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PROGRESS REPORT

I. Project Summary

Over the past year, our efforts in The University of Michigan cross section project have been directed toward two general objectives: continuing measurements using our photoneutron facilities, and the development of equipment and a new laboratory to extend our capabilities into the 14 MeV neutron energy range.

The work in our photoneutron laboratory involves graduate students Dan Grady and George Baldwin. Both are within a few months of completing their doctoral dissertation experimental work, and this will conclude a major phase of our project activity. In the future, no new doctoral projects will be initiated using our traditional photoneutron sources and calibration techniques. Nonetheless, we will continue to devote enough attention to maintain these facilities because of their valuable and unique nature. In particular, we will continue to maintain our manganese bath source calibration facility because of the excellent documentation on its efficiency that we have accumulated over nine years of operation. Since it is one of only two or three such facilities currently in operation across the country, our plans are to continue to maintain this laboratory in a state that will allow its application to specific problems on short notice.

Looking to the future, our emphasis has shifted to the 14 MeV measurements. Graduate students Mehrzad Mahdavi and Ken Zasadny are currently supported by the project and are involved in the development of various components of this laboratory. Over the past year we have successfully refurbished and operated a 150 keV neutron generator in a well-shielded and low-scatter laboratory. Many aspects of the associated electronics and detection apparatus also have been designed and fabricated. Our initial measurements are aimed at fission cross sections of the common uranium and plutonium isotopes. As detailed in the attached proposal, we anticipate that we will investigate a number of other 14 MeV neutron-induced reactions in the future.

We continue our close collaboration with Prof. J. C. Robertson of the University of New Mexico in the 14 MeV program. Several components for these measurements are being developed jointly, and extended exchange visits were made by personnel from both campuses over the past year.

The following progress report is divided into four major sections. The first two concern the indium activation cross section and thorium capture cross section measurements now nearing completion in our photoneutron laboratory. Both of these measurements have significant technological interest both from a standards point of view and because of the nuclear fuel cycle importance of thorium. The third section briefly describes the fragment angular distribution measurements that will conclude our low energy fission cross section experiments. Finally, the fourth section describes our progress in establishing the 14 MeV facilities and the preliminary measurements that have been carried out in this laboratory.

II. In¹¹⁵ Capture Cross Section Measurements

The In¹¹⁵ capture cross section is being measured at the four following photoneutron energies: 23, 265, 770 and 964 keV. The reactions of interest are the following:

The measurements have involved the use of several different techniques and components. As in our previous fission cross section work, we have made use of the photoneutron sources, the manganese bath calibration system and the VES Monte Carlo computer program for the modelling of the photoneutron sources. In addition, we have developed a 4 π β gas flow proportional counter, beta-gamma coincidence counting, and an indium foil holder which allows for both the accurate placement of the foil during the irradiation and the precise determination of the source-foil separation. All experimental measurements have been completed, and we are now in the final stages of data analysis.

Indium Target Foils

A set of eight, 3/4" diameter indium foils with a thickness of 0.006 cm were prepared from 99.999% pure indium ribbon. Foil masses of about 0.14 gm were measured to an accuracy of +20 micrograms and have been observed to be constant throughout the handling required in the series of cross section measurements. Purity of the material has been confirmed by recording the gamma ray spectrum following neutron activation, in which only indium activation was detectable.

Foil Holders and Foil Activation

Three precision machined foil holder assemblies were constructed with adjustable stainless steel spacers to allow the activation of indium foils at spacings ranging from 0.4 cm to 9.0 cm between the foil and the source surface. These assemblies were attached to the removable source well bottom plate that supports the source, centered over the foil and fixed by a positioning ring within the source well. This design facilitated the precise measurement of the source surface-to-indium foil spacing. The measurements were performed using a dial gauge and gauge blocks at every 20° around the indium foil circumference. RMS deviations of less than 0.005 cm in all spacings were obtained indicating precise placement and near normal source-foil orientation.

The activations took place for between one and three hours and at several different spacings. As will be discussed later, the different spacings were required in order to obtain the room return contribution to the indium activity.

