INL/CON-10-18471
PREPRINT

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International Conference on Nuclear Data for Science and Technology 2010

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April 2010

AN INTEGRAL REACTOR PHYSICS EXPERIMENT TO INFER ACTINIDE CAPTURE CROSS-SECTIONS FROM THORIUM TO CALIFORNIUM WITH ACCELERATOR MASS SPECTROMETRY

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The principle of the proposed experiment is to irradiate very pure actinide samples in the Advanced Test Reactor at INL and, after a given time, determine the amount of the different transmutation products. The determination of the nuclide densities before and after neutron irradiation will allow inference of effective neutron capture cross-sections. This approach has been used in the past and the novelty of this experiment is that the atom densities of the different transmutation products will be determined using the Accelerator Mass Spectrometry technique at the ATLAS facility located at ANL. It is currently planned to irradiate the following isotopes: ^{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.

KEYWORDS : Integral Experiment, Actinide Capture Cross-Sections, AMS, ATR

1. INTRODUCTION

This paper presents an on-going effort at INL/ISU/ANL to perform an integral reactor physics experiment whose objective is to infer actinide neutron capture cross-sections. 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 TRU in a LWR, leads to increased neutron emissions that could impact the fuel fabrication process. As a consequence, higher mass transuranics poorly known nuclear data should be significantly improved.

The principle of this experiment is to irradiate very pure samples in the Advanced Test Reactor (ATR) at INL and, after a given time, determine the amount of the different transmutation products. The determination of the nuclide densities before and after neutron irradiation will allow inference of energy-integrated neutron cross-sections, i.e. $\int_0^\infty \sigma(E)\varphi(E)dE$, where $\varphi(E)$ is the neutron flux "seen" by the sample.

This approach has been used in the past and the novelty of this experiment is that the atom densities of the different transmutation products will be determined using the Accelerator Mass Spectrometry (AMS) technique at the ATLAS facility [2] located at ANL. This technique is sensitive for measuring quantities of long-lived, rare isotopes with high discrimination in the presence of more abundant ones using very small amounts of material. While AMS facilities traditionally have been limited to the assay of low-to-medium atomic mass materials, i.e., A < 100, there has been recent progress in extending AMS to heavier isotopes – even to A > 200. The detection limit of AMS is orders of magnitude lower than that of standard mass spectrometry techniques (abundances as low as 10^{-12} can be detected), thus allowing more transmutation products to be measured and consequently more neutron cross-sections to be inferred from a single sample.

2. SAMPLES IRRADIATION IN THE ATR

It is currently planned to irradiate the following isotopes: ^{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. These enriched isotopes are available at INL and their precise characterization is underway. Only a few mg of each material will be needed in order to prepare the experimental samples.

The ATR is a 250 MWth water-cooled reactor, with a thermal neutron spectrum. Using neutron filters (cadmium and boron) it is however possible to modify the spectrum and thus meet specific needs. A MCNP model was used to calculate the effective one-group cross-sections and flux levels in the samples that will be inserted in large B positions in the beryllium reflector (B9, 10 and 11, see [3]).

The neutron capture reactions on ^{10}B and ^{113}Cd have large cross-sections and strongly impact the neutron spectrum (Fig.1). It is presently planned to irradiate three sets of actinide samples: the first one filtered with cadmium and the other two filtered with enriched ^{10}B of different thicknesses (~ 8 mm and 4 mm).

Furthermore, based on the results of the analysis presented in chapter 3, the cadmium and boron-filtered samples will be

irradiated, respectively, for 50 days, 100 days and 150 days. Loading of the samples in the ATR is scheduled for Feb. 2011.

