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Actinide Targets for Neutron Cross Section Measurements

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ABSTRACT

The Advanced Fuel Cycle Initiative (AFCI) and the Generation IV Reactor Initiative have demonstrated a lack of detailed neutron cross-sections for certain "minor" actinides, those other than the most common (^{235}U, ^{238}U, and ^{239}Pu). For some closed-fuel-cycle reactor designs more than 50% of reactivity will, at some point, be derived from “minor” actinides that currently have poorly known or in some cases not measured (n,γ) and (n,f) cross sections. A program of measurements under AFCI has begun to correct this. One of the initial hurdles has been to produce well-characterized, highly isotopically enriched, and chemically pure actinide targets on thin backings. Using a combination of resurrected techniques and new developments, we have made a series of targets including highly enriched ^{239}Pu, ^{240}Pu, and ^{242}Pu. Thus far, we have electrodeposited these actinide targets. In the future, we plan to study reductive distillation to achieve homogeneous, adherent targets on thin metal foils and polymer backings. As we move forward, separated isotopes become scarcer, and safety concerns become greater. The chemical purification and electodeposition techniques will be described.

CONTENTS

FIGURES

TABLES

ACRONYMS

AFCI	Advanced Fuel Cycle initiative
DOE	Department of Energy
Gen-IV	Generation-IV
INL	Idaho National Laboratory
LANL	Los Alamos National Laboratory
LANSCE	Los Alamos Neutron Scattering Center
NE	Nuclear Energy
NERAC	Nuclear Energy Research Advisory Committee
ORNL	Oak Ridge National Laboratory

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1. INTRODUCTION

The United States Department of Energy (DOE), Office of Nuclear Energy (NE) has embarked on a long-term program to significantly advance the science and technology of nuclear energy. Key aspects of the DOE-NE agenda are embodied in the Generation-IV (Gen-IV) advanced nuclear energy systems development program and in the Advanced Fuel Cycle Initiative (AFCI) program. The planned efforts involve near-term and intermediate-term improvements in fuel utilization and recycling in current nuclear power reactor systems as well as the longer-term development of new nuclear energy systems that offer much improved fuel utilization, proliferation resistance and reduced requirements for waste disposal, along with continued advances in operational safety.

One of the most important improvements will be the fundamental nuclear databases, especially the evaluated neutron interaction cross section files that serve as the foundation of all reactor system designs, operating strategies, and fuel cycle engineering activities. In particular, the new concepts for reactors and fuel cycles involve the use of transuranic nuclides that were previously of little interest, and where measured information is limited and in some cases does not exist.

There is a recognized need for improvement of the neutron cross section database for several of the actinides, especially in the case of nuclides (minor actinides) other than ^{235}U, ^{238}U, and ^{239}Pu, which of course already have been extensively studied and characterized. As discussed in the DOE-NE Long Term Nuclear Technology Research and Development Plan, published in June, 2000 by the DOE Nuclear Energy Research Advisory Committee (NERAC), there is a clear recognition in the U.S. community that, "*Advances in reactor concept design can be expected to require additional data for basic nuclear properties, such as neutron and gamma spectral data, microscopic cross sections (and other information). The existing data base is only marginally adequate for present applications and is unlikely to be sufficient for future applications*." A similar consensus exists in the international community[1].

Therefore, the Idaho National Laboratory (INL) and the Los Alamos National Laboratory (LANL) have initiated a program to fabricate actinide isotope targets and measure the (n,γ) and (n,f) cross sections with neutrons in the energy range of 10^{-2} – 10^{6} eV, using the DANCE detector[2] (Detector for Advanced Neutron Capture Experiments) and various fission detectors[3], located on collimated neutron beam lines at the Los Alamos Neutron Scattering Center (LANSCE).

2. EXPERIMENTAL

2.1 Preparation of Reagents and Target Backing Foils

The chemical reagents were all ACS grade and solutions were made using deionized water (18 MΩ). The 2.5 μm and 4 μm titanium foils (99.6% pure), used to support DANCE targets, were purchased from Goodfellow Corporation and were attached to Al rings using Devcon 5 minute epoxy #14250. The Al rings have an O.D. of 1.6" and an I.D. of 1". VYNS, a copolymer of 90% vinyl chloride and 10% vinyl acetate was purchased from The Dow Chemical Co. Solutions of VYNS were made using cyclohexanone, as described by YAFEE[4]. The one-sided, fission foils are 12.5 μm thick stainless steel that is sandwiched between stainless steel rings that are 7" O.D. and 6" I.D.

