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AUTHOR(S): M. S. Moore, P. E. Koehler, A. Michaudon, A. Schelberg, Y. Danon, R. C. Block, R. E. Slovacek, R. W. Hoff and R. W. Lougheed

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EXPERIMENTAL ASSESSMENT OF THE PERFORMANCE OF A PROPOSED LEAD SLOWING-DOWN SPECTROMETER AT WNR/PSR*

M. S. Moore, P. E. Koehler, A. Michaudon, and A. Schelberg**
University of California, Los Alamos National Laboratory
Los Alamos, NM 87545 USA

Y. Danon, R. C. Block, and R. E. Slovacek
Rensselaer Polytechnic Institute
Troy, NY 12181 USA

R. W. Hoff and R. W. Lougheed
University of California
Lawrence Livermore National Laboratory
Livermore, CA 94551 USA

ABSTRACT

In November 1989, we carried out a measurement of the fission cross section of ^{247}Cm, ^{250}Cf, and ^{254}Es on the Rensselaer Intense Neutron Source (RINS) at Rensselaer Polytechnic Institute (RPI). In July 1990, we carried out a second measurement, using the same fission chamber and electronics, in beam geometry at the Los Alamos Neutron Scattering Center (LANSCE) facility. Using the relative count rates observed in the two experiments, and the flux-enhancement factors determined by the RPI group for a lead slowing-down spectrometer compared to beam geometry, we can assess the performance of a spectrometer similar to RINS, driven by the Proton Storage Ring (PSR) at the Los Alamos National Laboratory. With such a spectrometer, we find that it is feasible to make measurements with samples of 1 ng for fission 1 μg for capture, and of isotopes with half-lives of tens of minutes. It is important to note that, while a significant amount of information can be obtained from the low resolution RINS measurement, a definitive determination of average properties, including the level density, requires that the resonance structure be resolved.

I. THE SLOWING-DOWN SPECTROMETER AT RPI

The Rensselaer Intense Neutron Source (RINS) is generally recognized as the highest intensity neutron spectrometer in the world.[1] It consists of a (1.8 m)^3 block of highly pure lead, in the center of which is placed a neutron production target so that the spectrometer can be driven by electron pulses from the RPI electron linear accelerator. Four penetrations are provided for the insertion of experiments; these are horizontal channels located 60 cm from each of four faces of the block of lead.

The earliest use of a lead slowing-down spectrometer (SDS) was reported at the first Geneva conference (1955) by Bergmann et al.,[2] who also calculated the performance characteristics. In a lead SDS, the neutron pulse is time and energy

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**Present address EG&G, Inc., P. O. Box 809, Los Alamos, NM 87544.

focused in the slowing-down process (lower energy neutrons make fewer collisions per unit time, and thus lose less energy than higher energy neutrons in the pulse). The neutron intensity is much higher than in a standard time-of-flight experiment because the sample can interact with the same neutrons many times in the slowing-down process. A rough rule-of-thumb based on the experience of the RPI group[3] suggests that the SDS gives an intensity about 10,000 times that of a standard time-of-flight measurement at the equivalent flight path, 5.6 m, which is characteristic of the SDS made of solid lead. The energy dependence of the enhancement factor is shown in Table I.

Table I. Energy dependence of the flux enhancement of a lead slowing-down spectrometer over that of a conventional time-of-flight experiment at a 5-meter flight path, as determined by the RPI group.[3]

Neutron Energy (eV)	Φ_{RINS}/Φ_{TOF}
1000	5800
100	7800
10	10500
1	14300

According to Bergmann's calculations, the resolution of the SDS should be about 30% in energy. In practice, the RPI group found that 35-40% in energy is to be expected, because of the presence of light element contaminants. Even a small amount of hydrogen degrades the resolution to the point that the instrument is no longer useful as a spectrometer.[4]

While Bergmann et al., used the SDS for radiative capture cross-section measurements, the RPI group has concentrated on fission. One of the early triumphs was the first demonstration of subthreshold fission in (^{238}U + n).[5] More recently, they have completed a series of measurements of the resonance fission cross sections of the curium isotopes,[6] using a special hemispherical fission chamber suggested by N. W. Hill[7] to minimize alpha pileup.

