the transuranic isotopes are known, the rate of (α,n) reactions is very difficult to predict, since they depend on factors such as fuel particle size and boron concentration. Thus a crucial step in the passive neutron measurement technique was a calibration step in which the neutron emission rate from representative fuel samples was measured. This neutron emission rate was used in the analysis of measurements in the RV to convert the number of neutrons measured into a fuel quantity.

Since the measurement of neutron emission from fuel samples was so important to the estimate of fuel in the RV, staff from the Pacific Northwest Laboratory (PNL) performed an independent measurement of the fuel samples to determine the neutron emission rate. The measurements were performed at Idaho National Engineering Laboratory (INEL) where the fuel samples are now stored. INEL staff also performed measurements, using an instrument designed at INEL, in parallel with the PNL study. This report documents the PNL measurements and compares them to the values determined by INEL and by the licensee.

DESCRIPTION OF MEASUREMENT EQUIPMENT

The measurements performed by PNL used the Precision Long Counter (PLC) (DePangher and Nichols, 1966), which consists of moderating material surrounding a detector that responds to thermal neutrons. A diagram of a PLC, shown in Figure 1 (Page 4), illustrates that the detector is a BF_3 tube, and the moderator is polyethylene. The PLC was described in detail in DePangher and Nichols, and it has been used extensively as a neutron flux meter for over 25 years.

Signals from the BF_3 tube were routed through a preamplifier, then through a linear amplifier, and collected in a multichannel analyzer (MCA). In many routine uses of the PLC, signals are collected in a simple scaler rather than an MCA, because the neutron counting depends on a simple count rate from the detector rather than a distribution of pulse heights. However, an MCA was used for this experiment to ensure that an appropriate region of interest (ROI) was chosen for the count. Using this equipment, the experimenters could be sure that the neutron counts would not be contaminated by gamma counts, which is a concern with fuel samples that emitted high levels of gamma radiation. This equipment also allowed the experimenters to set an ROI that ensured the best selection of events for the very-low-count-rate measurements and guarantee that all legitimate neutron events were counted.

MEASUREMENT OF NEUTRON CALIBRATION SOURCES

The first series of measurements was performed to determine the neutron counting efficiency of the PLC. The efficiency value relates the number of neutron counts recorded by the detector to the actual neutron flux at the detector.

The efficiency measurements were performed using two neutron sources, AmBe and ^{252}Cf. The AmBe source emits neutrons with an average energy of approximately 4.5 MeV, and the ^{252}Cf source emits neutrons with an average energy of about 2.2 MeV. Therefore the two sources cover the energy range of neutrons that will be emitted by the fuel samples, and comparing efficiencies for the two sources gives an indication of the energy-related variability in the PLC's counting efficiency. Previous experience and characterization of the PLC shows that the detector has a very flat energy response curve over the neutron energy range of 1 to 5 MeV, so it should be a reliable instrument for measuring these sources.

The known AmBe and ^{252}Cf sources were counted in the "Chopper" laboratory of the MTR Building in INEL's TRA Area. Each source was placed in a lead and steel mock-up that duplicated the materials and geometry of the three storage casks that housed the TMI fuel samples. While in this mock-up each source was counted with the PLC at five or six different distances from the source. The distance from the detector face to the source was measured as shown in Figure 2 (Page 5). The counting data for these measurements are shown in Table 1.

Figure 1.