2.2 Actinide Target Isotopes and Chemical Purification

^{239}Pu, ^{240}Pu and ^{242}Pu was purchased from ORNL, isotope sales department. Each of these Pu isotopes was the highest enrichment available. Table 1 shows the isotopic abundance for each of these enriched isotopes on 03/30/2006. The Pu target material was chemically purified by either the classic anion exchange technique[5] (adsorb Pu (IV) to anion exchange column from 10 M HCL or HNO_3, wash with same acid and remove Pu by reducing acid strength to 0.1 M or reducing the Pu (IV) to Pu (III) with HCL/HI) or use of a TEVA-resin® anion chromatography column. These 2 ml, 100-150 μm particle size, resin columns were purchased from Eichrom Technologies, Inc and the purification technique is described by Horwitz[6].

Table 1. Isotopic content of each of the Pu target materials.

Enriched Isotope	238	239	240	241	242	244
^{239}Pu	0.0345	99.077	0.881	0.002	0.005	0.000
^{240}Pu	0.007	0.735	98.835	0.141	0.282	0.000
^{242}Pu	0.002	0.016	0.025	0.012	99.943	0.002

The purification of the enriched isotopic Pu target material, ~5 mg, using the TEVA column is as follows. The Pu was dissolved in 5 ml of warm 8 M HNO_3/0.01 M HF. We added 0.5 ml of saturated H_3BO_3 to complex the fluoride ion and the solution was evaporated to dryness. The Pu was redissolved in ~2 ml of 2 M HNO_3. This solution was passed through a TEVA-resin column along with one 2 ml rinse of the beaker. The Pu (IV) is strongly adsorbed to the resin while Am (III) and U (VI) does not. The column was washed with 10 ml of 2 M HNO_3. The TEVA column is further washed with 5 ml of 6 M HCl which removes any Th (IV) and displaces the HNO_3. Finally, the Pu is removed from the column with 10 ml of 10.8 M HCL/0.39 M HI, a 9:1 mixture of concentrated HCl and concentrated HI.

An example of the purification of ^{240}Pu (the Pu isotope with the most ^{241}Pu) is shown in Figure 1. It is evident that the ^{241}Am, the dominant gamma-ray emitting impurity, has been reduced to a not detected level, a factor of ~10^5. This chemical purification was performed for each enriched Pu isotope prior to target fabrication.

2.3 Actinide Target Electrodeposition

We used the electroplating technique described by MAYA[7]. Maya used 0.4 M $(NH_4)_2SO_4$ adjusted to pH 2.6 to electroplate U as the hydrated UO_2 onto Ni substrates. The effluent from the TEVA column, containing Pu (III) in HCl/HI was evaporated to dryness several times with concentrated HNO_3 and

redissolved in 2 ml of 1 M HNO_3. We would take 0.25 ml of this solution and very gently evaporate to dryness and then redissolve the Pu in 8 ml of 0.4 M $(NH_4)_2SO_4$ adjusted to pH 2.6. Rapidly, this solution was transferred to the electrodeposition cell, using a Ti foil for the cathode and a Pt wire, bent into a ring at the end, as the anode. The ring was parallel to the Ti cathode and ~5 mm above it. A dc power supply provided constant current of 0.35 A (0.70 A/cm^2) at ~15 V. The cell was rotated relative to the Pt wire during electroplating which lasted for ~80 minutes. At the end of the electroplating, but prior to turning off the voltage, ~ 1 ml of concentrated NH_4OH was added to the electrolyte to minimize redissolution of the Pu hydrated oxide. The electrolyte was poured into a bottle for later recovery of unplated Pu as was a deionized H_2O wash of the electroplating cell. Finally, the electroplated Pu, still in the cell, was washed with ethyl alcohol and the cell was inverted to let the deposited Pu dry. Table 2 shows the results of 4 Pu electrodepositions, one with a lesser amount of Pu and the last 3 are replicate with a electroplating efficiency of ~70% when electroplating ~1 mg/cm^2 of Pu.

Table 2. Pu Electroplating Results.