Absolute Activity Determination and the Beta Detector Efficiency

Once activated, the indium foil absolute disintegration rate must be determined. This was done in two steps. The foils were first counted in the $4\pi\beta$ gas flow proportional counter. The second and much more difficult step was the determination of the beta detector efficiency for the decay of the $\text{In}^{116\text{m}}$. This step was accomplished using a combination of $4\pi\beta$ - γ coincidence counting and beta detector efficiency variation. The experimental technique involved the determination of the apparent beta efficiency for a series of modifications in this efficiency created by the use of aluminium absorbers covering the indium foils. Graphical analysis of these data gave rise to a spectrum or "K" correction factor which accounts for the extremely complicated interaction of $\text{In}^{116\text{m}}$ decay scheme components with the beta detector.

The result of the absolute efficiency determination for the beta detector was 0.5471 ± 0.00521 , with a "K" correction of 0.97448 ± 0.0081 when using the 1.293 MeV photopeak for the gamma channel. This "K" value is in excellent agreement with the value obtained by Robertson¹ (0.9806 ± 0.006) using a different efficiency variation technique.

Further experiments were performed to determine possible effects on the efficiency due to variation in foil thickness or non-uniformity in the activity across the foil caused by variations in the flux profile. The second point has no apparent effect on the efficiency. The data to answer the first point are still being analyzed. However, since thickness variations are only a few percent, any effects should be small.

Neutron Source Strength

As in all of the past cross section measurements, we rely on the manganese bath for our neutron source strength determinations. Uncertainties in the source strength of about $\pm 0.7\%$ are limited by the uncertainties in our local standard, a Cf^{252} neutron source. Plans are underway to indirectly recalibrate this local standard against NBS-I using an intermediate standard Cf source shipped from the National Bureau of Standards.

Correction Factors

Several correction factors will also be required for this measurement series. Neutron scattering from the experimental package and containment is handled with a discrete element approximation that accounts for neutron scattering anisotropies. Scattering from the target foil backing is handled by a Monte Carlo path length procedure due to the extremely close proximity of the scatterer and target.

The contribution of competing neutron-induced reactions in indium to the $\text{In}^{116\text{m}}$ activity must also be accounted for. Inelastic scattering reactions in In^{113} and In^{115} give rise to metastable states whose decay may be detected in the proportional counter. Beta detector efficiencies of 0.2% and 30.7% for these

two isotopes ($\text{In}^{113\text{m}}$ and $\text{In}^{115\text{m}}$ respectively) have been determined using absolute gamma counting techniques.

Additional corrections for the contribution of room return activity to the total indium activity and neutron energy spectrum effects are also being determined in the capture cross section measurements.

Present Status

With all of the necessary experiments completed, the analysis of the results has begun in earnest. The In^{115} capture cross section determinations are expected to be completed by the end of this contract year.

III. Absolute Measurement of the $^{232}\text{Th}(n,\gamma)$ Cross Section at 24 keV

Experimental measurements are currently underway to absolutely determine the neutron radiative capture cross section of thorium-232 by an activation method using an antimony-beryllium photoneutron source. Recent thorium data base evaluations^{2,3} have indicated a high-priority need for more accurate determinations of this cross section, to resolve appreciable discrepancies between previous experimental values. Unfortunately, additional measurements at higher energies with other available photoneutron sources at this laboratory are not feasible, primarily owing to the low time-integrated neutron yield of those sources.

Overview of Method

The photoneutron source activates a relatively large cylindrical target of natural thorium for approximately two weeks. The flux is calculated by Monte Carlo modeling. The source strength is calibrated against a Cf-252 standard as in previous measurements using a manganese sulfate bath system. Neutron captures in thorium initiate the sequence:

The cross section is obtained from a determination of the absolute amount of ^{233}Pa produced. A Ge(Li) detector is used to count the 312 keV gamma line in ^{233}Pa decay, for which the branching ratio is well known.⁴ To improve the geometry of the counting and isolate the low-level ^{233}Pa activity from high natural thorium background, the protactinium is first separated from the target chemically. A solvent extraction is performed with diisobutylcarbinol,

with another isotope (^{232}Pa) employed as a tracer to measure protactinium recover.

