TOWARDS BENCHMARK MEASUREMENTS FOR USED NUCLEAR FUEL ASSAY USING A LEAD SLOWING-DOWN SPECTROMETER

B. Becker[1], N. Thompson[1], J. Thompson[1], D. Beller[2], S. Bowyer[3], A. Gavron[4], G. Imel[5] and Y. Danon[1]

[1]*Rensselaer Polytechnic Institute, Troy, NY 12180*
[2]*University of Nevada, Las Vegas, Las Vegas, NV 89054*
[3]*Pacific Northwest National Laboratory, Richland, WA 99352*
[4]*Los Alamos National Laboratory, Los Alamos, NM 87545*
[5]*Idaho State University, Pocatello, ID 83204*

The use of a Lead Slowing-Down Spectrometer (LSDS) is considered as a possible option for non-destructive assay of fissile material in used nuclear fuel. The primary objective is to quantify the ^{239}Pu and ^{235}U fissile content via a direct measurement distinguishing them through their characteristic fission spectra in the LSDS. Threshold fission chambers are employed to detect fission neutrons due to their insensitivity to lower energy interrogation neutrons and their favorable behavior in an irradiating environment.

In the framework of the LSDS Collaboration for the Material Protection and Control Technology (MPACT) Campaign we performed several assay experiments at the Rensselaer Polytechnic Institute (RPI) to demonstrate the feasibility of such method and to provide benchmark experiments. A fresh UO_X fuel rod from the RPI Criticality Research Facility and a PuBe source were assayed consecutively in the LSDS. The characteristic fission spectra were measured with ^{238}U and ^{232}Th threshold fission chambers. Despite the constant neutron and gamma background from the PuBe source, the LSDS system was able to measure a ^{239}Pu response. This is the first time that Pu was assayed in the US using a LSDS system. Both measurements were found to be in good agreement with calculations done with MCNP5. Experiments with a combination of a fuel pin and the PuBe source were performed to simulate to some extent burn-up conditions. The distance of the PuBe source with respect to the detectors was varied to simulate different Pu contents and several mixed ^{239}Pu and ^{235}U response spectra were measured and compared to calculations.

I. INTRODUCTION

Lead Slowing-Down Spectrometers (LSDS) have been researched for non-destructive assay of nuclear fuel pins to analyze their fissile content. Interatom and the Karlsruhe Nuclear Research Center, Germany designed and operated a LSDS to assayed fresh mixed oxide (MOX) fuel pins for fabrication control (Refs. 1,2). In the US, the LSDS of Rensselaer Polytechnic Institute (RPI) was used in the past to assay fresh ^{233}U and ^{235}U enriched fuel pins (Refs. 3,4).

Due to its ability to assay non-destructively, the LSDS is well suited not only for fresh fuel assay but also for spent fuel testing. The characterization of spent fuel is particularly important for nuclear safeguards and for determining the fuel burn up level in view of reprocessing and recycling of used fuel. Several studies have been made to assess the potential of the LSDS for spent fuel assay (Refs. 1, 4-9).

In the framework of the Office of Nuclear Energy, Fuel Cycle R&D, Material Protection and Control Technology (MPACT) Campaign the LSDS is investigated among other methods as an option for the accurate, direct, and independent assay of the fissile isotopes.

As part of the LSDS Collaboration for the MPACT Campaign we assayed a fresh fuel pin from the Rensselaer Critical Facility (RCF) and a PuBe neutron source in order to demonstrate the feasibility of the LSDS assay method and as a first step towards benchmark measurements. In this paper, a brief overview of the preformed experiments is given. Experimental results using a ^{238}U assay detector are presented.

Recent design studies of the LSDS system (e.g. Refs. 5-7, 9) are mainly based on Monte Carlo simulations. Therefore, the experimental results are compared to Monte Carlo calculations of the assay in order to investigate the validity of such Monte Carlo simulations.

II. EXPERIMENTAL CONDITIONS

In the following discussion a summary of the basic components of the RPI LSDS fuel assay system is given. Detailed information on the system can be found in (Refs. 3, 10).