Date	4/19/2005	4/19/2005	4/20/2005	4/21/2005
Electrolyte concentration	0.4 M $(NH_4)_2SO_4$	0.4 M $(NH_4)_2SO_4$	0.4 M $(NH_4)_2SO_4$	0.4 M $(NH_4)_2SO_4$
Acidity (pH)	2.6	2.6	2.6	2.6
Mass Pu added to cell (μg)	46.2	577	577	577
Time (min)	60	79	82	73
Current (A)	0.35	0.35	0.35	0.35
Voltage (V)	15.4	15.4	~17	15.7
Mass Pu Electroplated (μg)	40.9	423	389	400
Percent Pu Electroplated	87	73	67	69
Color of Electroplated Pu	Green	Green	Green	Green

2.4 Actinide Target Assay

All of the Pu targets have been assayed using a carbon window HPGe detector that has been efficiency calibrated to 2-3% from 40 KeV to ~800 KeV. The initial chemical purification and electroplating studies, including Figure 1, were analyzed using a standard Al window HPGE detector. Since the ^{240}Pu and ^{242}Pu targets are rather thick and the emitted gamma-rays are low in energy, the gamma-ray attenuation in each source was modeled and found to be ~1%. We have combined, in quadrature, the detector efficiency uncertainty (3%), the source attenuation uncertainty (1%) along with the largest emission probability uncertainty[8] for the gamma-rays used in the quantification (1.7% for ^{240}Pu and 3.9% for ^{242}Pu), giving a total uncertainty of 3.7% for ^{240}Pu target masses and 5.0% for the ^{242}Pu target masses.

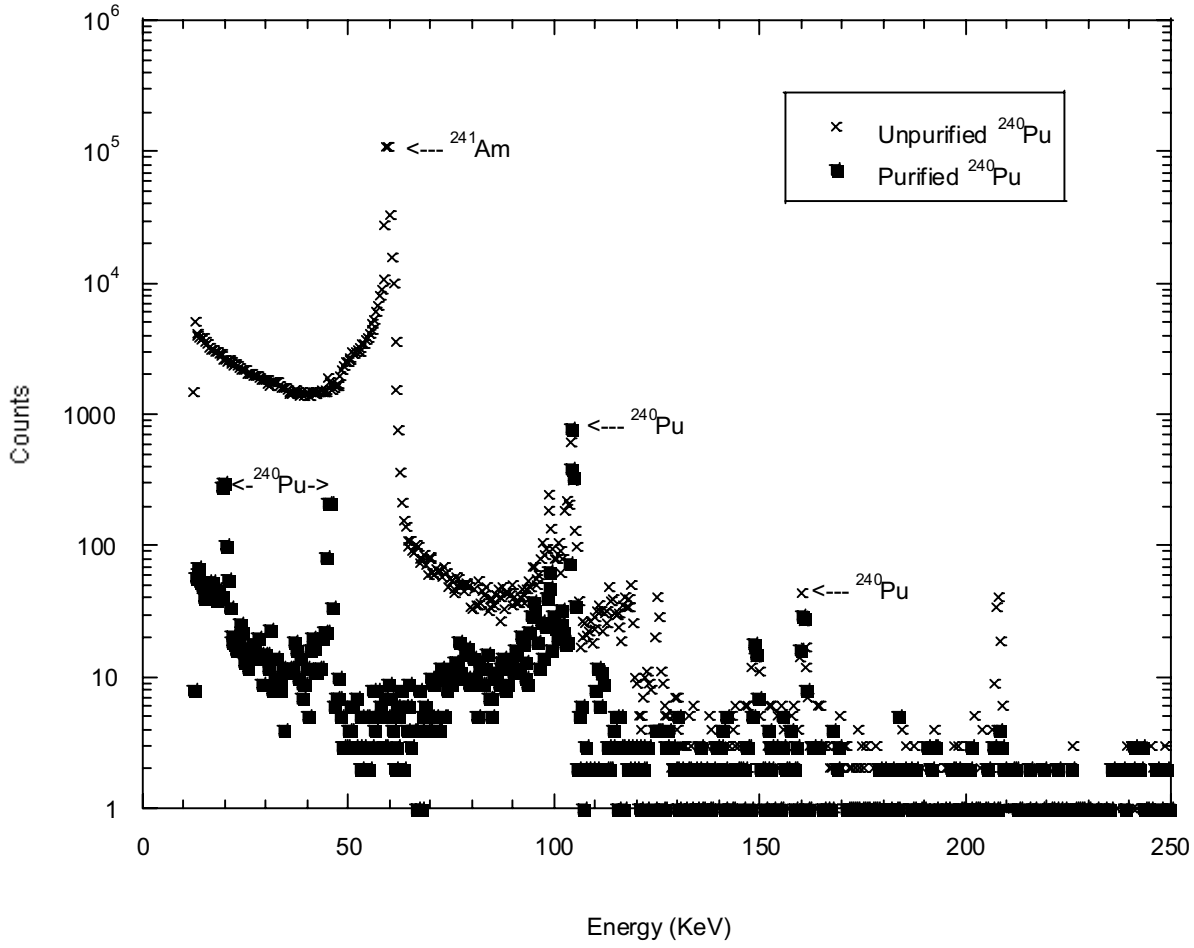

Figure 1. Gamma ray spectra of ^{240}Pu before and after chemical purification.