Thorium Target

Three targets of natural thorium metal have been obtained from the Isotope Target Laboratory at the Oak Ridge National Laboratory. The analytical chemistry division at ORNL determined the thorium content to be 98.5% purity. Each target is a flat sheet 0.05 cm (0.020") thick, which is rolled into cylindrical shape (12 cm. high x 4 cm. radius) and clamped in place on a supporting framework.

The supporting framework has been designed and constructed. It consists of a 5 cm. O.D. brass source well with the 8 cm. I.D. target mounted concentrically. Two aluminum rings top and bottom hold the target to the source well in this geometry. The source is supported in the well at the midpoint of the target by an aluminum crosspiece insert. Much of the wall of the brass source well was machined away to reduce the scattering perturbation.

Target Flux Calculation

A computer code was written to model neutron transport specifically for the source-target geometry used in this measurement. A spherical source of finite extent emits neutrons uniformly but distributed in polar angle according to the description provided by the VES code. Neutron histories are traced to the enclosing thorium cylinder, where their path lengths through the target are assumed. This sum, appropriately normalized, gives the average scalar flux at the target. The code is a one energy group model, but accounts for scattering in the target and in the brass source well between source and target. Results of the code have been checked against the analytically-derived flux expression for the Sb-Be source used by Engdahl.⁵

Flux Sensitivity

The cylindrical target geometry has an advantage of being relatively insensitive to error in source position. A quantitative analysis was made to determine the deviation of the average scalar flux seen by the target as a function axial and radial source position offsets from center. This study was performed for a variety of target dimensions before selecting the present configuration.

Chemical Separation

A carrier-free solvent extraction scheme to chemically separate protactinium from thorium has been developed using reactor activated thorium. Excellent radiochemical purity has been achieved, and recovery yields on the order of 70% have been accomplished with approximately 2 μCi of ^{233}Pa in 100 grams of thorium. Refinements are now being made with the expectation of further increasing the yield percentage.

Chemical Yield Measurement

^{231}Pa had initially been considered as a tracer isotope for the yield measurement. It could be used directly, requiring thin source preparation and alpha counting, or more easily with reactor activation to ^{232}Pa (200 barn activation cross section), enabling gamma counting. Although ^{231}Pa is available for purchase, it is classified as byproduct material and would first require amendment to our NRC license. This would likely have entailed an interminable delay.

Instead, in order to expedite matters, an alternate idea was conceived and tried successfully. This involved producing the ^{232}Pa isotope directly by the $^{232}\text{Th}(p,n)^{232}\text{Pa}$ reaction. Feasibility calculations were made which indicated that the method looked possible with 10-12 MeV protons, although a fission rate six times higher was also to be expected. Thorium foil was readily available, and arrangements were made to make use of the tandem van de Graaff facility at Western Michigan University. A sample was irradiated for 3 1/2 hours at 0.5 μA 11 MeV beam current. Ge(Li) spectroscopy verified the presence of relatively large numbers of fission products, but chemical separation (the same procedure described previously for obtaining protactinium from thorium) retrieved the ^{232}Pa in excellent purity. The ^{232}Pa solution is used immediately after extraction for tracing the $^{233}\text{Pa}/^{232}\text{Th}$ separation. It is sampled volumetrically by micropipette: one aliquot is used to spike the dissolved neutron-activated thorium target; one or more other are used as control samples (do not go through the separation). After separation, the liquid volumes are made equal and the samples counted in the same geometry. The ratio of the counts in the prominent 969 keV gamma peak gives the yield directly.

Absolute ^{233}Pa Activity Measurement

The ^{232}Pa tracer isotope has only a 1.3 day half-life, and therefore quickly decays to reduce the Compton continuum background that underlies the 312 keV peak of the 27 day half-life ^{233}Pa . The 312 keV peak is then counted again by a relative count against another liquid solution sample of ^{233}Pa (equal volume, same geometry) used as a reference. The reference sample in turn is produced in advance of the activity measurement by equal-volume sampling of a ^{233}Pa stock solution (from a chemical separation of reactor activated thorium) both into the liquid counting container and onto a planchet which is dried. The evaporated deposit is then standardized by absolute Ge(Li) counting (also considering $4\pi\beta\text{-}\gamma$ coincidence). The Ge(Li) detector efficiency is calibrated using a mixed radionuclide standard obtained from Amersham-Searle and traceable to NBS.

