FISEVIER

Contents lists available at ScienceDirect

Nuclear Inst. and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb



Laser ablation positive-ion AMS of neutron activated actinides



R.C. Pardo^{a,*}, T. Palchan-Hazan^c, R. Scott^a, M. Paul^c, O. Nusair^a, W. Bauder^b, R. Vondrasek^a, D. Seweryniak^a, S. Baker^a, R. Talwar^a, P. Collon^b, F.G. Kondev^a, G. Youinou^d, M. Salvatores^d, G. Palmiotti^d, J. Berg^d, J. Giglio^d, M.T. Giglio^d, G. Imel^e, C. Nair^a, C.L. Jiang^a

- ^a Argonne National Laboratory, Argonne, IL, USA
- ^b Nuclear Structure Laboratory, University of Notre Dame, Notre Dame, IN, USA
- ^c Racah Institute of Physics, Hebrew University, Jerusalem, Israel
- d Idaho National Laboratory, Idaho Falls, ID, USA
- e Idaho State University, Pocatello, ID, USA

ARTICLE INFO

Keywords: Actinide AMS Laser ablation Neutron capture Cross section

ABSTRACT

At Argonne we have enhanced the AMS capabilities of the Argonne Tandem Linac Accelerator System (ATLAS) to attempt to minimize crosstalk between samples and to quickly move from one (M/Q) setting to another in order to make measurements on a large number of samples provided by the MANTRA (Measurement of Actinide Neutron Transmutation Rates by Accelerator mass spectrometry) project. Those improvements include the use of a picosecond laser to ablate actinide material into the source, a new 20-sample holder that can switch samples within 1–2 min, and a number of accelerator configuration improvements that allow quick and precise switching between species. In principle, AMS can provide production yields of actinide isotopes produced during the irradiation period at a sensitivity exceeding other mass spectrometry techniques. A total of 27 irradiated samples of a variety of actinides have been provided for measurement. We discuss our experience with these facility improvements and how well we have met our performance goals. In addition, we present preliminary results on a number of the irradiated actinide samples with this approach and compare those results to Multi-Collector ICPMS measurements.

1. Introduction

Neutron cross-sections characterize the way neutrons interact with matter. They are essential to most nuclear engineering projects and, even though theoretical progress has been made as far as the predictability of neutron cross-section models, measurements are still indispensable to meet tight design requirements. In particular, the assessment of advanced fuel cycles requires an extensive knowledge of transuranics cross sections. Plutonium isotopes, but also americium, curium and up to californium isotope data are required with a small uncertainty in order to optimize significant features of the fuel cycle that have an impact on feasibility studies (e.g. neutron doses at fuel fabrication, decay heat in a repository etc.).

The Measurement of Actinide Neutron Transmutation Rates by AMS (MANTRA) project aims to improve the quality of neutron capture cross section data by making a series of energy-integrated cross section measurements on a variety of actinide isotopes. Isotopically pure actinide samples were irradiated in the Advanced Test Reactor (ATR) at Idaho National Laboratory (INL) under a set of 3 neutron filters: 1 mm

cadmium, 5 mm boron (70% 10B), and 10 mm boron (70% 10B). These filters cut out various portions of the thermal neutron spectrum and isolate a region of the epithermal or fast energy regime.

Due to the irradiation, a series of isotopes build up in the samples from successive neutron captures. From starting nuclei of mass A, products will build up with mass numbers A+1, A+2, etc. By measuring the isotopic ratios of these capture products before and after irradiation, the energy integrated capture cross section can be extracted by:

$$\sigma_{A}^{c} = \frac{\frac{N(T)_{A+1}}{N(T)_{A}} - \frac{N(0)_{A+1}}{N(0)_{A}}}{\varphi T}$$

where σ_{A}^{c} is the energy-integrated one neutron capture cross section, $\frac{N(T)_{A+1}}{N(T)_{A}}$ is the isotopic ratio of A + 1 to mass A after irradiation, $\frac{N(0)_{A+1}}{N(0)_{A}}$ is the isotopic ratio before irradiation and φT is the neutron fluence. Similar equations can be derived for 2 and 3 neutron capture cross sections. Thus, from only isotopic ratio measurements and the total neutron fluence, the neutron capture cross sections can be extracted.

E-mail addresses: pardo@anl.gov, pardo.richard@comcast.net (R.C. Pardo).

^{*} Corresponding author.