II.A. RPI - LSDS + LINAC

The RPI LSDS consist of a cubic lead pile (1.8 m side length) with a total weight of 72 tons (figure 1). The RPI linear accelerator (LINAC) directs 40-60 MeV electrons on an air cooled tantalum target located in the center of the cube. Neutrons are produced via a (e,γ) and a (γ,n) reaction within the target. The energy distribution of

the neutrons is given approximately by an evaporation spectrum with a peak energy of about 0.46 MeV. The created neutrons loose energy in successive collisions with the lead isotopes as time progresses. The neutron energy-time correlation is given by (Ref. 3):

$$E = \frac{k}{(t + t_0)^2}$$

where k ≈ 165000 eV/ μs^2 and t_0 ≈ 0.3 μs. E is the average neutron energy (in eV) and t is the slowing down time (in μs). This direct energy-time correlation is valid shortly after the source pulse until neutrons are completely slowed down. During the slowing down process the neutron energy is approximately Gaussian distributed with an energy dependent resolution [full width at half maximum (FWHM)] given by:

$$\left[\frac{dE}{E}\right]_{FWHM} = \left[0.0835 + \left(\frac{0.128}{E}\right) + 3.05\ 10^{-5}\ E\right]^{1/2}$$

where E is the neutron energy in eV. This energy resolution increases significantly with the introduction of light nuclei impurities such as hydrogen. Therefore, the RPI LSDS was build with high purity lead (Ref. 3).

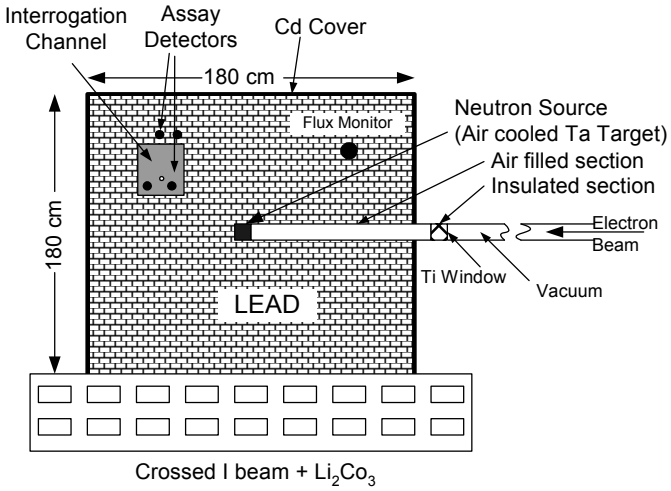

Fig. 1: RPI Lead Slowing-Down Spectrometer

When fissile material like a fuel pin is introduced into the LSDS fission neutrons are created. Since these fission neutrons have a significantly higher energy than the interrogating neutrons, they can easily be distinguished from the interrogation neutron flux by using a fast neutron detector such as a threshold fission chamber. Each fissile isotope gives, depending on its resolution broadened fission cross section a characteristic fission response as function of time.

II.B. Assay Detectors and Flux Monitor

In the performed experiment, two ^{238}U and two ^{232}Th threshold fission chambers were used to detect fission neutrons. Fission chambers are essentially insensitive to gamma radiation. They are therefore particularly appropriate for fuel assay in a LSDS in order to overcome the expected high gamma background caused by the spent fuel and the target Bremsstrahlung.

The assay detectors contain each about 200 mg of ^{238}U or ^{232}Th with an active length of about 20 cm. In case of the ^{238}U detectors, highly depleted ^{238}U is used (residual ^{235}U content ≈ 4.1 ppm (Ref. 3)). Such high grade is necessary to lower the detector sensitivity to the interrogation neutrons. The ^{232}Th detectors have a significantly smaller fissile content which improves the signal to background ratio. However, due to the lower fission cross section the efficiency of the ^{232}Th assay detectors is about 1/3 of the ^{238}U.

The neutron flux is monitored with a ^{235}U probe chamber located at the front upper corner of the LSDS (see figure 1). This monitor was used to normalize assay detector signals to the neutron flux in the LSDS.

II.C. Assay Specimen

In the presented experiment two different assay specimens were used: a fuel pin from the RCF reactor and PuBe source.

The low enriched UO_2 fuel pin from the SPERT (Special Power Excursion Reactor Test) program contains about 35.2 g ^{235}U. Since the RCF operates near zero power conditions the burn up of the fuel pin is negligible. The PuBe source serves two different purposes. Firstly, an assay of ^{239}Pu can be performed. Secondly, the sensitivity of the assay detectors to a constant neutron and gamma background can be investigated. The PuBe source contains about 96 g of ^{239}Pu and has an activity of about ~22 10^{10} Bq (~1.1 10^7 n/s).