One problem the RPI group has experienced involves target cooling. A hydrogenous coolant such as H_2O cannot be used, so in RINS the target is cooled by flowing helium gas. However, bremsstrahlung heating of the lead limits the beam power on target to <1 kW. In an experiment in which the background is dominated by radiation from the sample (alpha pileup or spontaneous fission in a fission measurement, or gamma emission in a capture measurement), the signal-to-noise ratio is maximized if the beam power per pulse is as high as can be achieved, with the repetition rate adjusted to give the maximum allowable average power. In a recent typical measurement,[8] the RPI linac operated at 60 MeV with an average current of 15 μA (giving 0.9 kW on target), achieved with 200 ns wide electron bursts at a repetition rate of 90 Hz. Under these conditions, it took about 24 hours of data collection to discover the 0.53 eV resonance in ^{250}Cf, using a 150 ng sample.

II. A PROTON-DRIVEN SLOWING-DOWN SPECTROMETER

When we first proposed adding a Proton Storage Ring (PSR) to the Weapons Neutron Research (WNR) facility at the Los Alamos National Laboratory, we recognized that a lead slowing-down spectrometer driven by bursts of 800 MeV protons would have some unique features. A proton spallation target is much more

efficient at producing neutrons than is an electron bremsstrahlung target: we estimate 10-15 neutrons produced per 800 MeV proton on a tungsten target, compared with 0.01-0.02 neutrons for each 60 MeV electron on target. So if the performance of the lead SDS is limited by the problem of average heat removal from the target, we expect a factor of 50-100 higher average neutron population in a proton-driven spectrometer. However, if the problem is primarily due to gamma heating of the lead, the improvement factor could be much larger than this estimate because the gamma pulse is very much smaller with a proton-spallation source. We also expect a dramatic improvement in the signal-to-noise ratio with a proton-driven SDS at WNR/PSR. The proton storage ring, currently used to provide neutrons for the Los Alamos Neutron Scattering Center (LANSCE) at Los Alamos, typically delivers 50-60 μA of 800 MeV protons in 250 ns wide pulses at 20 Hz.

In summary, based on the foregoing performance calculations, we might expect the average performance of a proton-driven lead slowing-down spectrometer at WNR/PSR to show an enhancement of a factor of 50-100 in flux, compared with RINS, at the same average power on target. The signal-to-noise ratio, if the noise is dominated by sample activity such as spontaneous fission, alpha pileup, or gamma radiation associated with sample decay for a capture measurement, should show an enhancement of a factor of about 10^4 over that at RINS. If this conclusion is correct, it suggests that we could consider making fission cross-section measurements on samples that are less than 1/10000 as large as those currently being done (~1 μg) or on samples with more than 10000 times the specific activity.

III. AN EXPERIMENTAL TEST

In November 1989, we fielded a hemispherical fission chamber with microgram sized deposits of ^{235}U, ^{247}Cm, ^{250}Cf, and ^{254}Es for a measurement in the RINS slowing-down spectrometer.[8] Eight months later (almost exactly one half-life for ^{254}Es) we fielded the same chamber in a measurement in conventional geometry at the LANSCE facility. The two experimental configurations are compared in Table II. If the calculations and the conclusions reached in the previous sections are correct (that a PSR-driven SDS should have 10^4 times the flux of RINS), and if the experience at RPI summarized in Table I obtains for PSR/WNR (that the SDS flux is about 10^4 times that of a conventional experiment), then in our conventional experiment at LANSCE we should see the same count rate per burst as we saw with the RINS spectrometer, if we correct to the equivalent flight path.

We choose the 1.25 eV resonance in (^{247}Cm +n) to make the comparison; it is very nearly resolved in the RINS measurement, and thus allows a reasonable comparison to be made. The raw count rate comparisons are given in Figs. 1 and 2. From these data, it is evident that the raw count rates per burst are nearly the same, 0.21 c/burst in this resonance with RINS and 0.29 c/burst at LANSCE. If one corrects to the equivalent flight path, by multiplying the resonance area observed at LANSCE by $(7.25/5.65)^2 = 1.65$, one can conclude that, at a 5.65 m station at LANSCE, the neutron intensity should be roughly a factor of 2 larger than with RINS.