This technique has been used before to extract energy integrated cross sections [1]. The key innovation of the MANTRA project is that many isotopic ratios are measured using Accelerator Mass Spectrometry (AMS). Because AMS has a much higher sensitivity than conventional mass spectrometry techniques like ICP-MS many more cross sections can be extracted from a single sample while only irradiating for 50–100 days. This allows the measurements to be completed quickly and limits the radiological risk of handling the samples after irradiation.

The predicted isotopic ratios built up from 50 days of irradiation with a 5 mm boron filter, assuming a nominal average cross section of 500mb for actinides is approximate $\sim\!10^{-4}$ for 1-neutron capture, $\sim\!10^{-8}$ for 2-neutron capture, and $\sim\!10^{-11}$ for 3-neutron captures. In most cases, one neutron capture cross sections are accessible through AMS or conventional mass spectrometry. However, the two or three neutron capture cross sections are much weaker channels and are difficult to access with other techniques.

The ability to extract multiple cross sections from a single sample combined with the irradiation of many different actinides allows MANTRA to make a rather comprehensive set of cross section measurements across the actinide region. Eight actinide materials have been identified as prime candidates for measurement by AMS: ²³²Th, ²³⁶U, ²³⁸U, ²³⁷Np, ²⁴²Pu, ²⁴⁴Pu, ²⁴³Am, ²⁴⁸Cm. The technique used to measure the isotopic ratios with AMS will be discussed in this paper as well as some preliminary example results.

2. AMS at ATLAS and facility modifications for MANTRA

Many heavy ion AMS experiments have been successfully performed at ATLAS which is ideally suited for heavier ions because of the ability to reach much higher energies [2,3]. However the ATLAS facility is also much larger and has much less automation than smaller AMS facilities meaning that systematic uncertainties associated with small changes in beamline parameters are much harder to manage and measure. Past experiments have been able to overcome these challenges because the experiment typically only measures 1 isotopic ratio in 3-5 samples. Thus, the demands on the accelerator in terms of scaling between many different masses and maintaining long term stability are much lower. However, the MANTRA project, by contrast wants to measure up to 15 samples in a single 5 day experiment and each sample will require the measurement of 2-3 different isotopic ratios for each sample. The need to measure so many different samples and isotopic ratios in a single experiment in a very limited amount of facility time requires changes in ATLAS equipment and procedures. Several key improvements have been made to the ATLAS facility for this project in order to manage the number of samples, move between accelerator tunes, normalize isotopic ratio measurements with electrical pico-amperes (epA) beam currents, and reduce cross talk between samples in the ion source.

When the MANTRA project began, nearly ten years ago, the use of low voltage tandems had been demonstrated but was not in widespread use for actinides at that time and there was some uncertainty concerning normalization and background limits to sensitivity. Although the ratios for 1n and 2n are high by AMS standards, the total beam current was expected to be quite low and total efficiency is important. That combined with collaborating with a 'sister' laboratory made ATLAS an interesting facility for this work.

Fig. 1 shows a floorplan of the ATLAS facility. The active significant portions used for the MANTRA AMS measurements are highlighted in black boxes.

Ions are produced in the Electron Cyclotron Resonance (ECR) ion source and are extracted in high charge states, typically 37 + to 40 + tor 40 + t

delivered by the beam transport system to the FMA where they are stripped to a higher charge state by a $50 \,\mu g/cm^2$ carbon stripping foil. This stage of stripping reduces the beam rigidity, slightly, and provides one additional stage of background reduction. The ions are then mass separated and identified in the FMA.

2.1. Automated accelerator scaling and system transmission

At the ATLAS accelerator, configurations are initially set, in most cases, by scaling from a past similar (M/Q) beam. Then the transmission for the beam of interest is optimized manually from the reference setting. In this case, transmission is defined as the current from a Faraday Cup just after the second large dipole magnet directing the beam into the radio frequency quadrupole (RFQ) shown in Fig. 1 to a cup at the end of the FMA immediately in front of the detector system. Thus the fractionation from the stripping foil at the FMA target location is included. For AMS experiments, the real beams of interest are much too weak to allow such tuning and optimization, so a similar (M/Q) 'guide' beam is used for this initial configuration and then a slight shift is made for all elements to the actual (M/Q) of interest. In this case 82Kr13+ or was used for the initial guide beam and the transmission was measured for that beam. For these actinide measurements, this transmission must also be measured for the entire range of M/Q for the actinide measurements and a small correction factor applied to each measurement (typically a few percent). This was done by also using another krypton isotope, ⁸⁶Kr¹⁵⁺. The two isotopes completely spanned the (M/Q) range of all the actinides. A third check of transmission in the (M/Q) range of interest was made with 83Kr14+. These tunes are then saved in a central database which records all the accelerator and beamline element parameters for that mass.

