NEUTRON-CAPTURE STUDIES ON ^{235}U AND ^{238}U VIA AMS

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Major nuclear data libraries exhibit some discrepancies for the neutron capture reactions ^{235}U(n,γ) and ^{238}U(n,γ) at keV energies. These differences reflect also the scatter of experimental data that are primarily based on time-of-flight measurements (TOF) with detection of prompt γ-rays. We report here on an independent approach for studying such reactions: Neutron activations with subsequent accelerator mass spectrometry (AMS) measurement represent an independent technique, where interference from fission is completely excluded. Activations of natural uranium samples were performed with cold neutrons (Budapest), thermal (Vienna) and with neutrons of 25 and 500 keV (Karlsruhe) for the measurement of ^{235}U(n,γ) and ^{238}U(n,γ). The produced long-lived ^{236}U and the decay product of ^{239}U, ^{239}Pu, respectively, were subsequently counted by AMS at the Vienna Environmental Research Accelerator. This method for measuring the neutron capture cross section has the advantage that the involved systematic uncertainties are in no way correlated with those inherent to the TOF technique. Preliminary data for these neutron energies indicate good agreement with evaluations and seem to support in the keV energy range the reported lower cross-section values for both capture reactions.

KEYWORDS: neutron capture cross section, ^{235}U, ^{238}U, accelerator mass spectrometry (AMS), nuclear data

1. INTRODUCTION

Improved and highly accurate nuclear data are urgently required for the design of advanced reactor concepts. This demand holds for minor actinides but also for the main fuel materials. Existing data for the capture channel have been measured e.g. by time-of-flight techniques via detection of the prompt capture γ-rays. A major difficulty in these experiments is the safe discrimination against the strong γ-background from the competing fission channel. Some indications exist, that previous data might suffer also from multiple scattering corrections. Moreover, recent studies exhibit some discrepancies at keV energies between major nuclear data libraries for ^{235}U(n,γ) and ^{238}U(n,γ). Both capture reactions are listed in the NEA High Priority Request List [1] and are recommended for re-measurement.

Neutron activation with subsequent accelerator mass spectrometry (AMS) measurement represents a technique, where interference from fission is completely excluded. Within the European EFNUDAT project [2], two neutron activations were performed with the goal to determine the capture cross section of ^{235}U for thermal and keV-neutrons [3]. The use of natural uranium samples allowed us also to measure the ^{238}U(n,γ) capture from the very same samples.

The ^{197}Au(n,γ) cross section is established as standard at thermal energy and between 0.2 and 2.5 MeV. However, at 25 keV a difference of some 6-8 % was found [4] between the standards evaluation and the reference value used for capture studies e.g. for nuclear astrophysics [5,6]. A similar difference for the cross section ratio ^{238}U(n,γ)/^{197}Au(n,γ) was found between data based on TOF and prompt γ-detection. The new data obtained in this work cover also this energy region and deliver precise data for ^{235,238}U(n,γ) again relative to Au; but utilizing an independent technique.

Activations were performed with cold neutrons (Budapest Research Reactor) [7], thermal (Atominstitut, Vienna) [8] and with neutrons of 25 and 500 keV (KIT Karlsruhe) [6], the latter with an energy spread of 20 and 50 keV FWHM, respectively. The produced long-lived ^{236}U and the decay product of ^{239}U, ^{239}Pu, were subsequently counted by AMS at the Vienna Environmental Research Accelerator (VERA) [9]. VERA was recently upgraded to reach high sensitivities also for actinides [10]. This method for measuring the neutron capture cross section has the advantage that the involved systematic uncertainties are in no way correlated with those inherent to the TOF technique. Therefore, this experiment provides important and independent information for these key reactions of reactor physics with uncertainties expected below 5% [11]. The high sensitivity of AMS requires only very small samples.

2. NEUTRON ACTIVATIONS

Fig. 1. Zoom into the chart of nuclides [12] relevant for neutron capture on ^{235}U and ^{238}U. Long-lived ^{236}U ($t_{1/2}$ = 23.4 Myr), and the decay product of ^{239}U ($t_{1/2}$ = 23.5 min) and ^{239}Np ($t_{1/2}$ = 2.36 days), the long-lived ^{239}Pu ($t_{1/2}$ = 24 kyr), were measured by AMS.