Present Status

The pre-run calibration of the Sb-Be photoneutron source has been completed. Final details of the chemical separation procedure should be completed very shortly. At least two and possibly three measurements will be made, with the first target irradiation expected to begin before the end of November 1980. Experimental work should be complete by the end of this contract year.

III. Conclusion of the Photoneutron Fission Cross Section Measurements

Angular Distribution of Fission Fragments

Accurate knowledge of the fission fragment angular distribution for the various neutron energies at which our fission cross section measurements have been made is very important. The need for this information at the specific energies of our photoneutron sources arises from the fact that we utilize limited solid angle counting to determine fission rates. Our policy continues to be that we will make these anisotropy measurements using the Tandem Dynamitron Neutron facilities at ANL.

Work on the Pu^{239} fission fragment anisotropy has been completed. The measurements were made at the neutron energies of 265, 770 and 964 keV; our three highest energy photoneutron sources. In the analysis of the angular distribution data, corrections were made for solid angle effects, neutron scattering, lower energy neutron groups and smearing effects caused by the finite angular resolution. The data were fitted to the expression

$$W(\theta) = 1 + A \cos^2 (\theta)$$

where " $W(\theta)$ " is the normalized angular distribution, " θ " is the center-of-mass angle between the incident direction and the fission fragment direction and " A " is the anisotropy coefficient (the quantity of interest). Our results are tabulated below and also have been entered on Figure 1, together with results of other anisotropy measurements in the 0-2 MeV neutron energy region.

<u>ENERGY</u>	<u>ANISOTROPY</u>
265 keV	.035 $\pm$.012
770 keV	.076 $\pm$.011
964 keV	.106 $\pm$.020

Experimental work on the U^{233} fission fragment anisotropy has been completed. Here again the measurements were made at the neutron energies of 265, 770 and 964 keV. The track recorder films have been etched and gridded and counting and analysis is expected to be completed by January of 1981. The experimental work on the Np^{237} fission fragment anisotropy has also been completed. In this case, the measurements were made at only 770 and 964 keV because of the threshold-like behavior of the fission cross section near 600 keV. Again the films are now being counted and the final anisotropy results are expected to be completed in January.

Absolute Measurement of Fast Neutron Fission Cross Section for ^{233}U

Track counting for the fission rate determination at all five photoneutron source energies is complete. Analysis is in progress and should be ready for publication early in 1981. Final cross section determination is awaiting the analysis of the fission fragment anisotropy measurements for ^{233}U outlined above.

Figure 1

IV. Facilities and Measurements at 14 MeV

Overview

Since the last progress report, our efforts in the 14 MeV cross-section project have been in the following major categories:

1. Construction work, equipment moving and general setup of radiation laboratory. This phase has almost been completed and only minor work is left to be done.
2. Finishing the design and production of various components of the accelerator, its setup on the low-mass platform, and testing the operation and optimization of its electronic controls. An existing leak detector was renovated and installed as an important part of the vacuum system. This part of the operation has been completed and we have routinely produced a 14 MeV neutron yield of up to 10^9 n/s. Higher yields will be possible with alternative targets.
3. Setup and testing of diagnostic equipment for various purposes. Included are: long counter, Bonner sphere, surface barrier detector and related electronics, multi-channel analyzer, tritium monitors, computer terminal, leak detector, survey meter, etc. The long counter and Bonner sphere are set in fixed positions to give relative flux levels and can also be used for dose rate estimation. The silicon detector, with its associated electronics, is used for accurate neutron energy and yield measurement. More detailed descriptions are given later in this report.
4. Development of safety procedures for handling and monitoring radioactive materials, specifically tritium handling and storage. Ventillation of the area used for handling targets has been provided along with monitoring of exhaust air. Shielding has been installed as required for the 14 MeV neutrons. Warning signals and an interlock system have been put into place for use during the accelerator operation.
5. Preliminary measurements have been started to provide an essential basis for our first fission cross section determination. Included are neutron energy measurement, relative and absolute flux measurement, angular distribution of fission fragments, iron foil activation and detector efficiency determination, and determination of room return flux. The status of our progress towards each one of the above major tasks will be outlined in the remainder of this report.