The two sets of data were analyzed in the same way: the ^{235}U fission count rate, measured at the same time in the experiment, was used to derive a flux shape and magnitude. This then permits the fission cross sections for the ^{247}Cm, ^{250}Cf, and ^{254}Es samples to be determined. The cross section comparisons are shown in Figs. 3, 4, and 5 for ^{247}Cm, ^{254}Es, and ^{250}Cf, respectively. Resonance parameters from area analysis are compared for ^{247}Cm and ^{250}Cf in Table III. For ^{254}Es, no distinct resonances were seen in either measurement.

Table II. Comparison of the November 1989 RINS experimental determination of the fission cross section of ^{247}Cm, ^{250}Cf, and ^{254}Es with the July 1990 LANSCE determination.

	RINS	LANSCE
Geometry	Pb SDS	Conventional
Flight path	5.65 m[a]	7.25 m
Beam	60 MeV e^-	800 MeV H^+
Target	He-cooled Ta	H_2O-cooled W
Pulse width	200 ns	125 ns FWHM
Repetition rate	90 Hz	20 Hz
Average power	0.9 kW	55 kW
Duration of expt.	19 hours[b]	96 hours
	(6.2 • 10^6 bursts)	(5.9 • 10^6 bursts)
Sample sizes:		
^{247}Cm	3.16 μg	3.16 μg
	(30.86 f/s)	(30.6 f/s)
^{250}Cf	0.149 μg	0.139 μg
	(441.9 f/s)	(412.6 f/s)
^{254}Es	0.237 μg[c]	0.117 μg
	(13.03 f/s)[d]	(338.3 f/s)[d]

[a]5.65 m is the equivalent flight path for a solid lead SDS.
[b]19 hours is a partial run, used for the present comparison.
[c]A recalculation of the sample size in November 1989, based on the observed spontaneous fission rate in July 1990 for a 276 day ^{254}Es half-life, gives 0.237 μg instead of the 0.21 μg of Ref. 8).
[d]The increase from 13 to 338 fissions per second is due to the ^{250}Cf content.

The resolution associated with the LANSCE measurement is much better than that with RINS, although not as good as that associated with the 1969 Physics-8 nuclear explosion measurement of Moore and Keyworth.[9] The resolution of our LANSCE measurement should be sufficient to give a reasonably complete set of parameters below 20 eV, however, where no data were obtained on Physics-8 because the moderator was moving up the line-of-sight pipe faster than the escaping neutrons.

A surprising feature of the present comparison of the two facilities and the data sets we obtained is that the low resolution lead SDS gives a reasonable, although not complete, picture of the average resonance properties of fissionable nuclei. In the LANSCE measurement, we confirmed the parameters of the lowest energy resonance in ^{250}Cf. We confirmed a net average cross section of ~4 barns between 16 and 80 eV in ^{250}Cf, but saw no additional structure. We confirmed the average cross section of ^{254}Es, and seem to see the same little wiggles, but no distinct resonances. In this sense, we confirmed the conjecture we made earlier[8] that the fission cross section of ^{254}Es has no resolvable resonances. However, for ^{247}Cm we observed many more resonances in the LANSCE measurement, enough to be able to extract the level density below 20 eV and to complete the multilevel analysis[9] in this region.

In conclusion, we feel that the present two experiments provide a sufficient data base to enable us to project what could be done with a lead slowing-down