Natural uranium samples were used for our project, despite the low concentration of ^{235}U (0.72% in natural uranium). Samples enriched in ^{235}U likely might contain higher concentrations of ^{236}U (e.g. from previous neutron exposure). We used two different types of natural uranium: (1) a material from IRMM (BC02061) [13], specified in its isotopic composition, with a well known stoichiometry (pure U_3O_8), and which was measured to contain low ^{236}U and ^{239}Pu concentrations. (2) for comparison a second material ('MB' - prepared from pre-nuclear uranyl nitrate) was also used for ^{238}U(n,γ).

2.1 Cold neutrons at Budapest Research Reactor

Cold neutrons were produced by means of a neutron guide beamline at the 10 MW Budapest Research Reactor. At two positions two uranium pellets, respectively, were activated under well-defined irradiation conditions [7]. The U_3O_8 powder of typically 50-70 mg was homogeneously mixed with a few mg Au powder and pressed to pellets of 6 mm diameter ('U+Au'). In addition, thin Au foils were placed to form a sandwich of Au–U(+Au)–Au. The ^{197}Au(n,γ) thermal cross section value was used for the determination of the neutron fluence, assuming the same 1/v energy dependence of the capture cross sections from thermal to cold energies for uranium and gold.

The Au foils and the Au powder delivered independent values for the neutron fluence, and confirmed a homogeneous Au powder distribution. The total neutron fluence was between 2.6×10^{12} and 1.4×10^{13} n cm^{-2} at the NIPS station (MB-samples); and 7.2×10^{13} and 2.8×10^{14} n cm^{-2} for BC02061 at a second irradiation position, respectively.

2.2 '25- and 500 keV-neutrons' at FZ Karlsruhe

Samples prepared from the same uranium reference material (BC02061) were used for the neutron activations at Karlsruhe Institute of Technology. Similarly, U_3O_8 powder was pressed into two pellets with a mass of 55 mg. Again, Au foils were used to form a stack of Au-U-Au, for the fluence determination. The activations were carried out at the 3.7 MV Van de Graaff accelerator: Neutrons were produced via the ^{7}Li(p,n) reaction by bombarding a Li target with protons at appropriate energies.

Two different neutron energies were selected. First, a neutron energy distribution peaking at 25 keV was produced. This neutron spectrum closely resembled an energy distribution of a Maxwell-Boltzmann type of 25 keV [6]. 500 keV neutrons with an energy spread of 50 keV FWHM were produced by higher-energetic protons through the same ^{7}Li(p,n) reaction. A proton beam intensity of typically 100 μA resulted in a fluence of 1.75×10^{15} ncm^{-2} (25 keV) and 4.34×10^{15} ncm^{-2} (500 keV), respectively.

Table 1 and 2 list the various samples irradiated at Budapest and KIT together with the neutron fluences. For an estimation of the expected production of ^{236}U and ^{239}U, ENDF/B-VII cross-section values were used.

Table 1 Isotope ratios ^{236}U/^{238}U produced at IKI (cold neutrons) and KIT (25 and 500 keV) using ENDF cross section values for ^{235}U(n,γ). [1)]calculated for ^{nat}U (0.72% ^{235}U); [2)]calculated for 25 keV Maxwell-Boltzmann distribution

sample	irradiation	fluence [n cm^{-2}]	cross-sec. [barn]	isotope ratio ^{236}U/^{238}U[1)]
IRMM-3	IKI	7.15×10^{13}	98.96	5.1×10^{-11}
IRMM-5	IKI	2.78×10^{14}	98.96	2.0×10^{-10}
MB-1	IKI	2.55×10^{12}	98.96	1.8×10^{-12}
MB-2	IKI	1.37×10^{13}	98.96	9.8×10^{-12}
IRMM-a	FZK-25	1.75×10^{15}	0.690[2)]	8.7×10^{-12}
IRMM-b	FZK-500	4.34×10^{15}	0.163	5.1×10^{-12}

Table 2 Calculated (ENDF/B-VII) number of ^{239}U (= ^{239}Pu) atoms produced at IKI (cold neutrons) and KIT (25 and 500 keV) using ENDF data; [1)]10^{20} atoms ^{238}U (≈ 40 mg); [2)]calculated for 25 keV Maxwell-Boltzmann distribution.

sample	irradiation	fluence [n cm^{-2}]	cross-sec. [barn]	^{239}Pu ats produced[1)]
IRMM-3	IKI	7.15×10^{13}	2.68	1.9×10^{10}
IRMM-5	IKI	2.78×10^{14}	2.68	7.5×10^{10}
MB-1	IKI	2.55×10^{12}	2.68	6.9×10^{8}
MB-2	IKI	1.37×10^{13}	2.68	3.7×10^{9}
IRMM-a	FZK-25	1.75×10^{15}	0.450[2)]	7.9×10^{10}
IRMM-b	FZK-500	4.34×10^{15}	0.109	4.7×10^{10}

3. SAMPLE PREPARATION for AMS

The various pellets (50-80 mg) were ground after the activation. A small fraction was directly used for AMS measurements of the $^{236}U/^{235}U$ isotope ratio without any pretreatment, i.e. for the direct measurement of $^{235}U(n,\gamma)$.