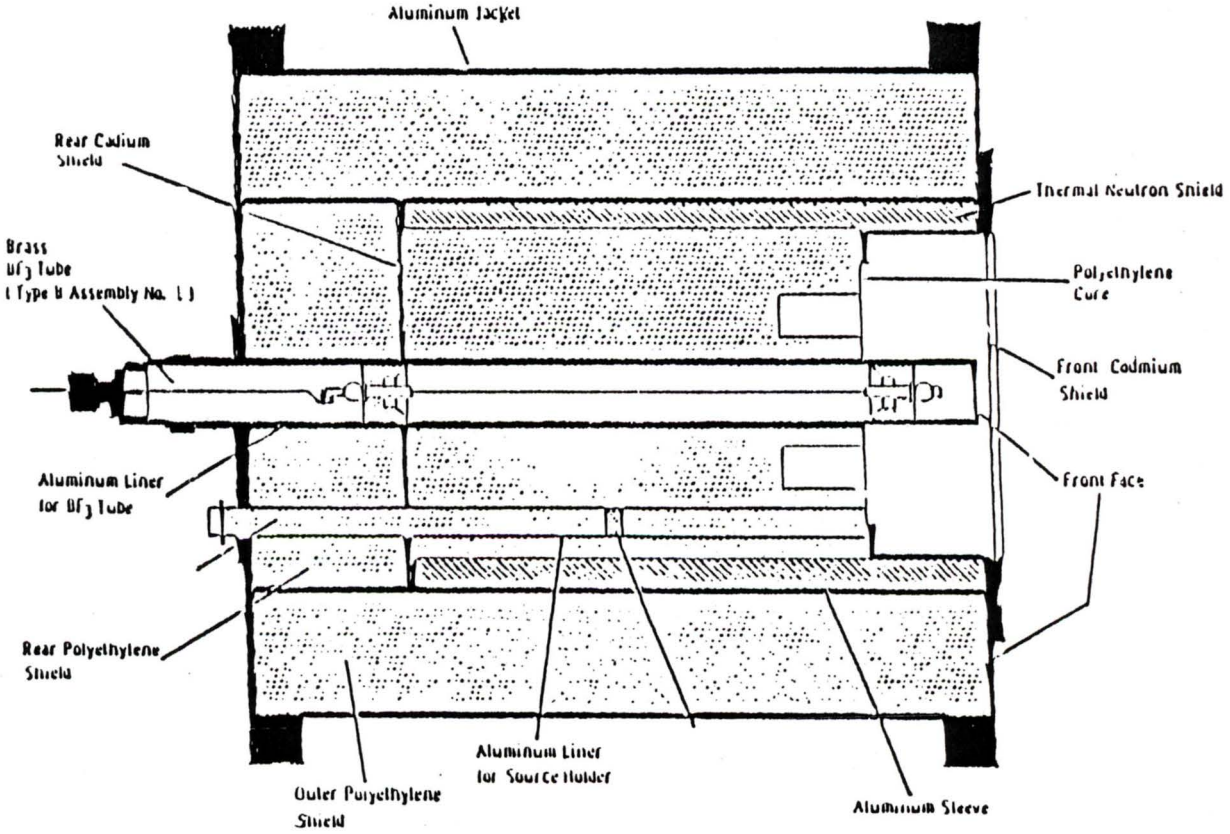

Aluminum Jacket
Rear Cadium Shield
Thermal Neutron Shield
Brass
BF3 Tube
(Type B Assembly No. 1)
Polyethylene Core
Front Cadmium Shield
Aluminum Liner for BF3 Tube
Front Face
Rear Polyethylene Shield
Aluminum Liner for Source Holder
Outer Polyethylene Shield
Aluminum Sleeve

Figure 2.

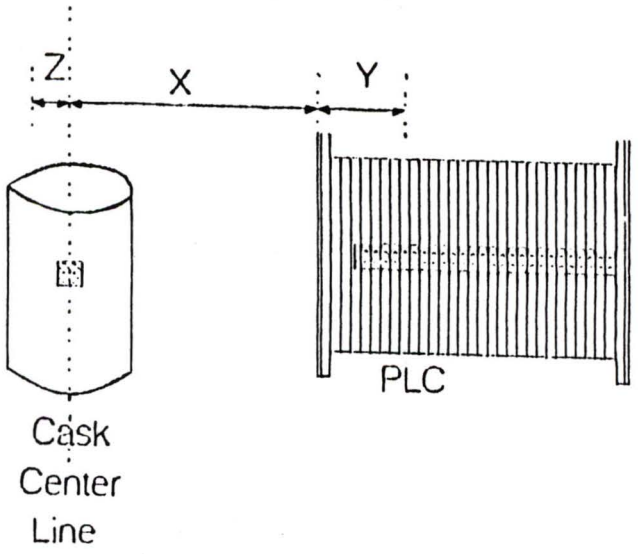


Table 1. Recorded Counts at the Neutron Calibration Sources

Neutron Source	Distance From Source (cm)	Count Time (sec)	Recorded Counts	Count Rate (cts/s)
^{252}Cf	42	300	431,865	1440
^{252}Cf	56	300	280,042	933.5
^{252}Cf	71	150	94,561	630.4
^{252}Cf	82	150	74,792	498.6
^{252}Cf	102	151	51,651	342.1
AmBe	42	300	47,300	147.7
AmBe	58	300	28,997	96.66
AmBe	71	300	20,444	68.15
AmBe	87	300	14,854	49.51
AmBe	104	300	10,928	36.43
AmBe	123	500	14,152	28.30