Once in the database, the setting can be loaded in a few minutes. Most of the time to load these differing tunes is due to the time required to cycle magnetic quadrupoles through their hysteresis loop and to directly set dipole magnets precisely based on Hall probe measurements. Typical machine changes between M/Q settings as well as selecting different samples can now be accomplished in about 10 min. A typical MANTRA measurement requires about 15 min of data collection. With this automated accelerator scaling, an isotopic ratio can be measured in approximately 30 min (admittedly still slow by small-accelerator automated AMS facility standards). This helps minimize systematic errors due to shifts in accelerator configuration and also makes it possible to measure many more samples in a single experiment. In particular, the overall beam transport efficiency, or transmission, was measured to vary by < 5% in all cases and typically the variation was ~2% for most cases (see Fig. 2). Other isotopes of Kr are available for transmission checks. We also measured the transmission for 86Kr14+ which agrees within the current uncertainties of these measurements of approximately 1% which is not shown in Fig. 2. Because other issues have limited our present accuracy, we have not pursued a more detailed check of transmission variation with M/Q, but other AMS measurements indicate this measured variation is typical for ATLAS. Compounding these issues of transmission normalization is the fact that, over time, beamline elements (focusing elements, steerers, etc.) will shift a small amount. Thus, the absolute transmission may be different between measurements. To understand these shifts, the transmission is always measured multiple times over the course of an experiment.

By measuring the beam current of the 'mother' material (the original actinide material subject to irradiation) at the FMA detector before and after each measurement, one also reduces the sensitivity to fluctuations in the transmission over time. Except for the variation in transmission for the mother (M/Q) compared to the neutron-capture (M/Q), the absolute transmission divides out and is not part of the ratio. Only the small variation with (M/Q) remains – assuming a constant value during the measurement period. Therefore a suppressed Faraday cup, coupled to a local Keithley 6485 femtoammeter [5] was used to measure the beam current of the mother material before and after each

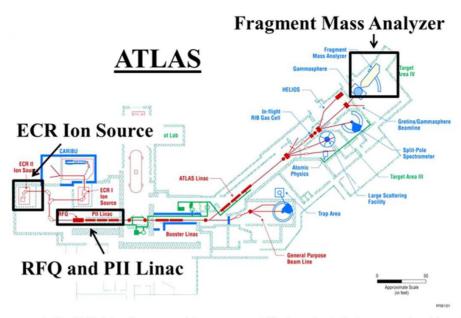


Fig. 1. A floor plan of the ATLAS facility highlighting the areas used for MANTRA. Highly charged actinide ions are produced from the ECRII ion source then accelerated to 1 MeV/u. The detection takes place at the Fragment Mass Analyzer.

measurement.

2.2. Multi-sample changer coupled to the ECR ion source

The MANTRA project involves not only a large number of isotopic ratio measurements but also requires measurements on a large number of samples. In previous experiments at ATLAS, only 3–5 samples were measured during a single 5–7 day AMS experiment. The ion source could only accommodate one sample at a time and, in order to change samples, the ion source would need to be stopped and the sample would be removed through an airlock. The new sample would then be inserted from atmosphere, the airlock is pumped out and then the source is restarted. With this process, because the vacuum is broken, there is also

typically a period of conditioning for the source and restoring the previous conditions. In an ideal scenario, this change takes at least 30 min. Not only is this process time consuming for MANTRA, which may measure 15–20 samples in a single experiment, but the process of stopping the ion source can introduce uncertainties because changes in the source conditions affect the ion production.

To solve this problem, a new multi-sample changer [6] has been installed parallel to the plasma axis of the ECR source. Fig. 3 shows a CAD drawing of the multi-sample changer attached to the injection side of the ECR chamber.

Inside the chamber, 20 samples are attached to retractable rods in a "Gatling gun" formation. One of a pair of stepper motors first rotates between samples and then the second motor moves the samples in and

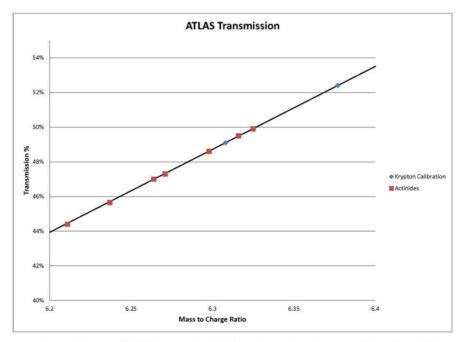


Fig. 2. Plots of accelerator transmission as a function of M/Q. The transmission is determined using Kr isotopes and then the actinide transmission for each actinide's M/Q is determined. The two isotopes of Kr shown here and $^{83}\text{Kr}^{13+}$ and $^{82}\text{Kr}^{13+}$.