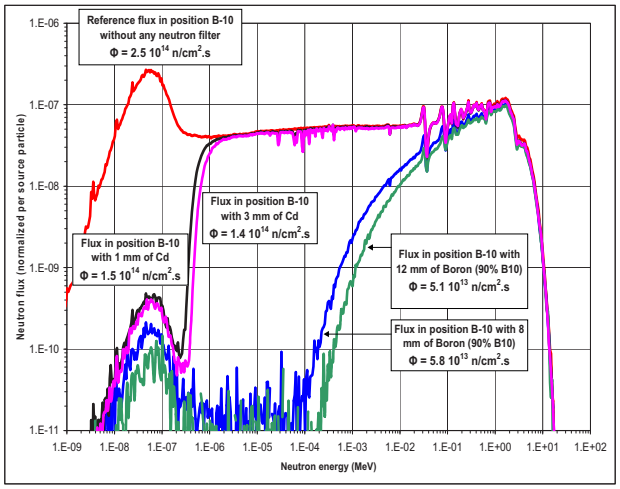

Fig. 1. Neutron flux in the samples with and without filters.

3. UNCERTAINTY ANALYSIS

Let us consider a very pure sample containing essentially (at least 99%) an isotope of mass number A, but also a few tenths of a percent of other isotopes present as impurities (let's say A+1, A+2 and A+3). Using a Taylor development of the solution of the Bateman equations, the atom density of isotope A+1 after an irradiation time T can be expressed as [3]:

$$N_{A+1}(T) \sim N_A(0)\,\bar{\sigma}_A^c[\bar{\phi}\,T] + N_{A+1}(0)\,\{1-\tilde{\sigma}_{A+1}^a[\bar{\phi}\,T]\}$$

where $\bar{\sigma}_A^c = \dfrac{\int \sigma_A^c(E)\,\varphi(E)\,dE}{\int \varphi(E)\,dE}$ is the effective neutron capture cross-section and $\tilde{\sigma}_{A+1}^a = \bar{\sigma}_{A+1}^c + \bar{\sigma}_{A+1}^f + \dfrac{\lambda_{A+1}}{\bar{\phi}}$ takes into account the neutron absorption (essentially capture and fission) as well as natural decay. Since the measurements will provide the time-integrated neutron flux $[\bar{\phi}\,T]$ as well as the relative atom densities $[\dfrac{N_{A+1}(t)}{N_A(t)}]_m \equiv [R_{A+1}(t)]_m$ both at t = 0 and t = T, the capture cross-section of A can then be inferred as:

$$\bar{\sigma}_A^c \sim \frac{[R_{A+1}(T)]_m\{1-\bar{\sigma}_A^f\,[\bar{\phi}\,T]_m - \lambda_A[T]_m\} - [R_{A+1}(0)]_m\{1-\tilde{\sigma}_{A+1}^a[\bar{\phi}\,T]_m\}}{[\bar{\phi}\,T]_m\left(1+[R_{A+1}(T)]_m\right)}$$

The time-integrated neutron flux $\int_0^T \phi(t)\,dt \equiv \bar{\phi}\,T$ can be determined experimentally from the amount of a fission product, ^{148}Nd, formed during irradiation in ^{235}U samples (well-known fission yield and fission cross-section) as:

$$\bar{\phi}\,T \sim [\frac{N_{Nd8}(T)}{N_{U5}(T)}]_m \frac{1}{\gamma_{Nd8}\,\bar{\sigma}_{U5}^f} \equiv [R_{Nd8}(T)]_m \frac{1}{\gamma_{Nd8}\,\bar{\sigma}_{U5}^f}$$

Using these expressions as well as the recommended, and widely accepted, laws of propagation of uncertainties, it is possible to estimate the experimental uncertainties that would allow the inferred effective cross-sections to be determined with a given target uncertainty, for instance 5%.

Let's consider the case of the capture cross-section of A. If the relative uncertainty of the measured A+1 relative atom densities in the initial as well as in the irradiated samples are the same, i.e. if $\dfrac{u([R_{A+1}(0)]_m)}{[R_{A+1}(0)]_m} = \dfrac{u([R_{A+1}(T)]_m)}{[R_{A+1}(T)]_m} \equiv \dfrac{u([R]_m)}{[R]_m}$, then it can be expressed as:

$$\frac{u([R]_m)}{[R]_m} \sim \frac{\sqrt{\left(\dfrac{u(\bar{\sigma}_A^c)}{\bar{\sigma}_A^c}\right)^2 - \left(\dfrac{u([\bar{\phi}\,T]_m)}{[\bar{\phi}\,T]_m}\right)^2}}{\left(\dfrac{\sqrt{[R_{A+1}(T)]_m^2 + [R_{A+1}(0)]_m^2}}{[R_{A+1}(T)]_m - [R_{A+1}(0)]_m}\right)},$$

with the uncertainty on the time-integrated neutron flux given by:

$$\frac{u([\bar{\phi}\,T]_m)}{[\bar{\phi}\,T]_m} \sim \sqrt{\left(\frac{u([R_{Nd8}(T)]_m)}{[R_{Nd8}(T)]_m}\right)^2 + \left(\frac{u(\gamma_{Nd8})}{\gamma_{Nd8}}\right)^2 + \left(\frac{u(\bar{\sigma}_{U5}^f)}{\bar{\sigma}_{U5}^f}\right)^2}$$

Similar expressions have been derived for the A+1 and A+2 capture cross-sections but are not reproduced here because they are too cumbersome. They can be found in Ref [3].

Though it might seem obvious, this expression shows that the smaller the difference, $[R_{A+1}(T)]_m - [R_{A+1}(0)]_m$, i.e. the smaller the term $\bar{\sigma}_A^c[\bar{\phi}\,T]$, the smaller the measurement uncertainty should be for a given target cross-section uncertainty. The same thing will apply to the inference of the capture cross-sections of the A+1, A+2… isotopes and is an issue especially in the hard boron-filtered spectrum where the capture cross-sections and the neutron flux are both small (~10^{-24} cm^{-2} and 5×10^{13} n.cm^{-2}.s^{-1}).

This is particularly important since, even though a very sensitive technique, the precision of AMS is limited to a few percents (2-3% anticipated for this specific application). Hence, in order to loosen the constraints on the required experimental uncertainties and, thus, take advantage of the high sensitivity of AMS, (1) the terms $\bar{\sigma}^c[\bar{\phi}\,T]$ should be maximized and (2) the initial level of A+1, A+2… impurities should be made as small as possible.

For a given neutron filter, condition (1) implies irradiation of the samples as long as possible within the reactor's operational constraints and provided that the atom densities stay away from saturation. These considerations led us to choose irradiation times for cadmium and boron-filtered samples of,

respectively, 50 days, 100 days and 150 days.

Condition (2) is most readily satisfied when A is the heavier long-lived isotope of an element because then the initial A+1, A+2… impurities belong to another element and are consequently easier to separate than when they belong to the same element as A. This category of isotopes includes ^{232}Th, ^{238}U, ^{237}Np, ^{242}Pu, ^{243}Am and ^{248}Cm and are given the highest priority for the AMS analysis because its 2-3% precision should be sufficient to infer cross-sections with an uncertainty of about 5%. The other isotopes, with a more important A+1, A+2… contamination, will be analyzed with the less sensitive but more precise methods available at INL (TIMS and ICP-MS), hence, providing another set of data.

Table 1, 2 and 3 show the calculated actinide build-up in ^{238}U, ^{242}Pu and ^{248}Cm samples (supposed to be 100% pure) filtered with cadmium and boron. Since the AMS can detect actinide abundances down to 10^{-12}, these nuclides could potentially be measured if the initial impurities are low enough that they don't mask the actual build-up. The purity of the samples will be essential for the success of the experiment.

Table 1. Example of actinides build-up in a U^{238} sample after 50 days with a cadmium filter and 200 days with a boron filter (U8 refers to ^{238}U, with similar abbreviations elsewhere)

	U8	Pu9	Pu0	Pu1	Pu2	Am3	Cm2
Cd	0.991	8E-03	2E-05	2E-06	2E-09	2E-11	1E-11
B	0.999	2E-04	7E-08	2E-11	-	-	-

Table 2. Example of actinides build-up in a ^{242}Pu sample after 50 days with a cadmium filter and 200 days with a boron filter

	Pu2	Am3	Cm4	Cm5	Cm6	Cm7
Cd	0.964	3E-02	9E-04	6E-06	5E-09	4E-12
B	0.999	3E-04	1E-07	2E-11	-	-

Table 3. Example of actinides build-up in a ^{248}Cm sample after 50 days with a cadmium filter and 200 days with a boron filter

	Cm8	Bk9	Cf9	Cf0	Cf1	Cf2
Cd	0.992	7E-03	4E-04	1E-04	8E-06	7E-08
B	0.999	1E-04	3E-05	2E-07	1E-10	-

4. EXPERIMENTAL DETERMINATION OF THE ATOM DENSITIES

4.1 Accelerator Mass Spectrometry at ANL

The requirements placed on the AMS measurements to be performed at ATLAS by this program are quite challenging. Challenges to a successful program include high-precision 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. The measurement configuration for ATLAS will use the Electron Cyclotron Resonance ion source, ECR-II, significantly modified as discussed below, as the source of ions. After acceleration and deceleration 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 by use of dedicated facilities, automated sample changes between unknown samples and absolute calibration standards, and principally by the fast cycling of the accelerator setup between the rare isotope (^{14}C) and a normalizing abundant isotope (^{13}C). Generally, spectrum 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 the final result uncertainty. This problem will be tackled by developing an automated sample changer to allow rapid, automatic changes between various samples and improved automation of the accelerator scaling required to measure the various isotopes required. 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 [4] 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 Between Samples. A major feature of AMS is the ability to analyze small samples. At ATLAS the AMS activities always are focused on samples of only 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 for ATLAS operation. The need to measure many small samples as quickly as possible pushes us to develop efficient sample feeding techniques for the ECR source.

We believe the best technique for this situation is to develop laser ablation for the feeding of sample material into the source. With laser ablation, a very small amount of sample material can be introduced into the source without introduction of extraneous material from the sample holder. Additionally, 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 AMS of heavy nuclides 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 separation or discrimination is best achieved at ATLAS by using the FMA which has a very large dispersive power and can analyze very heavy ions (in its acceptance region of electrostatic and magnetic rigidities). A proof of concept and first measurements of rare species in the actinide region were successfully performed [5] (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. A stripping process (typically the only one along the machine, improving thus efficiency) 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.

4.2 Methods available at INL

The materials to be irradiated in ATR will be prepared, characterized, and encapsulated at the Materials and Fuels Complex at Idaho National Laboratory. Pre-irradiation samples will be archived for concurrent post-irradiation analysis by AMS at ATLAS. The heaviest isotope of each mass chain will also be analyzed pre-irradiation by AMS at ATLAS.

Once purified, the materials would be precipitated in a form suitable for irradiation in the reactor and for measurement by AMS. As halides present a problem for the ECR source, the likely final form will be an oxide. To facilitate handling of these mg quantities, innocuous filler such as aluminium may be used. Once a solid form has been achieved, the material would be individually loaded and welded into a small stainless steel canister.

Pre-irradiation analysis of the materials at INL will be done using conventional analytical and radiochemical techniques. Isotopic ratios will be measured using both Inductively-Coupled Plasma Quadrupole Mass Spectrometry (ICP-QMS) and Thermal Ionization Mass Spectrometry (TIMS). Elemental contaminations will be measured using an automated gas-pressurized extraction chromatography (GPEC) system. This system has previously been used for measuring uranium and plutonium contamination in americium from dismantled AmBe sources [6].

5. CONCLUSION

The principle of the proposed experiment is to irradiate very pure actinide samples in the Advanced Test Reactor (ATR) at INL and, after a given time, determine the amount of the different transmutation products. The determination of the nuclide densities before and after neutron irradiation will allow inference of effective neutron capture cross-sections.

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6. ACKNOWLEDGMENTS

This work is supported by the U.S. Department of Energy, Office of Science, Office of Nuclear Physics, under DOE Idaho Operations Office Contract DE-AC07-05ID14517 as well as by the ANL Contract DE-AC02-06CH11357 and by the ATR National Scientific User Facility. The help provided by Antoine Petiot, summer intern from the École des Mines de Paris supervised by Joseph Nielsen from the Irradiation Testing Department at INL, in running the MCNP calculations is gratefully acknowledged. The involvement of Mitchell Meyer, scientific program manager at ATR-NSUF is also gratefully acknowledged.

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