3. RESULTS AND DISCUSSION

The DANCE neutron capture targets are typically ~1 mg in mass and 8 mm in diameter. The diameter was chosen such that the entire actinide deposit would be within the 10 mm diameter neutron beam. To date, these targets have been electroplated onto 2 μm and 4 μm Ti foils. All of the targets consisted of 2 Ti foils glued together with the actinide deposit sandwiched between the foils. We did not completely glue around the circumference so as to leave a path for air to escape during evacuation in the DANCE detector. In some cases, each of the foils contained electrodeposited target material in order to increase the total mass of target material.

Figure 2 shows the picture of an uncovered ^{240}Pu electroplated DANCE target (887 ± 33 μg). Figure 3 shows an assembled ^{240}Pu DANCE target with the sandwiched foils centrally glued inside of an Al cylinder having polyimide windows on either end which acts as secondary containment while inside of the DANCE detector. This outer container is called an RTH (radioactive target holder).

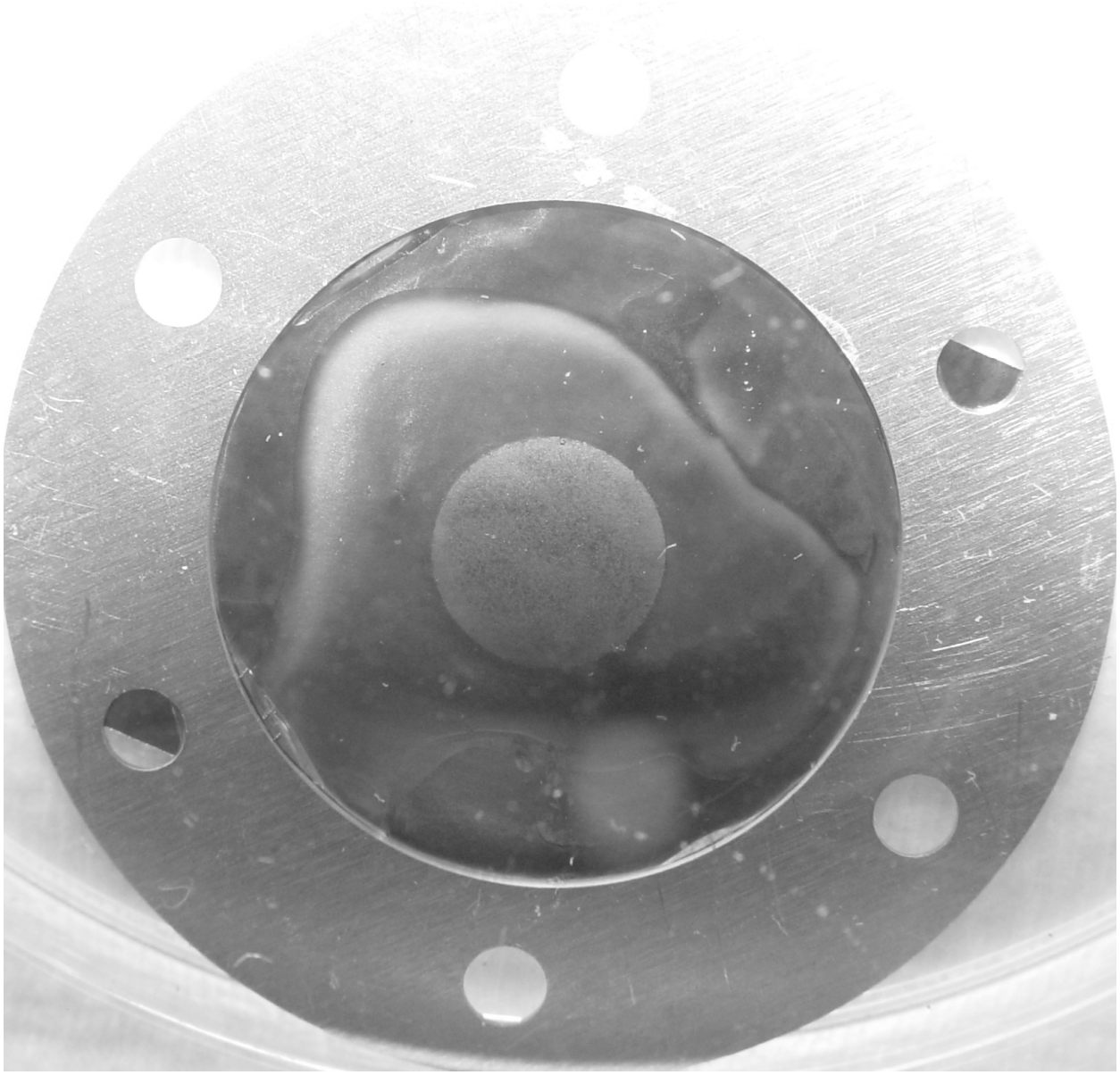

Figure 2. Picture of electroplated ^{240}Pu (887 ± 33 μg) on a 4 μm Ti backing foil.

Figure 3. Picture of DANCE Radioactive Target Holder containing ^{240}Pu target.

Initially, we fabricated and shipped a ^{239}Pu target. Upon receipt at LANL, it was found to be slightly contaminated on the outside of the RTH. The DANCE detector area, as most of the detector areas at LANSCE, see Figure 4, is not setup to be a contamination area and therefore, the targets must be free from contamination. To eliminate future target dusting, both the ^{240}Pu and the ^{242}Pu deposits were covered with several drops of liquid VYNS and allowed to dry. The mass of the adherent VYNS film is ~100 μg/cm^2 which has little to no effect on either the incident neutrons or the exiting gamma-rays. There has been no further contamination of the targets.

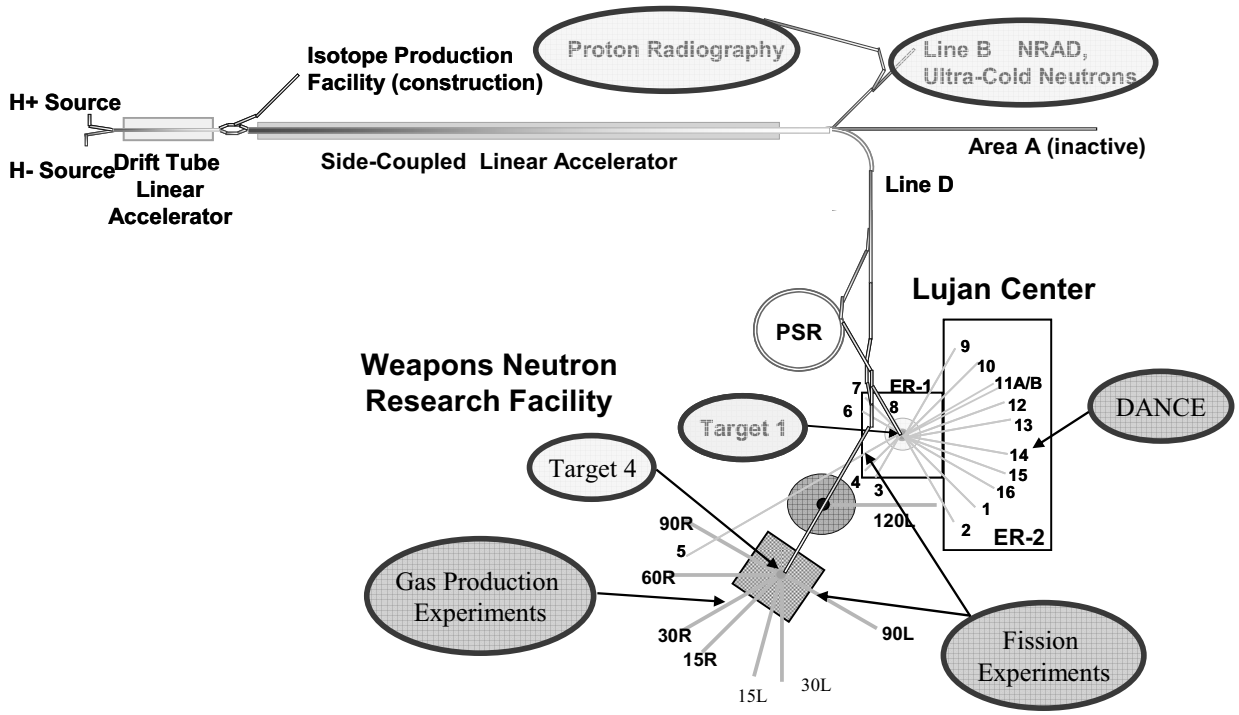

Figure 4. Schematic of LANSCE accelerator and spallation neutron source experimental areas indicating location of DANCE detector and fission detector areas.