DATA ANALYSIS FOR NEUTRON CALIBRATION SOURCES

While in the simulated cask, each source was counted with the PLC at five or six different distances from the source. In this way the data could be forced to fit the "point source/point detector" model. With a point source and point detector, the measured count rate would vary according to an "inverse-r-squared" principle. Thus if "x" corresponds to the distance between the source and detector as shown in Figure 2, an adjusted distance, x+a, could be found so that the count rate, CR, would vary as:

$$CR = k_i/(x+a)^2$$

$$\text{or} \quad k_i = CR(x+a)^2 \qquad (1)$$

where:

a = y+z;
y = the effective detection center of the PLC; and
z = the effective detection center of the source.

For each source, an initial value for "a" was assumed, and the resulting k_i values for all distances were found by applying Equation 1 to the data in Table 1. The statistical variance was then found for the set of k_i values resulting from the assumed value of "a". The value of a was then altered, finding new k_i values and the associated variance, and the process was repeated until the minimum variance was found. The value of "a" giving the minimum variance in k_i was then used to represent the adjusted distance.

Previous work with the PLC had determined the value of the effective detection center of the PLC as equal to:

$$y = (7.8 + 1.1E) \text{ cm}$$

where E is the average energy in MeV of the neutron spectrum. If we assume 2.2 MeV and 4.5 MeV for the average energies of the ^{252}Cf and AmBe spectra respectively, then the value of y for each source would be:

$$y_{Cf} = (7.8 + 1.1*2.2) = 10.2 \text{ cm}$$

$$y_{AmBe} = (7.8 + 1.1*4.5) = 12.8 \text{ cm}.$$

If these values are subtracted from the "a" values that were determined in the minimization of the variance we get:

$$z_{Cf} = 14.8\text{-}10.2 = 4.6 \text{ cm}$$

$$z_{AmBe} = 16.9\text{-}12.8 = 4.1 \text{ cm}.$$

The neutron energy dependence of the effective center of the simulated container is expected to be small and in the direction that would make the more energetic AmBe source appear closer. Thus the above numbers appear very reasonable.

The calibration constant for the PLC using the known neutron emission rate for the ^{252}Cf source is:

$$4.98\mu\text{Ci} \times 2.4\text{x}10^6 / 4.63\text{x}10^6 = 2.58 \text{ neuts/count-cm}^2;$$

and for the AmBe source:

$$1.27\text{x}10^6 / 5.39\text{x}10^5 = 2.36 \text{ neuts/count-cm}^2.$$

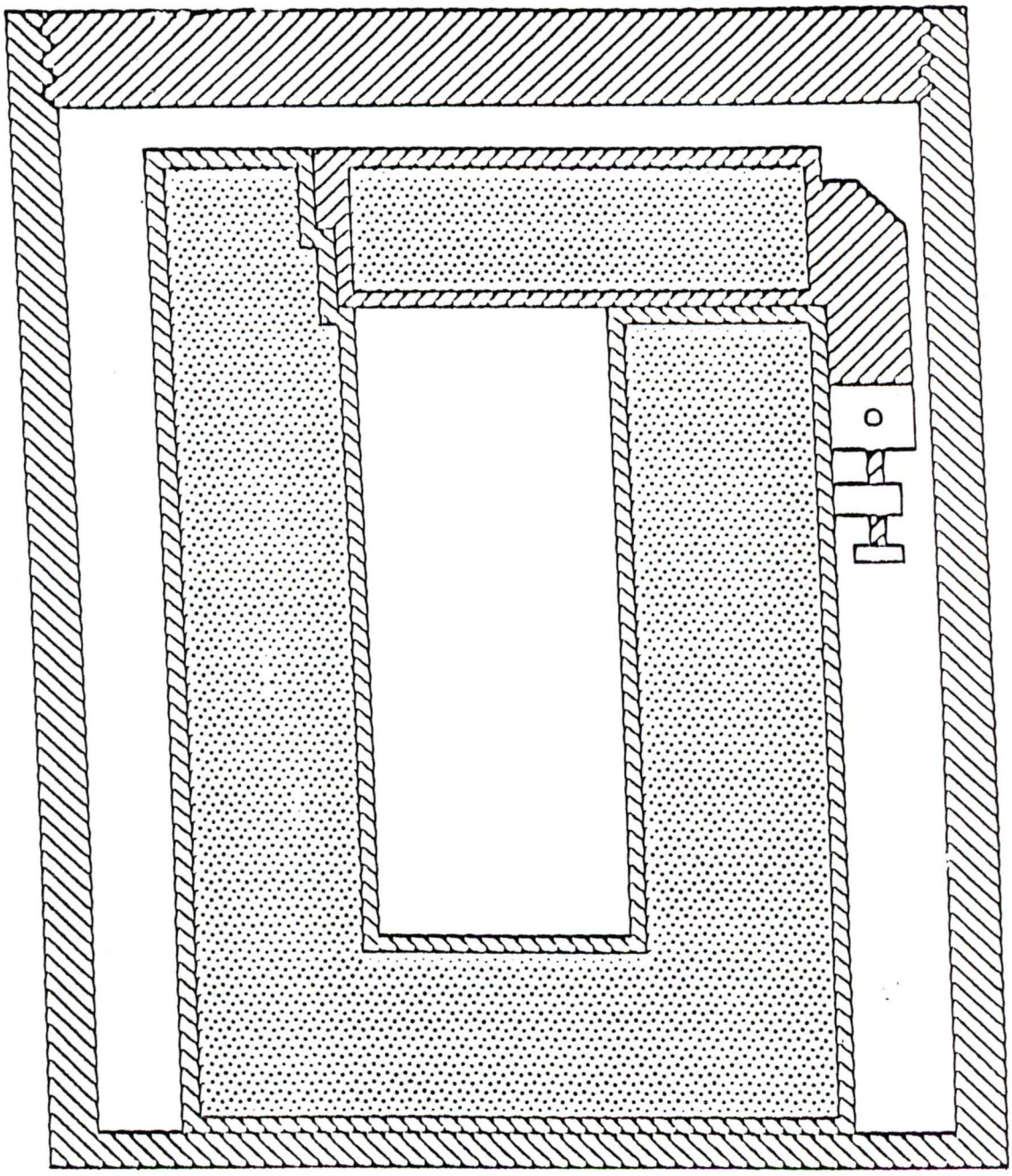
If we give equal weight to these two values and average them, we get efficiency of PLC = 2.47 neuts/count-cm^2.

MEASUREMENT OF FUEL SAMPLES

Measurements were performed on three TMI fuel samples in the ATRC Building, TRA Area at INEL. The nine fuel samples used in the original GPU study had been consolidated into three lead-and-steel casks. The samples could not be removed from their casks for the count because of the potential for some of the fuel particles to escape, so the fuel samples could not be counted individually as they were in the GPU study. The results of the counts could still be averaged, however, to obtain neutron emission rates that would be representative of fuel remaining in the TMI-2 RV.

The fuel samples were contained in a lead pig that was itself inside a type 2R shipping container, as shown in Figure 3 (Page 8). Only one 2R cask was brought to the ATRC Building at any one time to keep the neutron background as low as possible in the counting area. The configuration of the fuel samples in the 2R cask was similar to the cask mockup used with the calibration

Figure 3.



sources to eliminate any neutron attenuating effect of the lead and steel. During the fuel sample counts in the ATRC, additional lead blankets were draped over the cask, covering the sides not facing the detector to minimize personnel exposure. However, the lead blankets did not present any additional neutron attenuation that could influence the counts.

The counts that were made on the three unknown fuel samples were all done at a fixed distance from the source because the count rates were so low there was not time to do a full inverse square analysis for each source. Each fuel sample was counted until at least 1000 counts had been recorded. Two long background counts were also made. These data are shown in Table 2.

Table 2. Counting Data for the TMI-2 Fuel Samples

Sample	Distance from Source (cm)	Count Time (sec)	Recorded Counts	Count Rate (Cts/s)
Cask #1	73.8	15,648	2,576	0.1646
Cask #2	75.4	7,267	993	0.1366
Cask #4	75.1	16,501	1,742	0.1056
Bkgd	---	61,731	2,650	0.0429
Bkgd	---	62,028	2,630	0.0424

DATA ANALYSIS FOR FUEL SAMPLES

Using the recorded count data in Table 2, the neutrons per second emitted by each fuel sample cask can be determined by evaluating

$$CR * (x+a)^2 * E$$

where CR is the counts per second minus the average background count, (x+a) is the effective source-to-detector distance, and E is the PLC efficiency that was determined by counting the AmBe and ^{252}Cf sources. The value of "a" was also derived from the measurements of the known neutron sources.