II.D. Measurement Arrangement

An interrogation channel of the LSDS was used to introduce the assay specimen. One end of the channel was closed permanently with several lead bricks and covered with cadmium and boron plates. The remaining open end was partially covered with a brick and a cadmium plate. During the measurement the fuel pin was supported by fuel grid plates within an assembly box. ^{238}U and ^{232}Th assay detectors were placed directly into the fuel assembly box (figure 2). An additional ^{238}U and a ^{232}Th assay detector were introduced into the lead 2.54 cm above the interrogation channel. All detectors were located at mid depth of the interrogation channel. A guide tube is used for the insertion of the PuBe source.

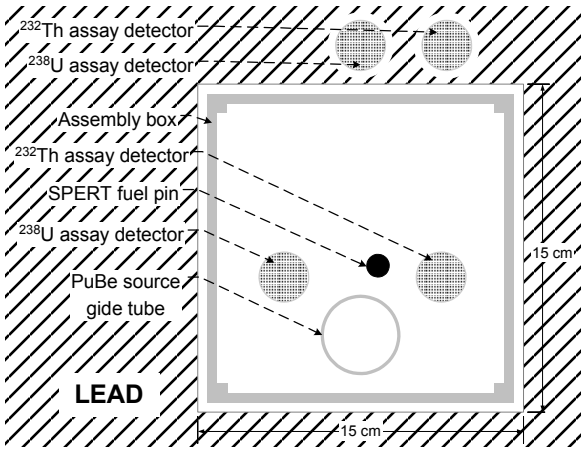

Fig. 2: Fuel pin, PuBe source and detector arrangement

III. SIMULATION OF DETECTOR RESPONSES

The Monte Carlo Code MCNP5 (Ref. 11) was used to simulate the fuel assay. The entire lead cube, assembly box, assay specimen and detectors were modeled. All neutrons escaping the cube were considered to be lost neglecting the room return. Unless otherwise stated the impurities in the lead were representatively modeled by assuming an H_2 content of 2 wt. ppm (see section IV.B). All cross sections are based on ENDF/B VII (Ref. 12) with exception of the cross section of the ^{238}U content of the detectors which is based on JEFF 3.1 (Ref. 13).

Several variance reduction methods were applied in order to decrease the computational time, namely importance zoning and time dependent weight windows. A typical computational time for one configuration was found to be several hours (7-10 h) on a 12 cpu windows cluster (~3GHz i7 processor).

IV. RESULTS

During all measurements the LINAC was operated with an electron current and energy range of about 14-15 μA, energy of 40-53 MeV, pulse width of 1 μs and repetition rate of 180 Hz, respectively. All measurements are normalized using a neutron flux monitor to 14.5 μA and 46 MeV conditions. In the following the responses of the ^{238}U assay detector located inside the fuel assembly box are presented. All shown experimental uncertainties correspond to the propagated uncertainty due to counting statistic.

IV.A. Detector Signals

The assay detector signal was measured without assay specimen in order to determine the response of the detector to the interrogation neutron flux. In addition, the detector efficiency can be determined using the known ^{235}U impurity content. The detector signal exhibits a distinct fission peak at about 10 to 20 μs (figure 3) which is caused by the ^{238}U subthreshold fission resonances in the 0.4 to 5 keV energy region. The response above 20 μs is mainly caused by the fissile ^{235}U content with the exception of a second ^{238}U subthreshold fission peak at about 90 μs.

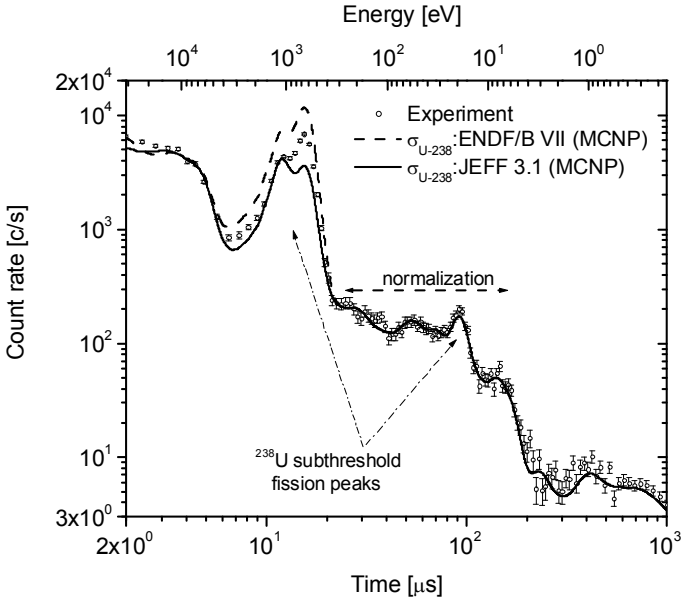

Fig. 3: ^{238}U assay detector signal caused by interrogation neutrons. MCNP calculations are using the ENDF VII or the JEFF 3.1 cross section library for ^{238}U.