The larger fraction of the samples was sub-divided into several independent fractions. They were dissolved in nitric acid. A spike of ^{233}U (IRMM-058) [14] was added to trace the uranium chemistry for each subsample. Neutron capture on ^{238}U forms ^{239}U which decays ($t_{1/2}$ = 23.5 min) to ^{239}Np and ($t_{1/2}$ = 2.355 days) to ^{239}Pu (Fig. 1). Similarly to U, a ^{242}Pu spike was added (IRMM-085) [15]. After an appropriate waiting time, the Pu was separated from the U bulk material. Iron (AAS standard solution) was added (between 5 and 20 mg per fraction) to the residues and finally, a Fe hydroxide co-precipitation was performed. After drying, the U / Pu and iron hydroxide powder was combusted to produce U and Pu oxides, embedded within the Fe_2O_3 matrix (see Fig. 2).

In this way sputter targets for AMS were produced from each fraction. Overall, from each of the irradiated pellets, material for about 20-30 sputter cathodes was available. A similar amount of non-irradiated blank samples was produced in parallel via the same chemistry. These blanks were also used for the measurement of the natural concentration of ^{236}U and ^{239}Pu and served as an internal check to exclude possible contaminations during chemistry and AMS measurements. Similarly, samples containing the spike materials ^{233}U and ^{242}Pu only, without uranium powder, were prepared to demonstrate that no significant ^{236}U was present in the spike and also verified the (small) amount of ^{239}Pu in the ^{242}Pu spike.

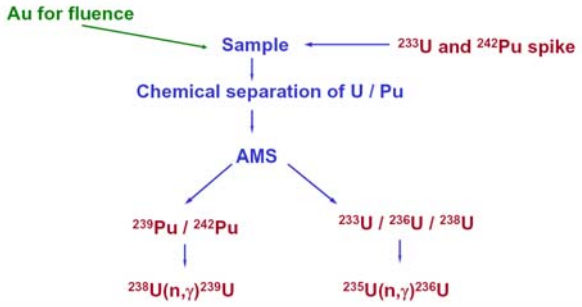

Fig. 2. Schematic view of the analytical strategy for preparation of AMS samples from the natural uranium samples.

4. AMS MEASUREMENTS

The AMS measurements were carried out at the VERA (Vienna Environmental Research Accelerator) facility, which is based on a 3-MV tandem accelerator. VERA represents a state-of-the-art AMS setup. Negative ions are produced from solid sample material in a cesium sputter source. A (small) fraction of the sputtered material will become negatively charged (≈ % for U and Pu) and is extracted from the source. Typical sample masses are mg of material.

The advantage of AMS is, that it does not suffer from molecular isobaric interferences due to the use of tandem accelerators and it can also be used for separating specific atomic isobars. An advantage is its flexibility for switching to different isotopes and elements because scaling is directly a function of the mass. It offers a powerful tool to measure cross sections of nuclear reactions leading to radioactive nuclides, independent of their decay times or schemes. The actually measured parameters in AMS are isotope ratios.

VERA has been recently upgraded to reach high sensitivities also for actinide isotopes [10,16]. To suppress interference from neighboring masses, the mass resolution of VERA was increased. Interfering ions which pass all beam filters are identified with a high-resolution time-of-flight (TOF) system and an ionization chamber. In switching mode, the various masses were measured sequentially; either the count rate of ^{236}U and the current of ^{238}U; or for Pu, by switching between ^{239}Pu and ^{242}Pu counting. Approximately, 20 to 30 sputter cathodes were prepared and measured by AMS in about 10 different beam times. The overall measuring time was of the order of 10 weeks of AMS runs.

One critical aspect was the stability of the measured natural $^{236}U/^{235}U$ isotope ratio of un-irradiated samples. The reason was that the natural ^{236}U content was comparable to the additional ^{236}U signal from the neutron activations. We measured a ratio $^{236}U/^{238}U$ of ≈ 7×10^{-12} for un-irradiated samples, while the activations generated an equivalent of a few 10^{-12} to at most 2×10^{-10} (see Fig. 3, Table 1). For ^{239}Pu, the natural concentration was negligible compared to the one produced during the neutron activations.