The calculated neutrons per second emitted by the fuel samples in each cask are:

Cask #1 $(.165-.043)(73.8+15.8)^2*2.47=2419$ neutrons/second

Cask #2 $(.137-.043)(75.4+15.8)^2 * 2.47=1931$ neutrons/second

Cask #4 $(.106-.043)(75.1+15.8)^2*2.47=1286$ neutrons/second.

Each cask held one or more fuel samples. The masses for the samples were derived from (GPU, 1991). Table 3 shows the samples in each cask and gives the mass of uranium that was measured by INEL staff in each cask:

Table 3. Contents of the TMI Fuel Sample Casks

Cask #	Sample ID #	GPU-Measured Sample Fuel Mass (grams)	INEL-Measured Cask Total Fuel Mass (grams)
1	RD-SSC-1-2	129.7	
1	RD-SSC-1-8	1534.5	
1	RD-SSC-1-9	657.0	
1	RD-SSC-1-10	239.7	
1	RD-SSC-1-11	851.7	
1	RD-SSC-1-12	433.5	
1			4170
2	RD-SSC-1-3	1608.1	
2	RD-SSC-1-5	1960.7	
2			3645
4	RD-SSC-1-7	2048.0	1828

GPU had measured the mass of each of the nine samples listed above (GPU Nuclear, 1991), and used the results of an INEL analysis (GPU Nuclear, 1989) to determine the uranium mass in each sample. This mass for each cask is listed in Table 3 for each individual sample. After INEL analyzed the nine samples for particle size and uranium content, INEL staff repackaged the samples into the three casks that were ultimately used for the PNL/INEL study. The repackaging kept the bulk of each of the nine samples together as listed in Table 3, but some of the sample material had been consolidated during the INEL analyses and was not necessarily repackaged with the bulk of the original sample. Thus the INEL-measured cask totals did not match the GPU values. It should also be noted that the sum of the uranium measured by INEL in the three casks is greater than the sum of the nine masses measured by GPU, which is surprising. INEL had stated, however, that uncertainties in the uranium contents were about 10%, and the disagreement in masses is less than 2%. This study will use the cask masses measured by INEL, since the casks were not opened after the INEL repackaging, so these values are most likely to represent the actual contents of the cask as they were used by the PNL/INEL study.

The neutron yield per gram of U is:

Cask #1: 2419/4170 = .58 neutrons per second per gram;

Cask #2: 1931/3645 = .53 neutrons per second per gram; and

Cask #4: 1286/1828 = .70 neutrons per second per gram.

MEASUREMENT UNCERTAINTIES

Uncertainties in the determination of neutron emission rates came from several different sources. The most important uncertainties are discussed in the following sections. In this report, all uncertainties are stated as one-sigma values.

Calibration Sources

The neutron emission rates from the ^{252}Cf and AmBe sources were important factors in the determination of the detector efficiency. INEL estimated the uncertainty in the sources' neutron emission rates as 5%.

Counting Geometry

Several different features in the geometrical arrangement of the sources, detector positions, and other materials create uncertainties in the measurements.

Detector Position

The distance between the face of the PLC and the centerline of the source cask was measured with a meter stick, and in some cases the positions were difficult to access. The detector location was known to within ±1 cm, which could produce errors as large as 3% in the range of positions used in this study.

Sample Position

The calibration sources were positioned in the center of the cask mockups, but the exact positioning of the uranium samples inside the 2R casks could not be known. In the INEL study this effect was studied by placing a ^{252}Cf source in a number of different positions inside the cask mockup, and a 22% variation was found in the detector efficiencies. The effect should be much smaller in the PNL measurement, since the detector was further from the cask. An inverse-square argument would show that a neutron source placed at the closest position in the 3-inch-diameter cavity would produce a count rate 23% higher than a source place in the position farthest from the 75-cm-distant detector. A 23% error would correspond to an implausible sample configuration however, so a more reasonable uncertainty would be 12%.

Neutron Moderation and Attenuation

As neutrons encounter scattering and absorbing material, some neutrons are removed from the field, some are scattered back into the detector region, and some experience changes in energy. All of these can effect the neutron counting.