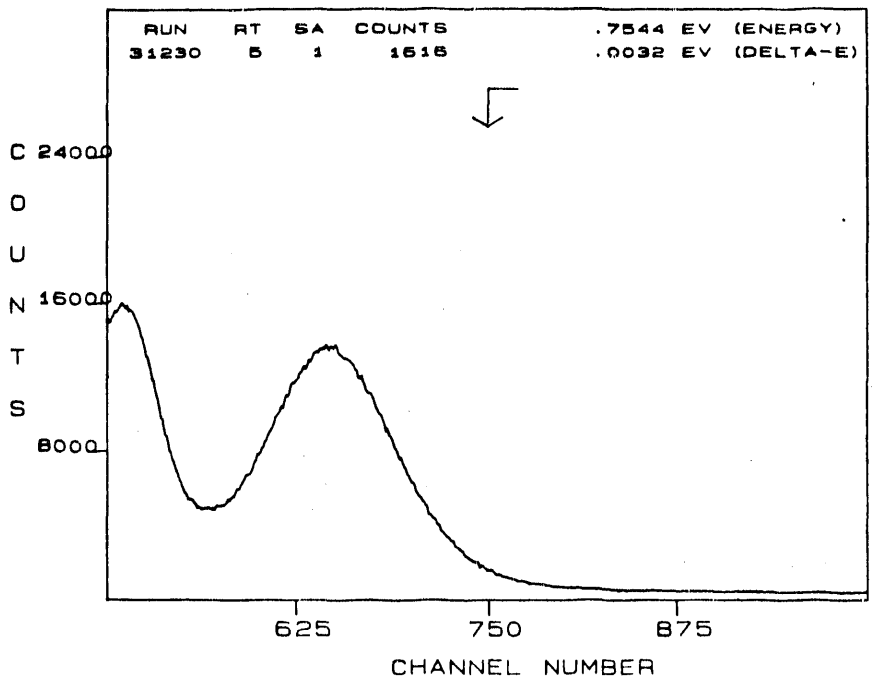

Fig. 1. Raw data plot over the 1.25 eV resonance in (^{247}Cm + n) as obtained with the RINS on 2-3 November 1989. This figure represents a summation of 19 one-hour runs (runs 312-330) at 90 Hz, or 6.2 • 10^6 bursts. The integral over this resonance is about 1.3 • 10^6 counts, giving 0.21 count/burst.

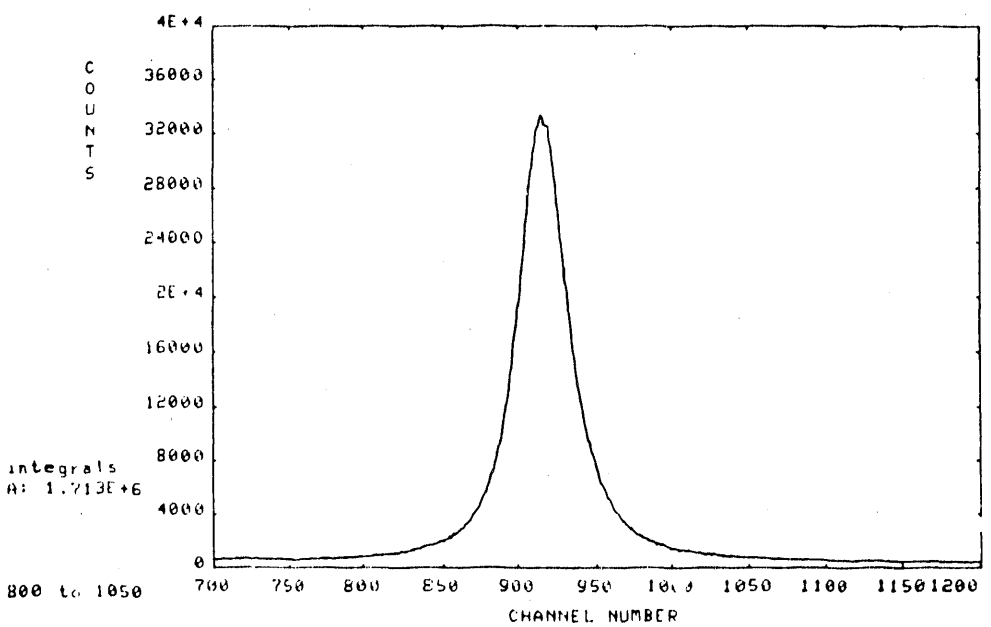

Fig. 2. Raw data plot over the 1.25 eV resonance in (^{247}Cm + n) as obtained in the conventional time-of-flight measurement at a 7.25 m flight path at LANSCE on 26-30 July 1990. This figure represents a summation of four 24-hour runs taken at 20 Hz, or 5.9 • 10^6 bursts. The integral over this resonance is about 1.7 • 10^6 counts, giving 0.29 count/burst.