The measurement was compared to MCNP simulations using ENDF/B VII or JEFF 3.1 cross sections for ^{238}U. While JEFF 3.1 under predicts the subthreshold fission peak (10 to 20 μs), ENDF/B VII over predicts the peak. A similar spectrum was measured by Slovacek et. al. (Ref. 14) using the same detector and by Danon et. al. (Ref. 15) using a different ^{238}U detector.

The MCNP calculations were normalized to the experimental data in the 20 - 200 μs time frame applying a least square method. It should be noted that there is a good agreement between simulation and experiment in the ^{238}U fission dominated 2-5 μs time frame as well as in the ^{235}U fission dominated 200-1000 μs time frame.

In general the MCNP calculations need to be normalized for comparison with obtained experimental result. As a first approach, we use the normalization factor obtained in this section for the following assay measurements which directly takes into account the detector efficiency and the neutron source strength.

IV.B. Fresh Fuel Pin Assay

A SPERT fuel pin was inserted into the assay channel and the detector response was measured (figure 4). Using

the normalization of the previous section an absolute count rate prediction is possible using MCNP. Since the resolution of the LSDS is dependent on the amount of light element impurities in the lead three different cases were considered with a representative H_2 contamination of 0, 2 and 4 wt. ppm. With increasing impurity content, the detector response signal tends to lose structure which is particularly visible at about 300 μs. A level of 2 wt. ppm of hydrogen is found to give the best agreement to the experimental response (c.f. Ref. 10). With exception of the previously mentioned sub threshold fission peak, the absolute detector response is in excellent agreement with the calculation.

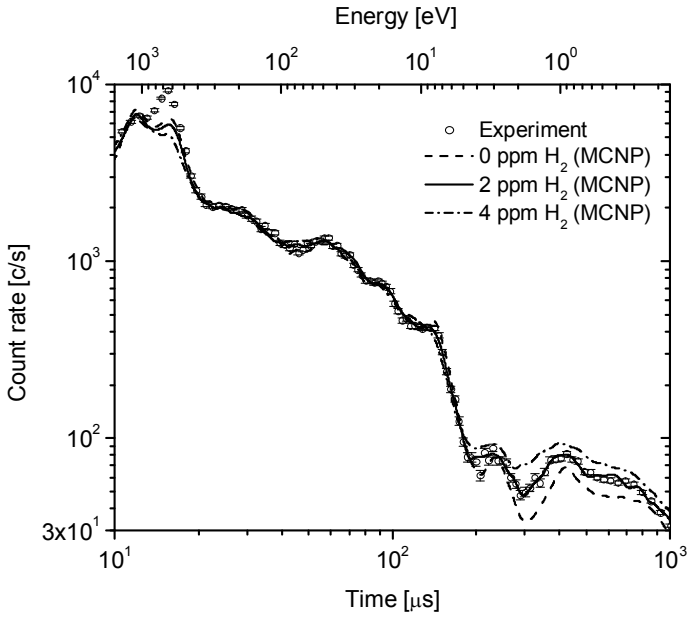

Fig. 4: ^{238}U assay detector response function of a fresh fuel pin assay. MCNP calculations are done with different H_2 impurity contents.

IV.C. ^{239}Pu Assay

The PuBe source was positioned at mid depths of the LSDS interrogation channel. Figure 5 shows the measured assay signal in comparison to the signal of a ^{239}Pu probe chamber. Due to strong self-shielding of the lumpy PuBe source the assay signal exhibits a decrease of the fission peaks at about 50, 130 and 800 μs. In addition, resonance shielding effects of ^{181}Ta and of ^{240}Pu are noticeable. While ^{181}Ta is present in the cladding of the PuBe source, small amounts of ^{240}Pu can be found within the PuBe source. Despite the self-shielding effects the absolute response function of the assay is well predicted by calculations.