Cask Walls

The walls of the 2R storage cask affected the neutron radiation passing through it. This effect should be nullified by the mockup of the 2R cask used around the calibration sources, but the INEL study used computer modeling to estimate that the difference between the 2R cask and its mockup could introduce a 5% uncertainty.

Room Scatter

Neutrons emitted by the sources could reflect from moderating surfaces and produce counts in the detectors. This effect will be dominated by the concrete floors, since the walls and other moderators were further from the detectors. The effect of room scatter should be minimized by positioning both the calibration sources and the uranium samples at the same height above the floors, but room scatter may have contributed a 3% uncertainty.

Neutron Energies

The response of the PLC is fairly uniform with respect to neutron energy. There is some variation in the response, however, as shown by the fact that different PLC efficiencies were determined for the ^{252}Cf source and the AmBe source. The energy distribution for the neutrons emitted by the uranium samples is similar to the calibration neutrons, but not well-characterized. Since the two efficiencies were 9% apart (2.36 versus 2.47), a 9% uncertainty is justified.

Counting Statistics and Background

For the calibration measurements, large number of counts were collected compared to a minimal background. Counting statistics uncertainties were thus less than 0.1%. For the uranium sample measurement, the worst case was a 3.4% uncertainty.

Uranium Sample Mass

The debris samples were weighed by GPU during their measurements in 1989, then analyzed by INEL for uranium content. GPU states that the sample weights were accurate within 5 grams (GPU 1991), and INEL places the uncertainty in the uranium content at 10% to 15% (GPU 1989). After the GPU emission rate measurements were performed, the samples were subjected to analysis and repackaging, and INEL measured the sample weights as they were repackaged. The uranium weights used in this study do not agree with the values used by GPU, but the value falls well within the stated uncertainty, and an uncertainty of 15% appears to be appropriate for this study.

Overall Uncertainty

A summary of the components for the measurement uncertainty is given in Table 4.

Table 4. Contributors to Measurement Uncertainty

Item	Uncertainty
Calibration source	5%
Detector position	3%
Sample position	12%
Cask walls	5%
Room scatter	3%
Neutron energies	9%
Counting statistics	3.4%
Uranium sample mass	15%
Total	23%

The total measurement uncertainty was determined by taking the square root of the sum of the squares of the other uncertainties. The overall uncertainty for these determinations of neutron emission rate is 23%.

COMPARISON OF PNL, INEL AND GPU NEUTRON EMISSION RATES

The neutron emission rates for the three casks as determined by the PNL and the INEL measurements are presented in Table 5.

Table 5. Summary of Neutron Emission Rates Measured by PNL and INEL

Cask #	Cask Fuel Mass (g U)	Measured Neutron Emission Rate PNL n/s per g U	Measured Neutron Emission Rate INEL n/s per g U
1	4170	0.580	0.466
2	3645	0.530	0.423
4	1828	0.704	0.473
Weighted Average		0.584±.134	0.451±.117

The weighted averages listed on the bottom lines of Table 5 were calculated using the cask fuel mass as a normalizing factor. The error bounds listed with the weighted averages were calculated using the uncertainty values of 23% for the PNL measurements and 26% for the INEL measurements.

The values in Table 5 were calculated based on the quantity of U in each fuel sample. If they were adjusted to be in terms of UO_2, the values would be lowered and they would compare to the value used by GPU as shown in Table 6:

Table 6. Average Measured Neutron Emission Rates

Study	Emission Rate n/s per g UO_2
GPU	0.62 to 0.70
PNL	0.40 to 0.63
INEL	0.29 to 0.50

The range of emission rates given in Table 6 was taken from the uncertainty values quoted by each study (6% for GPU, 23% for PNL, 26% for INEL).

DISCUSSION AND CONCLUSIONS

All three measurements, GPU, PNL and INEL, used similar types of neutron detectors. All three systems included a detector that responded to thermal neutrons surrounded by a moderator. The PLC used by the PNL study has been used extensively for measuring the neutron flux in a variety of counting situations in neutron fields. It has proven to be a reliable instrument in similar situations. The INEL instrument has been the subject of sophisticated computer modeling and its response to neutron fields is well-understood. Thus it is surprising that the neutron emission rates measured by the three different instruments did not show better agreement.