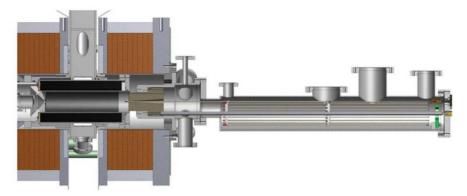


Fig. 3. A CAD drawing of the multi-sample changer attached to the injection side of the ECR. Retractable rods insert the sample through the iron plug in the back of the chamber. The motors are mounted outside the vacuum chamber on the right side of this drawing, but are not shown.

out of the plasma chamber. When inserted, the samples pass through an iron plug in the back of the ECR and protrude about 2.5 cm into the chamber. All the samples are held under vacuum. This allows us to seamlessly change between 20 samples without altering the source conditions or breaking vacuum. This reduces the sample change time to less than 5 min.

2.3. Laser ablation at the ECR

Apart from increasing the speed of measurements, the variety of samples requires us to better control cross talk due to contamination of the ion source. In order to produce ions with the ECR ion source, it is necessary to inject solid material into the plasma region where it is ionized by electron collisions between the plasma electrons and neutral atoms. In the past, AMS experiments have relied on injecting material using a solid slug or packed powder sample and an electrical sputtering technique [7]. This technique requires moving a sample radially into the ion source, near the edge of the plasma region, and biasing it to a negative voltage, typically 500-1000 Volts. This voltage attracts ions from the plasma which sputter material off the sample. The ejected material is then incorporated into the plasma and ionized. This technique can produce electrical microamperes of beam current but also produces a lot of contamination of the ion source. In particular, material will be sputtered onto the walls of the plasma chamber and that material can be recycled by the source. This residual material remains in the source even after the sample is removed and continues to be ionized, an effect known as cross talk.

Cross talk effects exist in all ion sources and are particularly problematic for AMS experiments which rely on measuring extremely small isotopic ratios. Any residual material can create a false signal which may swamp the isotopic ratio in the sample. In MANTRA these effects are compounded by the fact that the variety of samples creates additional mass conflicts. For example, a $^{237}{\rm Np}$ sample would directly interfere with mass 237 measurements made on a $^{236}{\rm U}$ sample. Even a part in 10^4 cross talk effect would significantly impact the ability to measure the one neutron capture cross section in $^{236}{\rm U}$.

We have attempted to limit the impact of cross talk for the MANTRA project, by developing a new material injection method as an alternative to electrical sputtering. In this new method, a short pulse diodepumped Nd:YAG laser at 1064 nm is used to ablate material from the sample into the plasma region. The laser has a variable energy per pulse of up to 4 mJ, a pulse width of 15 ps and a maximum repetition rate of 400 Hz. The spot diameter on target is 0.5 mm. Fig. 4 shows a diagram of the ECR source with the new laser ablation injection method.

In this scheme, the laser is directed through the plasma chamber onto a sample located parallel to, but slightly offset from the plasma axis on the injection side of the ECR. When the laser is focused on target it deposits $\sim\!10^{11} \text{W/cm}^2$ onto the sample surface. This energy

deposition creates an instantaneous heating of the top few layers of the sample and the heated material is ejected (ablated) back from the sample surface. This process is known as laser ablation [8,9]. The advantage of laser ablation for reducing cross talk is two fold. First the amount of material ablated is directly proportional to the intensity of the incoming laser pulse, in other words, the laser pulse energy. Thus, by adjusting the laser pulse energy, we can control the amount of material ablated into the source and thus decouple the material ablation process from the plasma operation. Second, the ablated material is ejected backward from the sample not isotropically but in a well-defined cone. Our off-line testing shows that the majority of material comes out in a cone within 20° of the normal [10]. By having this welldefined ejection cone, the material can be better directed into the plasma region and will be less likely to be deposited on the walls, particularly around the sample region. Thus, the laser allows us to better control the amount of material that's injected into the source and allows the material to be more efficiently used by the plasma, while producing less wall contamination. These effects can help reduce cross talk in the ion source.

To stabilize the laser power, we measure and control the laser energy in real time. A photodiode positioned after the first IR mirror (Fig. 4) measures a small portion, < 1%, of the laser intensity. This signal is calibrated to the full laser energy and read through a digitizer. The measured signal then runs in a feedback loop to correct the laser energy and maintain stability to within 2% [11].

In order to maintain beam production stability, the laser beam is continually rastered across the sample surface in a circular pattern. Moving the laser continuously helps prevent the laser from drilling craters in the sample surface which reduce the ablation efficiency [11,12]. In order to raster the beam, two piezoelectric actuators are attached to the mirror on the small optical platform as shown in Fig. 4. The combination of laser energy correction and rastering the laser beam allows us to improve the stability of the ablated beam current although inhomogeneities in the sample also cause beam current fluctuations.

The changes in laser operation and rastering significantly improved the stability of beam current, but even so there is typically a factor of 3–4 exponential fall off in the initial beam current during the first 10 min. After that period beam current stability varies depending on the sample. By counting the rate of actinides at the detector we can correct for the fall off in current over the period of the measurement which for these cases was usually less than 5 min. Measuring the 'mother' material beam current both before and after each run allowed us to compare those values with the observed actinide rate and compute an 'average' current for the run. For many of our cases, the current fluctuations were around 10–20% and were monitored and corrected as described. The current fluctuations certainly folds into our overall uncertainty for a measurement.

The cross talk with the laser was gauged by measuring the actinide

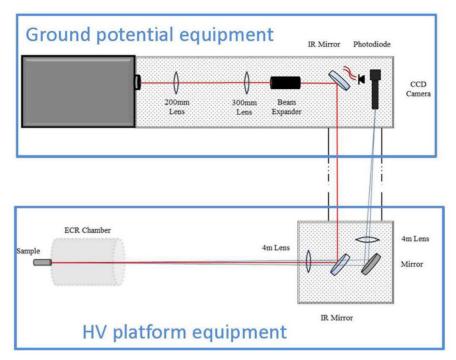


Fig. 4. The optics used for focusing and aligning the laser. A large optical bench off the high voltage platform collimates and expands the beam and a smaller optical bench on the platform redirects and focuses the beam onto target.

count rate was measured with and without the laser on. While laser ablation does not completely eliminate cross talk, it has demonstrated the ability to produce enA of actinide beams from <1% actinide samples while still maintaining low cross contamination between samples.

The total system efficiency was measured for a number of actinides. The average source efficiency was 2.2×10^{-8} . This is quite a bit lower than the efficiency seen for medium mass nuclei $(^{147}\mathrm{Sm}^{31}{}^+)$ which was approximately 4×10^{-5} . About a factor of 5 of this difference can be understood as due to the need to run at high charge states to match the acceleration requirements of the ATLAS radio frequency quadrupole (RFQ) which was not able to run at higher voltages which would allow the use of lower charge states.

2.4. Actinide counting at the fragment mass analyzer (FMA)

The final isotope identification is performed at the FMA. Although the accelerator itself is capable of distinguishing M/Q to within a part in 400, there are still many contaminants that come from the ion source. A schematic of the FMA is shown in Fig. 5.

The FMA uses a combination of two electric dipoles and a magnetic dipole to separate incoming ions based on their mass to charge ratio rather than their magnetic rigidity. At the focal plane of the FMA, ions are linearly dispersed horizontally by their (M/Q) compared to the FMA central value setting. The FMA has a mass resolving power of 340. At

the focal plane of the FMA a Parallel Grid Avalanche Counter (PGAC) measures the ions position on the focal plane, a measure of M/Q, and energy loss rates. A gas-filled ionization chamber measures the residual ion energy. The energy measurements help separate the actinide material from the lower mass contaminants. The FMA's mass resolving power and ability to separate heavy ions, including the actinides, makes it well suited to perform mass identification.