Preparation of the Laboratory - The Neutron Experimental Bay

The major construction work has been completed to convert the former large cyclotron bay into the "Neutron Experimental Bay" of the Department of Nuclear Engineering. Attention has been given to proper shielding, ventillation, and equipment installations. Figure 2 shows the layout of the lab after our construction work.

FLOOR PLAN OF THE
NEUTRON EXPERIMENTAL BAY

Figure 2

In the shielding design, calculations were made assuming a neutron strength of up to 10^{11} n/s. Addition of portable concrete block shielding to existing concrete walls has provided the necessary neutron attenuation.

For proper ventilation, two large existing air conditioning and circulating systems were renovated and are in full operational order at this time. Additional ducts have also been constructed as a part of independent ventilation of the accelerator area where the tritium targets are handled and stored. A fume hood was designed and constructed for use in storing contaminated parts of the accelerator and for other tritium handling tasks. A large and completely sealed glove box with proper filters and blowers was purchased and installed to store the tritium targets.

In future construction work we anticipate setting up a small machine and electrical shop in the large working area of the Neutron Experimental Bay. Additional shielding reinforcement may be required as we change to higher loading tritium targets and higher neutron yields, but the present configuration is adequate for our initial measurements.

Accelerator Setup and Operation

Since the last progress report, all components of the accelerator have been renovated and it is now in full operational order. Figure 3 shows an overall schematic of our system. A brief description of major parts of the system are given below.

The ion source generator is based on an r-f oscillator, and induced gas discharge. The accelerating tube is capable of high voltage up to 150 kV. Figures 4 and 5 show an overall view of high voltage terminal area and ion source with its related components.

The vacuum system consists of three major stages: first, a two stage mechanical pump to reduce the pressure to about 10^{-2} torr, second, a sorption pump to produce vacuum to 10^{-3} torr, and the third stage is an ion pump which is capable of producing a 10^{-6} torr static and 10^{-5} torr operating pressure. In order to test the system, a leak detector of mass spectrometer type was renovated and installed. This will be of continuous service to our existing vacuum system and in troubleshooting new vacuum chambers being designed. The exhausts from the mechanical and sorption pumps are routed to a tritium monitor for possible tritium gas contamination and eventually vented through special duct work.

The target assembly is attached to the drift tube by a bellows for added maneuvering of the target chamber and target head. The target head has been made from pure aluminum and kept as thin as possible to reduce scattering and undesired (n,2n) reactions. The target is cooled by forced air.

The accelerator is installed on a low-mass floor which was constructed specifically for this purpose. The neutron source is almost at the geometrical center of the room. In this manner a large distance from the thick concrete walls and floor will greatly help in reducing room return neutrons.

Figure 3

Figure 4

ION SOURCE
FIGURE

Figure 5

FIGURE

Neutron yields of up to 8×10^8 n/s have been produced with the generator to the present date. Additional work is planned to improve the beam transport efficiency and control by aligning the entire beam path and centering it with a carefully positioned laser beam. This step should reduce the demand on the deflection plates and focusing lens to steer the beam, and will result in higher achievable beam currents and better operating vacuum.

Diagnostic Equipment Installation and Operational Testing

We have installed two Bonner sphere neutron monitors to determine fast neutron dose rates at several locations throughout the laboratory. A standard long counter has also been assembled and is located in a fixed position 5 m from the neutron source in the accelerator room. The counting rate for the long counter is used as a direct gauge of neutron yield as a function of time, and has been absolutely calibrated against iron foil activations to provide an absolute scale.

A new tritium gas monitor has been purchased and installed in the vent line of our main vacuum system. A second flow-through monitor has been renovated and installed in the exhaust line from various pumps and glove boxes in which tritium targets and other contaminated equipment are handled. A general beta-gamma survey meter has also been provided for use in monitoring routine radioactive materials.