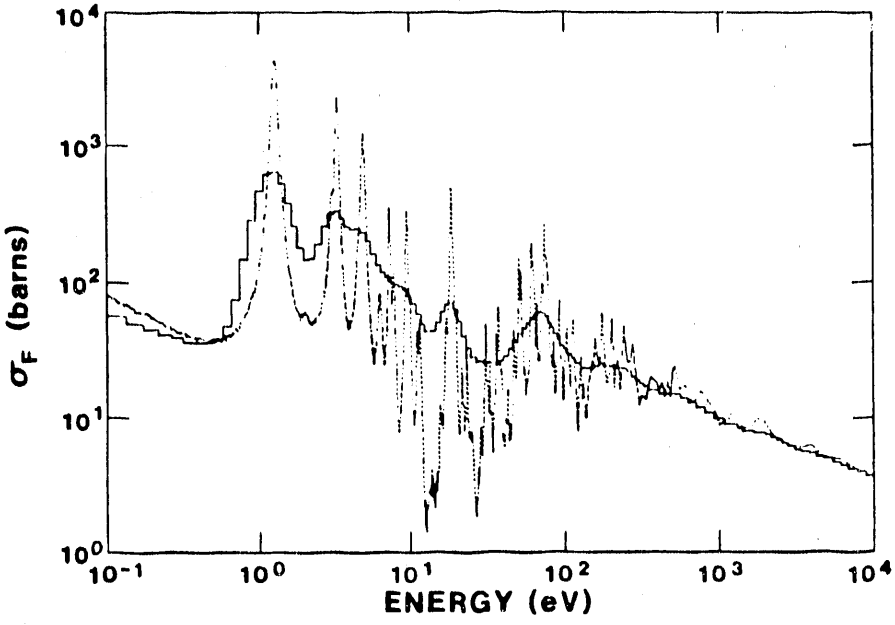

Fig. 3. Fission cross section of (^{247}Cm + n) as determined with the RINS, shown as the solid line, and in conventional geometry at LANSCE, shown as the dotted line, from 0.1 eV to 10 keV.

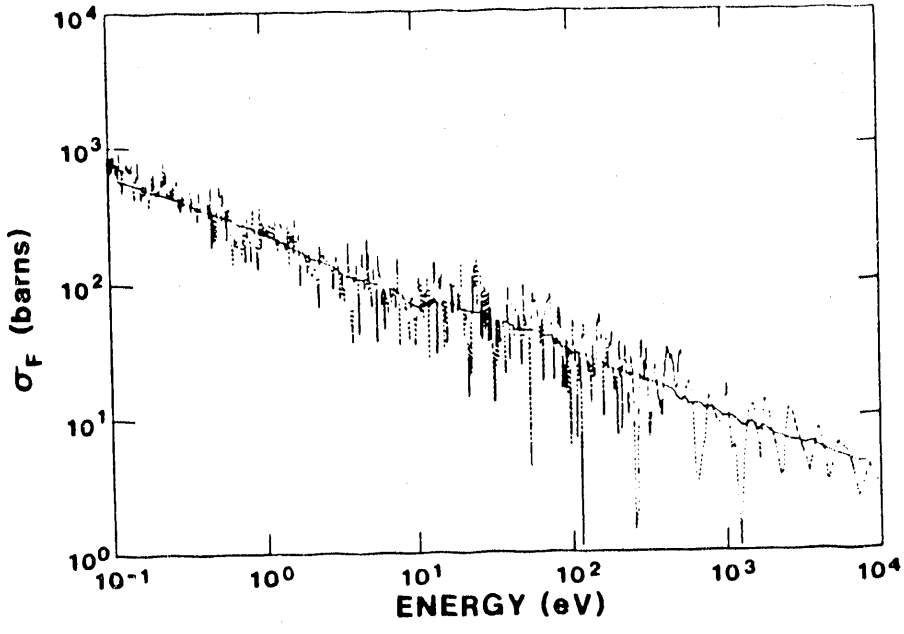

Fig. 4. Fission cross section of (^{254}Es + n) as determined with the RINS, shown as the solid line, and in conventional geometry at LANSCE, shown as the dotted line, from 0.1 eV to 10 keV. The RINS results have been scaled by 0.89, in accordance with a more recent determination of sample size as discussed in the text and in Table I.