However, the FMA's mass resolving power is not sufficient to separate all (M/Q) degeneracies from the accelerator. In order to provide additional separation, a 50 µg/cm² carbon stripper foil is placed at the entrance to the FMA, and disperses the incoming beam into a variety of charge states. Once the beam is stripped, the lower mass contaminant's alternative charge states will not match that of the actinide. This principle is demonstrated in Fig. 6. The left spectrum shows a plot of residual energy in the ionization chamber versus focal plane position, measured by the parallel grid avalanche counter (PGAC). This spectrum is taken after attenuated of the beam by a factor of 106. The actinide ions, in this case ²³⁷Np, are expected to be separated by energy loss from the lower mass lead and mercury contaminants. However the backgrounds are orders of magnitude more intense making the actinide difficult to detect. These background cannot be separated from the actinide in the original accelerator charge state because the (M/Q) match is too similar. However, in the alternative charge states, created by the stripper, the backgrounds are physically separated from the Np. A set of Ta slits are now moved into the focal plane to block out

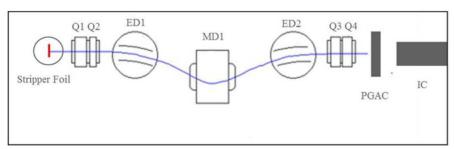


Fig. 5. A schematic of the FMA. The ions are separated based on their mass to charge ratio and are physically dispersed along the focal plane. Qi are quadrupole focusing magnets, ED1 & 2 are electrostatic dipoles, MD1 is a magnetic dipole. PGAC is the parallel grid avalanche counter, a position sensitive detector used for this measurement. IC is s gas-filled ionization chamber for measured the residual energy of the ions.

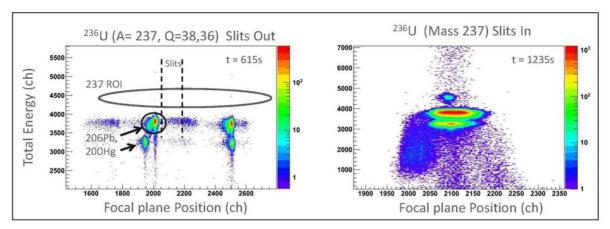


Fig. 6. A plot of residual energy (identified as total energy) versus horizontal position (M/Q) showing the dispersion of multiple charge states on the FMA focal plane. The scale for both axes is arbitrary channel number. The color code scale is number of events in each channel (pixel). In the left plot the slits are open accepting into the detector lead ions with two difference charge states (31 and 32) as well as mercury ions. In all cases the higher mass ions are separated by energy loss because they deposit more energy into the detector. The right plot is taken with the Ta slits closed to the point indicated by the vertical dashed lines and thus cutting out the dominant background lead and mercury species, allowing attenuation to be removed and the A = 237 ions to become visible as the upper small blob. Note that the right plot is expanded horizontally compared to the left plot. The time for each run is indicated in the upper right of each plot.

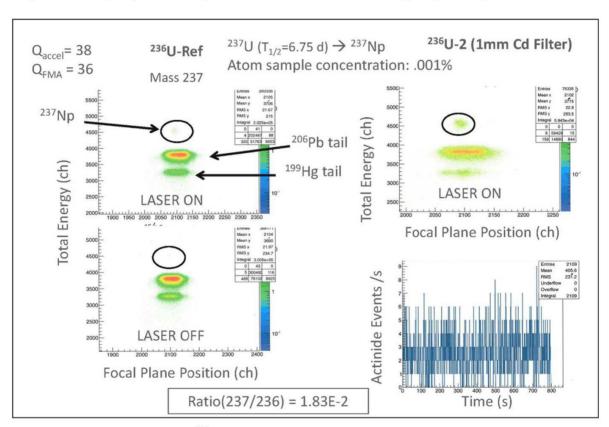


Fig. 7. Spectra obtained for 1-neutron capture case on 236 U. The left spectra show the level of A = 237 actinides in the un-irradiated samples with the laser on and with the laser off. The right spectrum shows the results for the irradiated sample. In all cases the run time was approximately 15 min, the actual run time and rate can be determined from the 1-D time spectrum for the irradiated case in the lower right of the figure.

the backgrounds and the attenuation can be removed, shown in right side of Fig. 6.

The tantalum slits successfully block out the lower mass contaminants and the Np peak is clearly visible. The remaining lower mass contaminants are due to charge exchange within the FMA. This method provides clear separation and unambiguous mass identification for non-actinide contaminants.

3. Example results of measurements

First measurements of irradiated samples were performed at ATLAS in March 2015 and a second run occurred in November 2016 using the system described above. Measure-ments were made of the one and two neutron capture cross sections in most of the seven actinide materials identified in the earlier in this paper. The full results are currently under analysis in collaboration with INL and will be presented at a later time.

