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ANALYSIS OF URANIUM AND THORIUM THIN TARGETS IRRADIATED AT THE PSI ACCELERATOR

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The aim of the ATHENA[1] programme at PSI is to provide experimental data for the validation of theoretical models in nucleon-meson transport codes used for accelerator-based transmutation studies. Emphasis is placed on the mass yield distribution of spallation and fission products for irradiated thin actinide targets. This paper presents results of an irradiation experiment carried out with $^{238}UO_2$ and $^{232}ThO_2$. Isobaric production cross-sections of fission and spallation products based on mass spectrometric measurements and γ-spectroscopy are compared with calculations carried out using the HETC[2] code and the RAL[3] high-energy fission model.

1 INTRODUCTION

The experimental ATHENA programme at PSI related to accelerator-based transmutation concerns thin-target proton irradiations of actinides. The purpose of the irradiations is to provide experimental data for the validation of high-energy fission models in nucleon-meson transport codes, with emphasis on the mass yield distribution of spallation and fission products. Semi-empirical models involving different types of adjustments are applied in the codes available. Due to the limitations of the models applied there is considerable spread in calculational results for the irradiation-product mass-yield distributions for different actinides. In the context of transmutation, especially for systems employing direct proton bombardment of minor actinide targets, such differences can be important due to the widely different toxicities and half-lives of the various fission and spallation products.

Results of a first thin-target irradiation experiment with $^{238}UO_2$ at PSI were reported earlier [1]. The present article gives results of a second experiment carried out with $^{238}UO_2$ and $^{232}ThO_2$ targets and was essentially presented at PHYSOR 96 [2]; some more recent findings are being included in Sections 2 and 6. The basic experimental setup and the analysis methods, viz. γ-spectroscopy, inductively coupled plasma mass spectrometry (ICP-MS) and total reflection X-ray fluorescence (TXRF), were described in detail [1] and are not repeated here except in the context of changes which have been made recently. The computer code used at PSI for providing comparisons with calculations is HETC [3] with the RAL high-energy fission model [4].

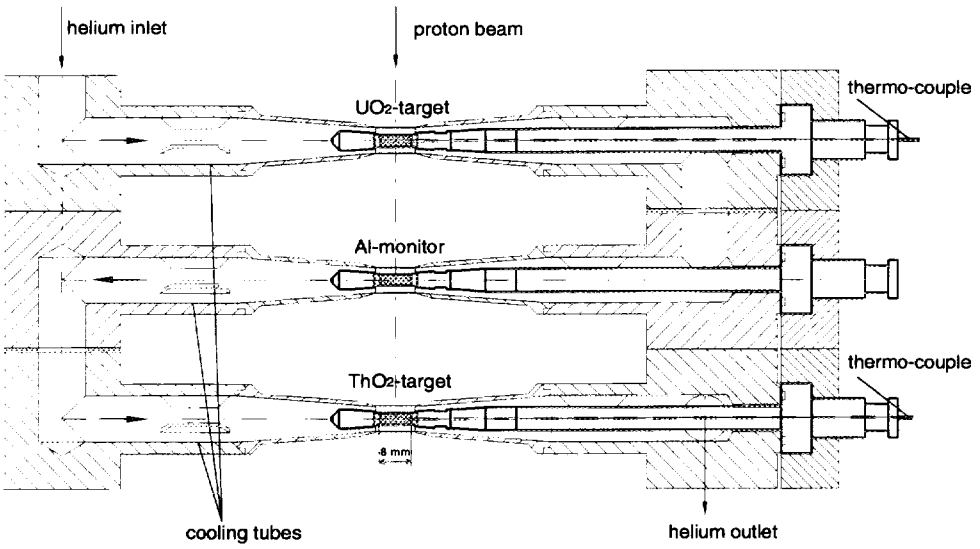

Fig. 1: Irradiation head with the three geometrically identical targets

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[1] *ATHENA* = actinide transmutation using high energy accelerators

[2] *HETC* = high energy transport code

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2 EXPERIMENTAL SETUP

The samples were irradiated in the PIREX (proton irradiation experiment) facility using 0.6 GeV protons from the PSI ring accelerator. In contrast to the first irradiation, the proton beam did not have to be shared with other experiments at the accelerator, thus providing fairly stable conditions and a total fluence about 100 times larger than the one obtained earlier. An irradiation head with three targets in series was used (Fig. 1), which allowed the simultaneous irradiation of $^{238}UO_2$, ^{27}Al and $^{232}ThO_2$, as the proton beam was not affected significantly by the targets themselves. Each actinide target consisted of ~ 240 mg of microspheres (~0.3 mm in diameter). The use of aluminium as fluence monitor via measurement of the Na-22 γ-activity from the $^{27}Al(p,x)^{22}Na$ reaction made it possible to obtained more objective results for the individual yields. During a 63-hour long irradiation a total fluence of $(4.19 \pm 0.07) \cdot 10^{19}$ p/cm^2 was achieved.

Approximately 95% of each irradiated actinide target was dissolved and diluted with HNO_3 for the different analysis methods. The rest was directly used for additional γ-measurements. To date, mass spectrometry and γ-spectroscopy have been applied to analyse each actinide target. Initial high-sensitivity TXRF measurements at the European Synchrotron Radiation Facility at Grenoble in August 1996 revealed that extra shielding is required in order to protect the detection system against background radiation. Special Al-Pb-Al sandwich devices are currently under construction for this.

3 MASS-SPECTROMETRIC ANALYSIS OF THE IRRADIATED SAMPLES

3.1 Prerequisites

The results obtained from the mass-spectrometric analysis are most important for the verification of calculated isobaric mass yield distributions since they are, at least for mass numbers < 204, totally independent of any model assumptions and rely only on the experimental accuracy. For each measurement, the mass spectrum of the irradiated target material was obtained in parallel to that for unirradiated material using identical chemical processing steps. This procedure revealed that certain mass ranges had to be excluded due to impurities present in the unirradiated samples. In addition, some other ranges had to be omitted because of matrix and/or argon ion interference.

As the expected yield for many of the mass numbers is close to the detection limit, two compensating effects need special attention: larger sample concentrations (and hence, larger trace element concentrations) are in competition with the increasing signal suppression of trace elements by the larger matrix element concentration (U and Th). It was shown in a separate investigation [5] that 0.1% - 0.3% uranium solutions gave best detection limits for a large number of trace elements. To avoid stability problems due to sample deposition on the cones of the ICP-MS, only small volumes (280 µl) of 0.1% U and Th solutions were analysed.

Element-dependent sensitivities were determined by measuring standard solutions containing the elements Li, Be, B, Mg, Sc, V, Mn, