PLANS FOR LASER ABLATION OF ACTINIDES INTO AN ECRIS FOR ACCELERATOR MASS SPECTROSCOPY*

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Abstract

A project using Accelerator Mass Spectrometry (AMS) at the ATLAS facility to measure neutron capture rates on a wide range of actinides in a reactor environment is underway. This project will require the measurement of many samples with high precision and accuracy. The AMS technique at ATLAS is based on production of highly-charged positive ions in an electron cyclotron resonance ion source (ECRIS) followed by linear acceleration. We have chosen to use laser ablation as the best means of feeding the actinide material into the ion source because we believe this technique will have more efficiency and lower chamber contamination thus reducing 'cross talk' between samples. In addition construction of a new multi-sample holder/changer to allow quick change between multiple samples is part of the project. The status of the project, design, and goals for initial off-line ablation tests will be discussed as well as the overall project schedule.

INTRODUCTION

Advanced nuclear fuel cycles are currently under evaluation in order to assess their potential to cope with new requirements of radioactive waste minimization, optimization of resource utilization, and reduced risk of proliferation. This assessment should account for several key features of the fuel cycle, including irradiated fuel processing, innovative fuel development and fabrication, waste characterization, and disposal. In some cases, the impact of nuclear data and their associated uncertainties can be crucial in order to further explore an option, or to reject it. The need for accurate data has been pointed out in recent studies devoted to Generation-IV systems, see e.g. [1]. The very high mass actinides can play a significant role in the feasibility assessment of innovative fuel cycles. As an example, the potential build-up of ^{252}Cf when recycling all transuranics in a light water reactor, leads to increased neutron emissions that could impact the fuel fabrication process. As a consequence, the poorly known nuclear data of higher mass transuranics need to be significantly improved.

At present, there is data to provide some information on the performance of these isotopes in reactor environments, but up to now, there has been little emphasis on the quality of these data and few reliable uncertainty estimates have been provided. This situation is due to the difficulty to make both integral and differential cross section measurements for these isotopes.

The objective of this project is to obtain valuable integral information about neutron cross sections for actinides that are of importance for advanced nuclear fuel cycles in a relatively short time compared to the more standard, and time consuming, route which consists of irradiating samples in a reactor and then performing chemical analysis to characterize the different isotopes produced during irradiation.

The proposed work intends to develop an original approach that takes advantage of two experimental facilities: the neutron irradiation capabilities of the Advanced Test Reactor (ATR) at the Idaho National Laboratory and the Accelerator Mass Spectrometry (AMS) capabilities of the Argonne Tandem Linac Accelerator System (ATLAS)[2] at Argonne National Laboratory.

The novelty of this approach relies on the use of AMS which is expected to provide very sensitive measurements of the production of different actinides that are built up during the irradiation, up to the highest mass isotopes. AMS at ATLAS can detect down to about 10^6 atoms in samples consumed in the ion source, which is out of the range of more classical chemical analysis traditionally used to analyze irradiated fuel samples.

In order to succeed in this project, the work can be decomposed into three major steps:

1. Preparation and irradiation of some pure actinide samples in ATR. The samples that are available at INL and which are of interest for advanced reactor fuel cycles are the following: ^{232}Th, ^{235}U, ^{236}U, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am and ^{248}Cm.

2. Measurements of the amount of the different isotopes produced in the irradiated samples at ATLAS.

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3. Using the results of the measurements, derive information on (n, γ), and (n,2n) cross sections on the target and daughter isotopes. An effort will be made also to derive, if possible, fission yields.

In this report we will focus on the requirements of the AMS program and the novel aspects that will be implemented at the ECR ion source at ATLAS to carry out this research program. The requirements of precision and accuracy to obtain useful results are discussed more fully in [2]

AMS AT ATLAS

The principle of AMS at ATLAS [4] differs from conventional tandem-accelerator AMS by the injection of highly-charged positive ions followed by their linear acceleration. In this scheme, the high-velocity electrons of the ECRIS plasma strip the ions to high charge states, a process in some sense inverse to the stripping of fast ions in a solid or gas target used in a tandem accelerator. Similarly, the ECRIS ion stripping dissociates any existing molecular ions, an essential feature of AMS.

The requirements placed on the AMS measurements to be performed at ATLAS are quite challenging. These challenges include high-precision isotope ratio measurements, minimization of cross-talk between samples, efficient use of milligram samples, and the processing of an unprecedented number of samples for a facility as complex as ATLAS. Unique element (Z) identification is desirable, but is not expected to be possible except for specific cases.

The measurement configuration for ATLAS will use the ECR-II ion source, significantly modified as discussed below, as the source of ions. After acceleration and deceleration (increasing the accelerator m/q resolution but keeping the ion energy within acceptance range of analytical elements) in the ATLAS linac to approximately 1 MeV/u, the actinide ions of interest will be counted in the focal plane of the Fragment Mass Analyzer (FMA). The major challenges are discussed in the succeeding paragraphs.

Precision Requirements

Only ^{14}C AMS has consistently achieved precision levels in the <1% regime. This has been achieved in dedicated facilities, principally by the fast cycling of the accelerator setup between the rare isotope (^{14}C) and a normalizing abundant isotope (^{13}C). Except for specific cases, counting statistics are not the problem. Knowing the transmission between the measurement point for the stable reference and the detector of the AMS isotope is often the limiting parameter for uncertainty in the final result. This problem will be tackled by developing an improved automation of the accelerator scaling required to measure the various isotopes or ion species and an automated sample changer to allow rapid changes between various samples. In many cases of interest, the measurement time for each sample will be only a few minutes for adequate measurement statistics. But the lack of known absolute standards, the possibility of cross-talk between samples, and instabilities in the accelerator are the significant problems that must be addressed to achieve the precision goals.

A recent AMS experiment on ^{146}Sm [5] has required that we develop an approach to measuring the absolute transmission of the accelerator and detector system and has highlighted the need to run ATLAS is an extremely stable mode. It has also demonstrated that it is feasible to switch rapidly (by computer control of the machine components' setup) between a rare and abundant isotope (in a way similar to that used for ^{14}C), thus improving control over the accelerator transmission. Because of this ongoing work and the additional techniques discussed here, we believe we can achieve the required stability and characterization of the transmission in order to enter this regime of precision.

Small Sample Size and Cross-Talk

A major feature of AMS is the ability to analyze small samples. At ATLAS the AMS activities always are focused on samples of a few milligrams. For this project, it is important that we deal with many small samples. The smaller the samples, the less are the radiological problems associated with handling α-emitting actinides for ATLAS operation. The need to measure many small samples as quickly as possible pushes us to develop efficient sample changing techniques for the ECR source and material delivery techniques which minimize source contamination.

We believe the best approach for this situation is to develop laser ablation for the feeding of sample material into the source. With laser ablation, a very small and controllable amount of sample material can be introduced into the source without introduction of extraneous material from the sample holder. Also the distribution of emission of ablated material in laser irradiation tends to obey a cos θ law which is expected to improve the efficiency of capture of ions into the plasma and thereby reduce wall contamination. Finally, the form of the sample material (metal, oxide, etc) is less critical than with the sputtering or oven technique.

The ECR-II source will also be equipped with a quartz liner. The quartz liner will keep the main body of the source relatively clean of actinides, thus simplifying cleanup. Furthermore, there is some operational evidence that cross talk among samples is reduced. This effect has been observed with other AMS projects at ATLAS. A negative to using a quartz liner is that source performance as measured by charge-state distribution and maximum beam intensity is somewhat reduced. But the beam energy is limited by the bending power of the FMA system and use of high charge state ions is not required. A mass-to-charge ratio of ~8-9 should be quite adequate for these measurements.

Ion identification and counting

One of the major challenges of actinide AMS at ATLAS is the need for separation or discrimination of a desired species from backgrounds of ions having similar mass-to-charge ratios present as chemical impurities or interference of ion-source materials. This discrimination is best achieved at ATLAS by using the FMA which has a very large dispersive power and can analyze very heavy ions. A proof of concept and first measurements of rare species in the actinide region were successfully performed [4] (since this experiment considerable improvement has been made in the focal-plane detector of the FMA). The typical setup consists in accelerating and decelerating a desired ion so that its energy matches the electric-rigidity acceptance of the FMA. Stripping at the FMA target position (the only one along the machine) allows the heavy ions to reach the high charge state needed for magnetic-rigidity acceptance of the FMA.

In the case of actinide nuclei, Z identification at the energies compatible with the FMA acceptance is likely to be impossible. However, the high redundancy of energy, energy-loss and time-of-flight signals available from that detector is expected to contribute to the mass-to-charge determination and to a high level of background discrimination.

LASER AND SOURCE CONFIGURATION

Laser Parameters and Configuration

Laser ablation into an ECR source was first developed at ATLAS [6] and used as a plasma diagnostic tool[7] and has since been used by a number of other labs to explore the coupling of laser produced ions into an ECR source. The technique has not been used routinely for ion production and will require development for this application. The controlled release of materials into the plasma by well-focused laser light will eliminate the significant material buildup often seen in the region of the oven throat or beside the sputter cones, two techniques widely used for sample feeding to the ECRIS. This inefficient, indiscriminate injection of material into the source not only reduces the overall sensitivity of the method but is a major source of cross-contamination between samples. Our experience with lasers in the past indicates that the laser ablation approach will be much cleaner, but must be shown to work for this application.

In this application we plan to use an axial geometry. The laser beam will be brought into the source through the extraction aperture as shown in Fig. 1. The properties of the laser to be used in this application are:

- λ =1064 nm
- ≤1011 W/cm2
- 8 ps pulse width
- Rep Rate up to 400 Hz
- Laser beam size ~7 mm maximum
- less than 1 mm diameter spot on sample
- Pulse energy: variable, up to 10 mJ/p.

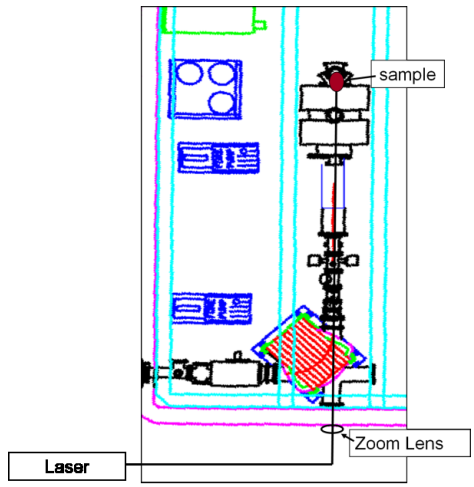

Figure 1: Axial geometry of laser ablation into the ATLAS ECR-II.

This project has an unprecedented number of samples that need to be characterized. Typically AMS experiments at ATLAS have needed to process less than 10 samples for any one experiment. This project envisions sample numbers of around 50 or more. An entirely new approach to sample processing is required. In addition, the desire to rapidly switch between measuring an irradiated sample and the original sample requires a different approach.

We have designed a multi-sample changer that will be installed in the injection side of the ECR ion source. This changer, shown in Fig. 2, can accept 20 samples and will be able to alternate between the various samples without breaking vacuum. The change time will be only a few seconds.

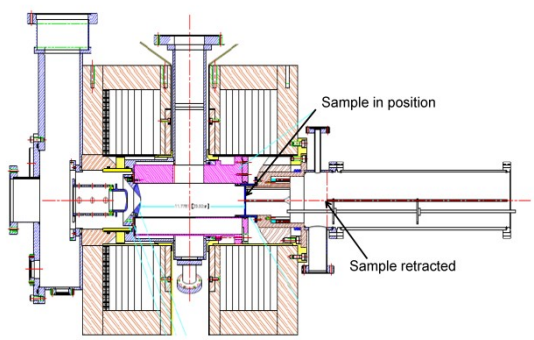

Figure 2: ECR-II cross-section with multi-sample changer shown

The laser is expected to be delivered in the Fall of 2010. Off-line ablation measurements will be made in late 2010 and installed at the ECR source by the Spring of 2011. Initial measurements on reference samples will be made in late 2010 to study backgrounds and

establish Z sensitivity of the detectors. The multisample changer will be ready for installation by the summer of 2011 and initial samples will be ready for measurement in late 2011.

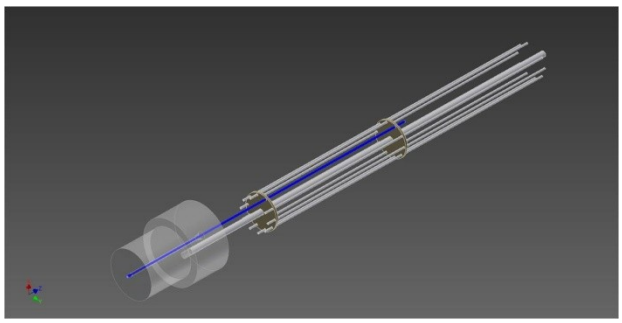

Figure 3: 3-D perspective of multi-sample changer assembly. The samples are pre-mounted on individual rods which can be rotated into position and inserted in the ECR ion source.

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