IV.D. Mixed ^{235}U and ^{239}Pu Assay Signals

A set of mixed ^{235}U and ^{239}Pu signals was measured by inserting both, the PuBe source and the fuel pin into the LSDS. The contribution of the ^{239}Pu signal to the detector response was varied by changing the insertion depth of the source i.e. by changing the distance of PuBe source to detectors. The mixed signals were compared to response signals of only the fuel pin or the PuBe source (Figure 6). The mixed detector signals exhibit characteristics of both, the ^{235}U and ^{239}Pu response functions. Even when the source is at a distance of 30 cm from the detector the mixed signal can clearly be distinguished from the clean ^{235}U response function.

The ^{239}Pu self-shielding within a used fuel pin is expected to be significantly smaller than in the presented case of the PuBe source which should improve the distinctness of the contribution of ^{239}Pu and ^{235}U fission to the combined signal.

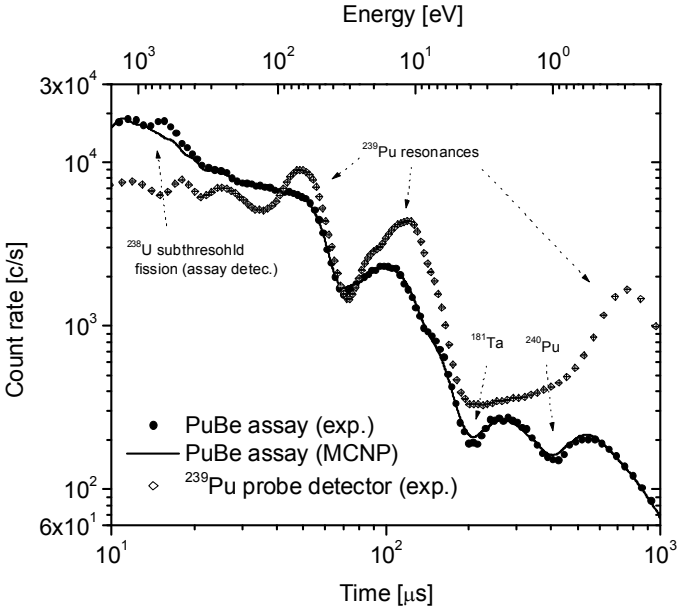

Fig. 5: ^{238}U assay detector response function of a PuBe source assay and characteristic ^{239}Pu probe chamber response (^{239}Pu probe chamber signal is scaled).

V. CONCLUSION

In the presented study a fresh fuel pin, a PuBe source and a combination of both were assayed in the RPI LSDS. Simulations of the different assay cases with MCNP were compared to the experimental results. Experiments and calculations were found to be in very good agreement with exception of a sub threshold fission ^{238}U peak. Using the known ^{235}U content of the assay detector, absolute count rate estimation with MCNP was possible. Even though the PuBe source exhibits strong self-shielding effects and emits a constant neutron and gamma background the calculated detector signal are in agreement with the measurement. The combined measurements of the PuBe source and the fuel pin demonstrate the superposition of the characteristic ^{239}Pu and ^{235}U response spectra.

The general agreement of the measurements and simulations indicate the validity of the Monte Carlo model and shows the potential of such simulations to be included in the benchmarking of de-convoluting algorithms for the fissile mass determination of LSDS assay measurements. In addition, the presented mixed ^{235}U and ^{239}Pu assay can be used as a first test case for such algorithm.

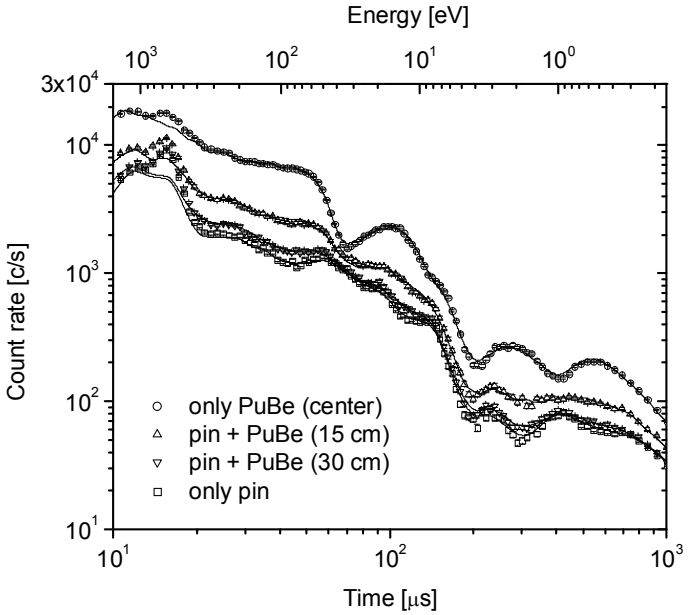

Fig. 6: ^{238}U assay detector response function of a fuel pin assay, of a PuBe source assay and an assay of both, fuel pin and PuBe source (at different positions). Solid lines are MCNP calculations.