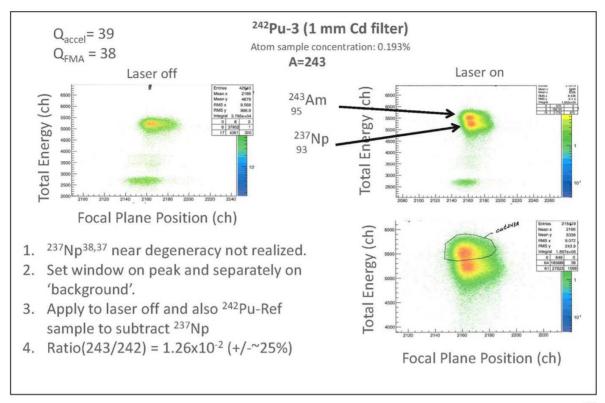


Fig. 8. Spectra from a much more difficult case. In this case, the goal is to measure the rate of A = 243 atoms. But there is a nearby (in M/Q) actinide, $^{237}Np^{+38,+37}$, that contaminates the region and must be subtracted. In this case, we use the laser-off spectrum to establish a background rate in the region of interest shown in the lower right spectrum.

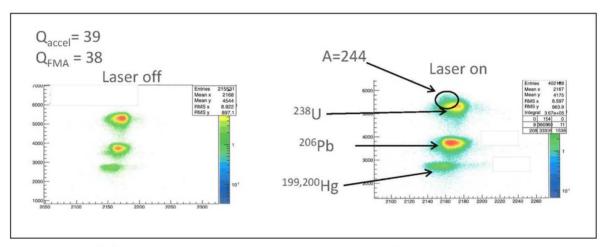


Fig. 9. The same sample as for Fig. 8, but here the system is set for A = 244. Because the concentration of the species of interest is around 1000 times less intense, the background for the nearby $^{238}U^{+38,+37}$ is even more difficult to subtract. The techniques used are similar to that discussed in for A = 243, The preliminary results for this measurement are Ratio (244/242) = 9.4×10^{-4} ($\pm 30\%$).

Two examples will suffice to give a sense of the possibility for this technique. First Figs. 6 and 7 show an example of a clean and thus relatively easy case for A=237, one neutron capture on 236 U. In this case there are no nearby actinides in (M/Q) space and the degenerate lighter nuclei are easy to separate from the difference in (M/Q) and by their energy loss in the PGAC detector and total residual energy in the IC. In addition, the baseline spectrum in the left of Fig. 7 shows the unirradiated material has very little A-237 actinide in the spectrum.

In all cases for MANTRA, the irradiated and unirradiated material has been heavily diluted into an Al_2O_3 matrix. In the case of ^{236}U , the sample material was 0.001% of the total material by atom count. That

resulted in very low 'mother' material beam currents and, we believe, contributed to the current high measurement uncertainties. In this case, the A=236 beam current at the FMA detector was 55 electrical picoamperes (epA).

The second example is for a case with a more difficult actinide background. This is shown in Figs. 8 and 9 and is for the cases of 1 and 2 neutron capture on ²⁴²Pu 'mother' material. Here, in both cases, the problem is from a nearly degenerate actinide and we do not have sufficient energy or delta energy resolution to cleanly separate the two species in the FMA detectors. This situation was due to not initially realizing the degeneracy from these other actinides and running

samples that contained these nuclei as 'mother' material. Thus this is a measure of the 'residual' sample crosstalk that still exists in our system even with the use of laser ablation. To establish a rate of background in the region of interest we chose to use spectra taken with the laser 'off'. Similar backgrounds information is also obtained from spectra taken with the unirradiated 'mother' material. In these two cases there was sufficient resolution and background quality to extract rates for the mass of interest, but significant uncertainties resulted in the present data (~ 25 –30%).

These ratios have been measured with ICPMS using two different ICPMS devices and the preliminary results for the ICPMS are within about 30% of our measurements. Thus the agreement is not good. As we solve some of our AMS uncertainties, we will reassess this comparison.

4. Conclusion

We have developed a new AMS technique at ATLAS to measured isotopic ratios in irradiated actinide samples. Modifications at ATLAS including a centralized clock program to rapidly scale the accelerator, a 20-sample changer to avoid source disruptions, a picosecond laser ablation system for injection of solid material into the ion source, and a highly sensitive Faraday cup at the FMA focal plane to allow beam current measurements and reduce the sensitivity to variations in transmission. This has allowed an unprecedented number of sample measurements in a small amount of allotted beamtime. Measurements on neutron-irradiated actinide samples have been made in two runs over the past two years at ATLAS.