The DANCE detector is a 4π BaF_2 scintillator that is composed of 160 BaF_2 crystals. There is a 6 cm shell of ^{6}LiH inside of the scintillator to minimize scattered neutrons interacting with the BaF_2. The neutrons come from a water moderated, tungsten spallation source and the collimation beam line is 20 m. The detector can measure neutron capture cross sections from 0.01 eV to ~ 500 KeV neutron energy. An example of the raw data that came from a DANCE measurement of ^{240}Pu is shown in Figure 5. The data set from which this graph was generated was acquired in 150 minutes. There will be a total of ~12 days of data acquisition.

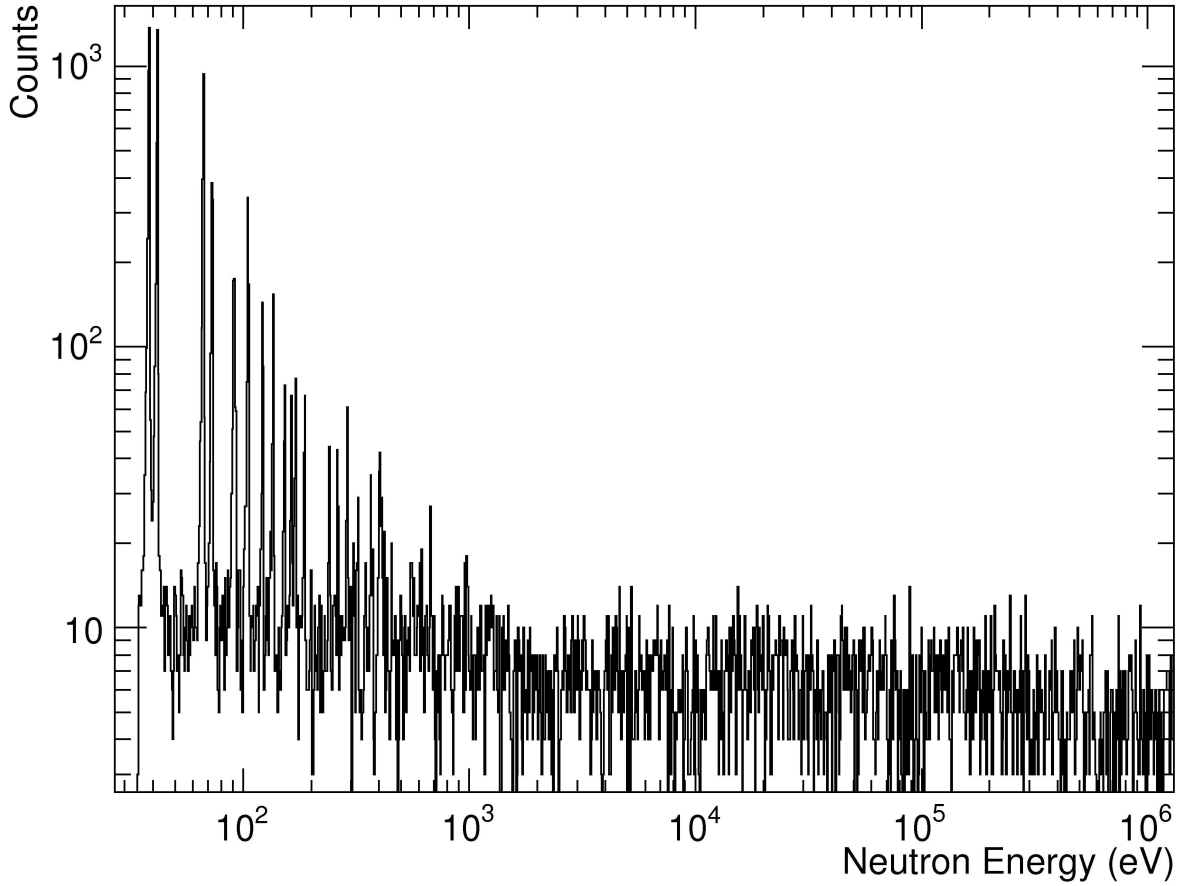

Figure 5. Plot of raw data (150 min) from DANCE detector for ^{240}Pu.