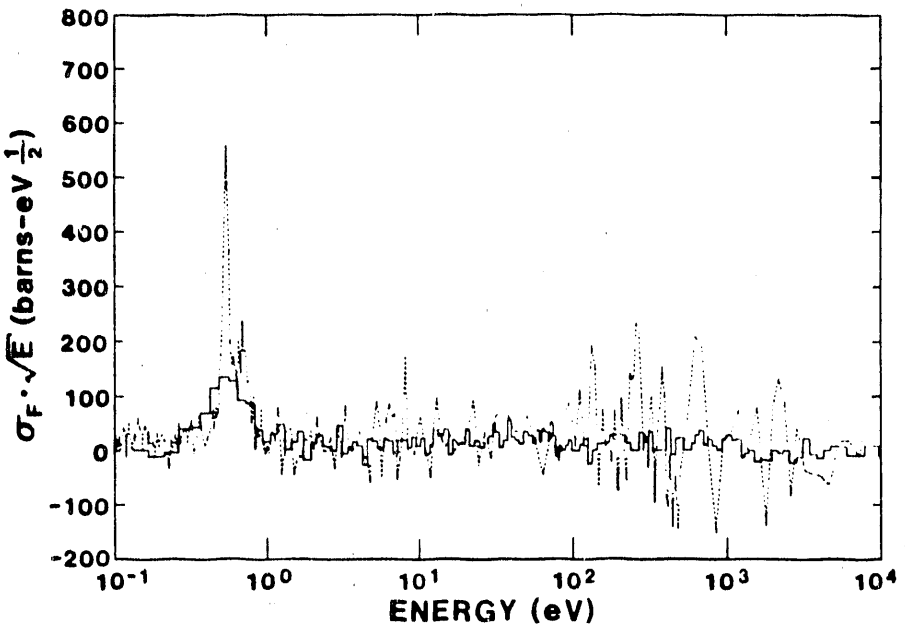

Fig. 5. Fission cross section of (^{250}Cf + n) as determined with the RINS, shown as the solid line, and in conventional geometry at LANSCE, shown as the dotted line, from 0.1 eV to 10 keV. The appearance of the 0.53 eV resonance as a possible doublet in the LANSCE results is considered to be spurious.

Table III Comparison of resonance parameters for ^{250}Cf and ^{247}Cm, as determined in the two experiments.

E_o	A_f (RINS) (b-eV)	A_f (LANSCE) (b-eV)
0.53 (^{250}Cf)	71 ± 9	80 ± 5
1.246	427 ± 34	636
2.97 + 3.18	444 ± 70	579
9.56 + 9.92	141 ± 70	182
18.24 multiplet	332 ± 46	463

spectrometer at WNR driven by 800 MeV proton pulses from the PSR. For each pulse, we expect 2 • 10^4 times as many neutrons in the SDS as are provided by RINS. How many fissionable isotopes are there whose average properties are of interest but which can be produced only in very limited (i.e., ng) quantities (2 • 10^{12} atoms)? There are perhaps 30 transactinium isotopes that have not been measured, which could be given consideration. Let us choose one of these, ^{235m}U, as an illustrative example.

There have been three recent measurements of the thermal and subthermal fission cross sections of 26 min ^{235m}U.[10-12] This 1/2+ isomer is produced in the alpha decay of ^{239}Pu. In the measurements of d'Eer et al.,[12] samples of 8 • 10^9

atoms were prepared by collecting recoil atoms from ^{239}Pu decay in a ^{235m}U generator, a 0.5 m D Al sphere coated on the inside with PuF_4. Mostovoi and Ustroyev[10] prepared somewhat larger samples ($2 \cdot 10^{10}$ atoms) in the same way--their cylindrical generator was larger. In the measurement by Talbert et al.,[11] the samples were still larger ($2 \cdot 10^{11}$ atoms) and were prepared by consecutive radiochemical separation of Pu and U.

All three measurements gave roughly the same result: the thermal (2200 m/sec) cross section of ^{235}U and ^{235m}U are about equal. However, this does not answer the question of primary interest: how do the reaction rates compare for fast neutrons? This question can be answered by determining average resonance parameters for ^{235m}U. A sample of $2 \cdot 10^{11}$ atoms is large enough to permit a measurement on a lead SDS at WNR/PSR: if the resonance fission cross section is comparable to that of ^{235}U, we should expect to observe 0.12 ^{235m}U counts/pulse with a fresh sample or 3700 counts between 1 eV and 80 keV for each sample fielded, if the SDS can be driven at 20 Hz. Here the estimate is made by assuming that event addresses are stored with a real-time clock address included and that data are taken for up to ten half lives; this permits one to use the sample until it is completely decayed, weighting each event as being due to ^{235}U or ^{235m}U fission by Bayes' equation.

This experiment can be considered as an effective lower limit on sample size and half-life for the SDS at WNR/PSR. While 3700 events are not an overwhelmingly large statistical sample for the determination of average properties, we feel that it would be adequate, noting that d'Eer at al[12] analyzed 1402 events in their measurement with thermal neutrons.

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