The dominant problem that we have experienced with the measurements to date is repeatability of the measurements. We are exploring a number possible explanations, but at this point we believe a large component of the problem is the very low beam currents that the samples are producing with this system and the low actinide concentration (typically 0.1 to 0.2%) with which each sample is made. The currents for the 'mother' material are generally in the few electrical picoamperes and thus require an very sensitive system with no offsets (or fully understood offsets), thus a very challenging measurement. We are looking into this question more and hope to make additional measurements to better understand this problem and complete the more sensitive channel measurements in the future.

If we can solve our problems with repeatability and understand beam current measurements, then we must also study the possibility of chemical fractionation in the source. That is the isotopes measured for (n+1), (n+2), etc. are not the same element as the mother material. Beta decay has occurred and so the elements are generally a higher Z than the original material. Thus, the charge state distribution for the 'mother' material may be slightly different than the elements detected in measuring the rates of creation for the capture channels. Some information exists which indicate that this is a small correction, but it will need to be addressed later if we are able to reduce our uncertainties that currently dominate the measurements.

This material is based upon work supported by the U.S. Department of Energy, Office of Science, Office of Nuclear Physics, under contract number DE-AC02-06CH11357. This research used resources of ANL's ATLAS facility, which is a DOE Office of Science User Facility.

References

- [1] G. Ferlay, J.-P. Dancausse, P. Leveque, S. Eymard, D. Carre, P. Bienvenu, C. Guy, C. Gautier, H. Isnard, F. Goutelard, P. Bourdot, L. Boucher, J.-M. Bonnerot, IOP Conf. Series: Mater. Sci. Eng. 9 (2010) 012021 available at iopscience.iop.org.
- [2] M. Paul, D. Berkovits, I. Ahmad, F. Borasi, J. Caggiano, C. Davids, J. Greene, B. Harss, A. Heinz, D. Henderson, W. Henning, C. Jiang, R. Pardo, K. Rehm, R. Rejoub, D. Seweryniak, A. Sonzogni, J. Uusitalo, R. Vondrasek, Nucl. Instr. Meth. Phys. Res. B 172 (2000) 688.
- [3] M. Paul, R. Pardo, I. Ahmed, J. Greene, D.J. Henderson, R. Janssens, C. Jiang, K. Rehm, R. Scott, D. Seweryniak, R. Vondrasek, Nucl. Instr. Meth. Phys. Res. B 294 (2013) 165.
- [4] C.N. Davids, B.B. Back, K. Bindra, D.J. Henderson, W. Kutschera, T. Lauritsen, Y. Nagame, P. Sugathan, A.V. Ramayya, W.B. Walters, Nucl. Instr. and Meth. in Phys. Res. B 70 (1992) 358.
- Keithley, https://www.tek.com/keithley-low-level-sensitive-and-specialty-instruments/keithley-series-6400-picoammeters.
- [6] R. Vondrasek, T. Palchan, R. Pardo, C. Peters, M. Power, R. Scott, 'A Multi-Sample Changer Coupled to an ECR Source for AMS Experiments", ECRIS2012: JACOW Proceedings of the 20th International Workshop on ECR Ion Sources, WEPP17, 146(2012), http://accelconf.web.cern.ch/AccelConf/ECRIS2012/papers/wepp17. pdf.
- [7] R. Harkewicz, J. Stacy, J. Greene, R. Pardo, Rev. Sci. Instrum 65 (1994) 1104.
- [8] E. Gamaly, Phy. Reports 508 (2011) 91.
- [9] A.L. Gray, Analyst 110 (1985) 551.
- [10] T. Palchan, R. Pardo, F. Kondev, S. Kondrashev, C. Nair, R. Scott, R. Vondrasek, M. Paul, W. Bauder, P. Collon, G. Youinou, M. Salvatores, G. Palmiotti, J. Berg, T. Maddock, G. Imel, Laser Ablation of Actinides into an Electron Cyclotron Resonance Ion Sources for Accelerator Mass Spectroscopy, in: JACoW ECRIS2012: Proceedings of the 20th International Workshop on ECR Ion Sources, FRXA03, 190(2012), http://accelconf.web.cern.ch/AccelConf/ECRIS2012/html/author.htm.
- [11] R. Scott, T. Palchan, R. Pardo, R. Vondrasek, F. Kondev, O. Nusair, C. Peters, M. Paul, W. Bauder, P. Collon, Rev. Sci. Instrum. 85 (2014).
- [12] O.V. Borisov, X. Mao, R.E. Russo, Spectrochim. Acta, Part B 55 (2000) 1693.