The Pu fission target chemical purification and electroplating was identical to the DANCE targets except the fission foil deposits are 3.8 cm in diameter, ~0.6 cm smaller then the neutron beam diameter. The mass density of the fission foil electrodeposited Pu is ~200 μg/cm^2 in order to minimize attenuation of the ejected fission fragments. These deposits were also covered with an ~100 μg/cm^2 VYNS film. The VYNS deposit has been calculated to attenuate a z = 53 particle (I), < 10%. Figure 6 is a picture of a ^{242}Pu fission foil (1086 ± 54 μg) covered with VYNS film.

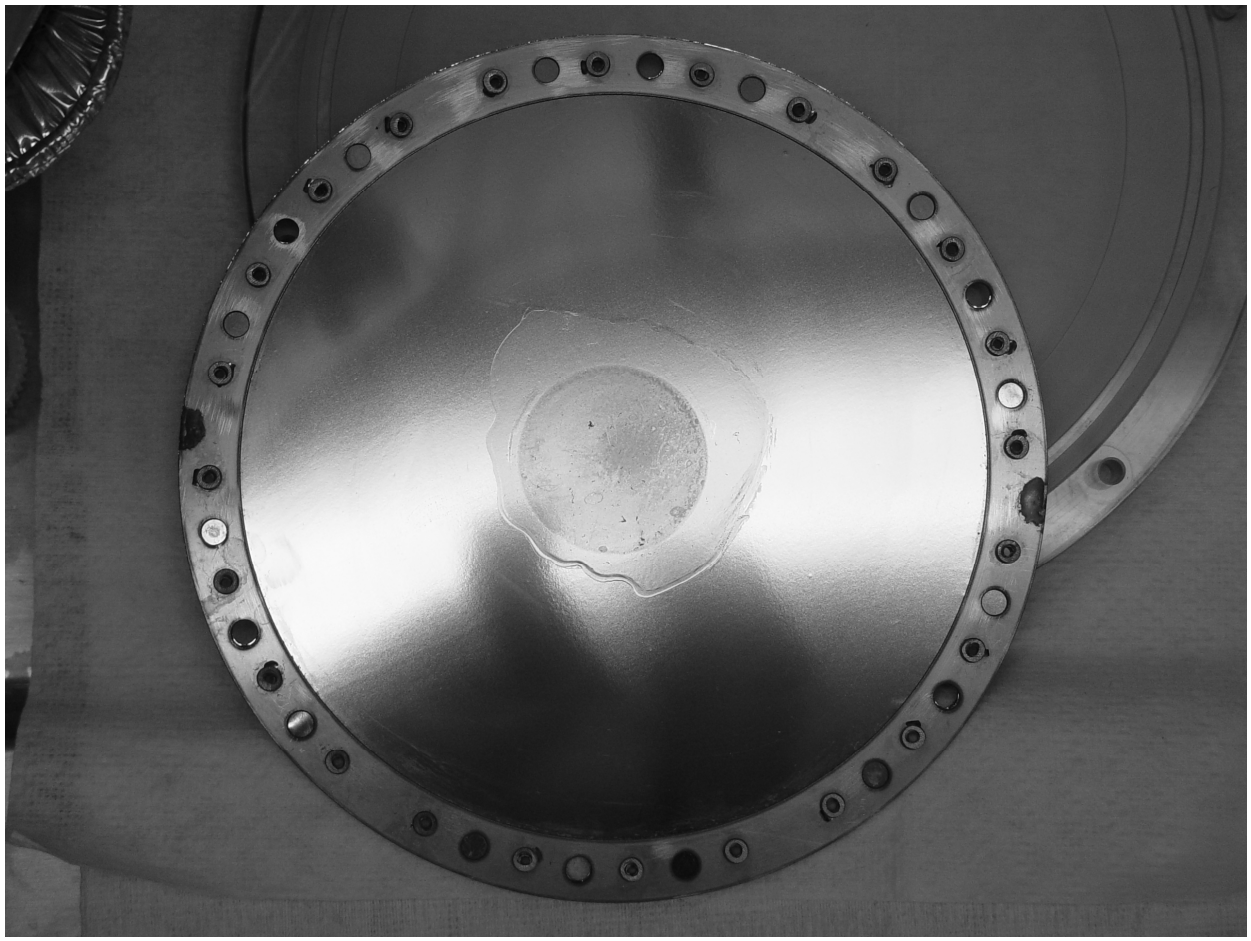

Figure 6. Picture of stainless steel fission foel containing electroplated ^{242}Pu (1086 ± 54μg).