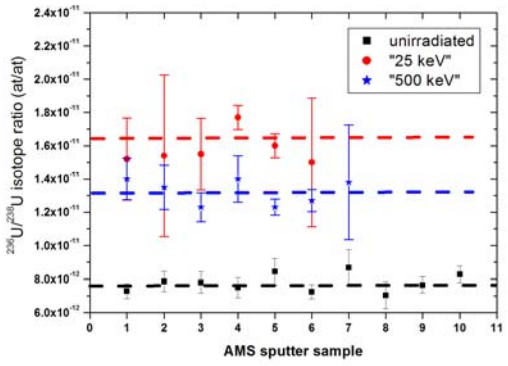

Fig. 3. Measured $^{236}U/^{238}U$ ratios in one measurement series for different sputter cathodes; shown are data for un-irradiated blanks (squares), 25 (circles) and 500 keV (stars) irradiations.

5. RESULTS

Cross-section values can be directly calculated from the neutron fluence and the isotope ratio measurements obtained by AMS. Adding a spike, results in a constant ratio relative to the ^{235}U or ^{238}U atoms. For quantifying systematic uncertainties within this work, several redundant data were available:

(1) In case of ^{235}U(n,γ)^{236}U, isotope ratios were generated relative to ^{235}U (and ^{238}U), and also relative to the spike ^{233}U (^{236}U/^{238}U and ^{236}U/^{233}U); (2) the irradiated pellets were split into sub-samples, and different amounts of ^{233}U were added, hence leading to different ^{236}U/^{233}U isotope ratios. (3) a different U material (two pellets) was irradiated with cold neutrons at a 2nd activation station. For those, data analysis is in progress.

For ^{238}U(n,γ)^{239}U, the AMS data were obtained relative to the ^{242}Pu spike. Again, different sub-samples (the same as for U) were processed and gave individual ^{239}Pu/^{242}Pu ratios, which finally represent the same cross section values.

These data rely on the ^{197}Au(n,γ)^{198}Au capture cross section values for the various neutron energies which is under debate for keV energies [4]. However, as we used the very same sample for both capture reactions, the neutron fluence was the same: the ratio of the reaction cross sections is equivalent to the ratio ^{239}U/^{238}U over ^{236}U/^{235}U, i.e. the neutron fluence cancels. Therefore, the corresponding AMS isotope ratios will directly give also the ratio of the cross sections, independent of Au.

The AMS measurements are mostly finished and data analysis is in progress. In particular, the neutron spectral distributions for activations with "25 keV neutrons" and 500 keV need to be folded with the energy-dependent cross sections. This is important for 25 keV: This spectrum is not symmetrical in energy, and the cross section is varying with neutron energy. At 500 keV, however, the energy distribution in the irradiation was symmetrical and, in addition, the cross section is fairly constant around 500 keV.

(1) **^{235}U(n,γ)^{236}U**: the ^{236}U data were normalized with our in-house ^{236}U standard (known to ± 4%). We obtain a preliminary value for the thermal cross section of 103±5 barn, respectively, for the two samples. The final data will be compared to the better known ^{233}U spike. First results for 500 keV indicate a value of 161 mbarn, in good agreement with ENDF and JEFF (163 mbarn), and slightly lower than JENDL (171 mbarn).

(2) **^{238}U(n,γ)^{239}U:** for 500 keV we obtain a preliminary value of 109 mbarn, in agreement with ENDF and JENDL (109 mbarn), and slightly lower than JEFF (119 mbarn).

(3) Ratio of **^{238}U(n,γ)^{239}U/^{235}U(n,γ)^{236}U** at "25 keV" (Maxwell-Boltzmann): If we take the AMS ratios, they translate into the cross-section ratio $\sigma_{238U\text{-}n\gamma}/\sigma_{235U\text{-}n\gamma}$. A value of 0.63 was obtained, which is in good agreement with both, ENDF and JEFF values (0.65).

All those data still are preliminary, as data analysis is not finished yet. Also, a systematic shift of a few percent is possible for the final results: up to now, data were normalized to our in-house reference material; and the spread of the neutron energy distribution was not taken into account yet. The uncertainty of the final data is expected to be at a level of 4-5%.

6. SUMMARY

AMS represents a powerful technique with excellent sensitivity for the detection of long-lived radionuclides via ultra-low isotope ratio measurements. The combination of activation and subsequent AMS detection is an independent and complementary technique, and thus depends on systematic uncertainties different to standard techniques [11,17]. At VERA the accuracy of AMS measurements of actinides is of the order of a few %. This technique can be extended to other isotopes in that mass range.

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