We have acquired a Nuclear Data Model 66 multichannel analyzer as the heart of our data recording system. This is a very versatile unit based on a LSI-11 microprocessor and can be configured in a variety of ways to suit different experiments. Current output is via a TI-700 terminal with cassette recording for local storage and direct connection to the Michigan Terminal System main computer via a modem telephone connection.

Safety Procedures

Because of the potential hazard of the 14 MeV neutrons and tritium-containing targets, operational procedures for routine facility use have been worked out in conjunction with the U of M Radiation Control Service.

Using present targets and shielding, fast neutron dose rates are well below 1 mr/hr at all accessible locations during operation. Monitoring of these levels will continue as new experiment conditions dictate.

During the high vacuum phase of operation of the accelerator, there is no exhaust of the system since we are relying on an ion pump in this vacuum range. However, during roughing-in periods following target change or other maintenance, the vacuum exhaust is monitored using a flow-through tritium monitoring unit. Abnormally high values have not yet been observed in these procedures. Future steps may require the installation of traps or filters in this line should appreciable tritium contamination be observed.

Special precautions are observed with changing tritium targets. Because substantial off-gassing can occur, all procedures are carried out using a small portable glove box which is sealed around the target area. Exhaust from the glove box is again monitored using a flow-through tritium monitor unit. Additional tritium detection capability is provided by a portable tritium sniffer used in conjunction with a liquid scintillation counter. New or used tritium targets are stored in a stainless steel glove box maintained at negative pressure. Again its exhaust is monitored for any tritium level.

Additional routine monitoring is carried out periodically of the entire laboratory area. Smear papers and liquid scintillation counting are used periodically monitor floors and working surfaces. Shoe covers and lab coats are required of all personnel working directly on the accelerator platform. Period bioanalyses are required of operating personnel, particularly following target changes.

Initial Measurements

Initial neutron irradiations have been carried out using both a U-235 fission foil and an iron activation foil target. These irradiations were conducted in a vacuum chamber designed for this purpose and illustrated in Figure 6. Main construction was of 20 mil aluminum to hold neutron in-scatter to a minimum. The target-detector aperture is also constructed of low-mass materials.

A preliminary fission irradiation was carried out to identify potential problems in the ultimate cross section measurements. The measurements were conducted at zero degrees with respect to the deuteron beam. Figure 7 shows the overall experimental arrangement. Alignment was carried out using a small helium-neon laser to ensure accurate centering of the U-235 target along the beam axis. Optical reflection from the backing of the target is also employed to ensure perpendicularity of the foil mounting. In order to support the vacuum system and provide means for its accurate positioning, a low mass positioning structure has also been provided.

In order to carry out an absolute measurement, both the source-to-target distance and the target-detector spacing must be defined to high accuracy. An inside micrometer is used to determine the spacing between the parallel planar surfaces of the tritium target and uranium target. Measurements are made both before and after the irradiation to provide some gauge of reproducibility. The uranium foil to detector spacing is determined using methods similar to those already developed in conjunction with our photoneutron series of measurements. Gauge blocks and dial gauges are used that are capable of accuracy within a few ten thousandths of an inch. Measurements are again carried out both before and after each irradiation.

We have carried out a total of 9 preliminary runs in which fission tracks were recorded on polyester film detectors. After etching and gridding we have observed track densities of approximately 15-25 tracks per frame. Projected to the irradiation geometry to be used in the final measurement, we estimate counting statistics will contribute less than 1% to the overall uncertainty

Figure 6

OVERALL SCHEMATIC OF SET UP FOR FISSION RUNS

of the measurement. The preliminary runs were also used to investigate the secondary particles and other backgrounds during the 14 MeV irradiation.

Irradiations of iron foils have also been carried out as a function of source-foil spacing. The objective of this series was to carry out a measurement of the room return neutron flux by observing departure from inverse square behavior. Because of the low activation cross section of iron at these energies, we are restricted to spacing of no greater than 15 cm using present thin targets. We plan a repeat of this series using a thicker target which, while degrading the neutron energy spectrum somewhat, will allow a greater range of measurements to be carried out. The induced iron activity is counted in the same proportional counter developed for use with our indium activation measurements described earlier. Methods for verifying absolute detection efficiencies are currently under active development.

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