Two possible differences have been identified which may account for the discrepancies in the neutron emission rates determined by GPU versus those determined by the PNL/INEL measurements.

First, there is some uncertainty in the total masses of the samples in the three measurement casks. The mass values used by GPU were determined during the 1989 measurements at TMI, but since then the samples were subjected to analysis and repackaging. The masses of uranium recorded by INEL staff as they repackaged the material were larger than the masses used by GPU staff. Thus the mass discrepancies contribute to the discrepancy in measured emission rates. However, the mass discrepancies are only about 2%, which is within the stated error bounds.

A second difference between the GPU measurements and the measurements performed by both PNL and INEL was that GPU measured the nine fuel samples individually, arriving at neutron emission rates for each of the nine samples. In the PNL/INEL study, the nine samples had been consolidated into three containers so that only three measurements were made. The results are still valid, of course, since GPU averaged the results of their samples to get one final neutron emission rate, and by consolidating the samples they achieved an average value. However, GPU had omitted the measured results from one of its samples, #RD-SSC-1-9, because they believed that the accepted mass of fuel in this sample was suspect (reviewing the analysis data reveals a probability that the mass of uranium in this sample is more than 657 g). PNL/INEL could not, of course, omit this sample, since it was sealed in Cask #1. Thus the comparison of neutron emission rates between PNL/INEL and GPU have this slight discrepancy in samples.

The PNL and INEL measurements were very similar to each other, and any bias that existed in the sample mass applied equally to both measurements. Tables 5 and 6 show that the error bounds for the measured neutron emission rates measured by the two studies overlap each other, but the results also showed that the PNL study consistently measured higher neutron count rates than the INEL measurements. The PNL results were nearly 30% higher than the INEL results.

One difference between the INEL and PNL measurement schemes was the strategy of detector placement. The INEL detector was located very near the cask to maximize the count rate, while the PNL detector was located about 75 cm from the cask. The PNL positioning resulted in a much lower number of neutron counts detected, but it decreased the influence of the positioning of neutron emitters inside the cask. The low number of counts recorded by the PNL detector resulted in a counting statistics uncertainty of 3.4% (while the corresponding uncertainty for the INEL counts was nearly 0). On the other hand, the INEL study found a 26% uncertainty in the measured efficiencies due to the positioning of the source inside the cask. PNL's corresponding uncertainty was only 12%.

A second possible reason for a discrepancy between the PNL and INEL results is the uncertainty in the location of the uranium samples inside the 2R casks. Both studies assumed that the casks were homogeneously loaded with sample material so that the effective center of the measured uranium was at the center of the cask. The PNL and INEL detectors were positioned at opposite sides of the cask from each other, so that if the effective center of the uranium in the counted cask were closer to the PNL detector than to the INEL detector, the PNL detector would experience a higher count rate. This effect may have been present, but there are two reasons why it is not likely to be a large effect. First, since PNL measured higher count rates than INEL for all three casks, all three would need to have hot spots that, when randomly positioned, ended up near the PNL detector. Second, the inner volume of the cask was 898 cm^3, and knowing the mass of sample weights and densities concludes that each of the casks contained at least 400 cm^3 of debris. Thus the only way to postulate a hot spot in the casks would be to assume a very non-uniform distribution of uranium in the sample material.

GPU expressed a 1σ error bound of 6%, which would make a 1σ range in neutron emission rates of 0.62 to 0.70. On the other hand, the PNL and INEL studies set uncertainty levels of 23% and 26% on their determinations of uranium neutron emission rates. The GPU error bound was based only on the spread of emission rates determined for the eight different samples that they studied. This small uncertainty ignored the fact that the uranium contents of the samples have an uncertainty of 15%. The GPU error bound should be much larger.

The results of this study do not provide any justification for choosing one of the measured neutron emission rates above another. The PNL authors believe that their extensive experience with the PLC in similar types of counting situations lends credence to their measurement, but it would require a careful comparison test to favor one measurement system over another, and any such test has not been performed.

Both the PNL and INEL measurements indicate that there is a chance that the neutron emission rate measured by GPU may be a high estimate. If the neutron emission rate were lowered, however, it would result in a higher estimate of the fuel remaining in the RV. Again, the PNL/INEL measurements did not provide conclusive proof that the RV fuel estimate should be increased. The study did show that the uncertainty assigned by GPU to its measurement of the neutron emission rate should be enlarged.

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