The fission detector is a multiple target, parallel plate ionization chamber where each event has its energy deposition and relative timing recorded. Using two different spallation source areas, Lujan and WNR (see Figure 4), neutron fission cross section data can be measured for neutron energy from 0.01 eV to 200 MeV. Figure 7 is a plot of 600 hr of the raw neutron fission data versus neutron energy for ^{242}Pu.

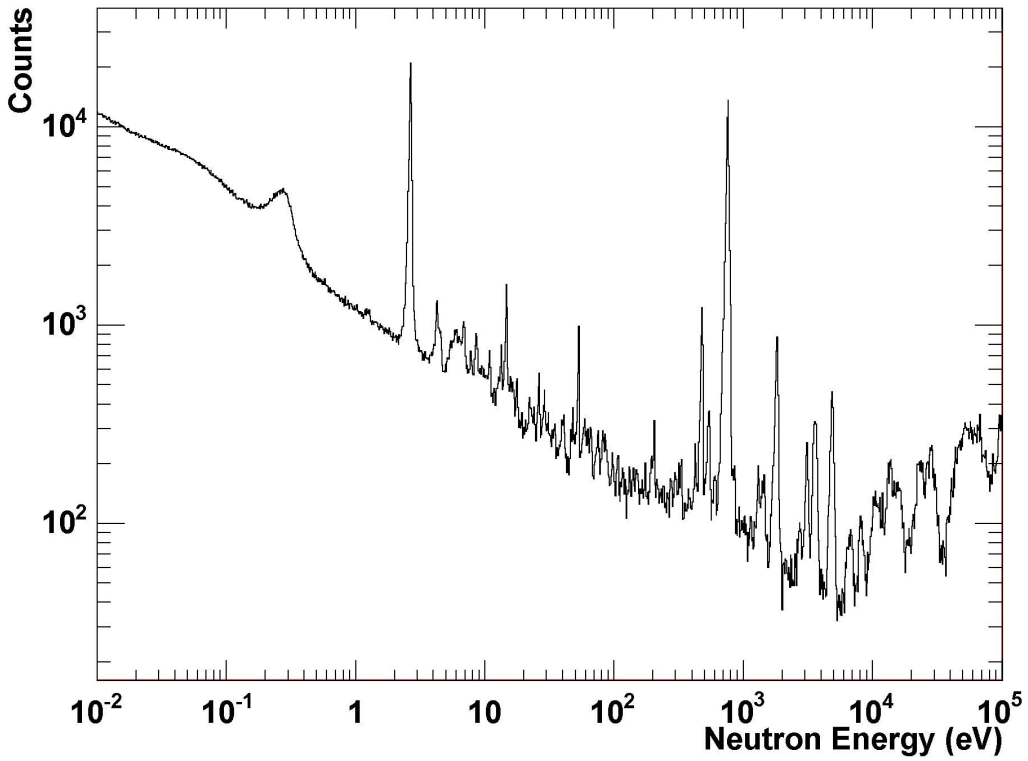

Figure 7. Plot of raw neutron induced fission data (600 hr) from parallel plate ionization chamber for ^{242}Pu.

4. CONCLUSION

In order to improve the uncertainty of the neutron cross section data for the minor actinides, INL and LANL are collaborating in the fabrication of ultra pure, highly enriched actinide isotope targets and the acquisition of neutron capture and fission data. The neutron measurement experiments take place at LANSCE. Neutron fission measurements from 0.01 eV to ~ 100 eV neutron energy, for threshold fission isotopes, such as ^{240}Pu and ^{242}Pu, are extremely difficult with the available actinide isotopes. Neutron interaction with impurity fission isotopes, such as ^{239}Pu and ^{241}Pu (see Table 1) overwhelm the threshold fission isotopes in this energy range, even when the impurity is only 0.01%. We plan to convert a small (10 μA) mass separator to allow for the high enrichment of ~1 mg masses of specific actinide isotopes to improve this issue.

5. ACKNOWLEDGEMENT

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