ACKNOWLEDGMENTS

The authors sincerely thank the technical staff at RPI LINAC (Peter Brand, Matt Gray, Martin Strock and Azzedine Kerdoun) for their efforts in operating and maintaining the LINAC and LSDS and assistance with setting up experiments.

REFERENCES

1. H. Krinninger, S. Wiesner, C. Faber, "Pulsed Neutron Method for Non-Destructive and Simultaneous Determination of the ^{235}U and ^{239}Pu Content of Irradiated and Non-Irradiated Reactor Fuel Elements", Nucl. Instr. Meth. 73 (1969), p. 13.
2. H. Krinninger, E. Ruppert, H. Siefkes, "Operational Experience with the Automatic Lead-Spectrometer Facility for Nuclear Safeguards", Nucl. Instr. Meth. 117 (1974) 61.
3. D.S. Cramer, R.E. Slovacek, E.B. Bean, R.G. Luce, Report KAPL-M-7449 (1976).
4. N. M. Abdurrahman, R.C. Block, D.R. Harris, R.E. Slovacek, Y.D. Lee, R. Rodriquez-Vera, "Spent-Fuel Assay Performance and Monte Carlo Analysis of the Rensselaer Slowing-Down-Time Spectrometer," Nucl. Sci. Eng. 115, 279 (1993).
5. A. Gavron, L.E. Smith, J.J. Ressler, "Analysis of Spent Fuel Assemblies Using a Lead Slowing Down Spectrometer Nuclear," Nucl. Instr. Meth. A 602, (2009).
6. Y. D. Lee, N.M. Abdurrahman, R.C. Block, D.R. Harris, R.E. Slovacek, "Design of a Spent-Fuel Assay Device Using a Lead Spectrometer," Nucl. Sci. Eng. 131, 45 (1999).
7. L.E. Smith, K.K. Anderson, J.J. Ressler, S.D. Kiff, M.W. Shaver, "Time-Spectral Analysis Algorithms for Lead Slowing-Down Spectroscopy of Spent Fuel," 2009 Institute of Nuclear Materials Management Conference, Tucson, AZ (2009).
8. E.C. Vanterpool, R.E. Slovacek, D.R. Harris, R.C. Block, "Fuel Assembly Assay by Neutron Interrogation in a Lead Slowing-Down-Time Spectrometer," Nucl. Sci. Eng. 110, 186 (1992).
9. C. Romano, Y. Danon, D. Beller, "Fuel Assembly Self Shielding of Interrogation Neutrons in a Lead Slowing-Down Spectrometer," Sixth ANS International Topical Meeting on Nuclear Plant Instrumentation, Control, and Human-Machine Interface Technologies, Knoxville, Tennessee (2009).
10. N. Abdurrahaman, "System Performance and Monte Carlo Analysis of Light Water Reactor Spent Fuel Assay Using Neutron Slowing Down Time Method", PhD thesis (1991), Rensselaer Polytechnic Institute
11. X-5 Monte Carlo Team, "MCNP – A General N-Particle Transport Code, Version 5 – Volume I: Overview and Theory", LA-UR-03-1987, Los Alamos National Laboratory (2003).
12. Chadwick et. al., "ENDF/B-VII.0: Next Generation Evaluated Nuclear Data Library for Nuclear Science and Technology " Nuclear Data Sheets, 107, 2931-3060, (2006).
13. JEFF Report 21, JEFF-3.1 Nuclear Data Library; NEA Data Bank (November 2006).
14. R.E. Slovacek, D.S. Cramer, E.B. Bean, J.R. Valentine, R.W. Hockenbury, R.C. Block, $^{238}U(n,f)$ Measurements Below 100keV, Nucl. Sci. Eng., 62, 455-462 (1977).
15. Danon et al., "Measurements with the high flux lead slowing-down spectrometer at LANL", Nucl. Instr. Meth. B 261, pp. 953–955 (2007)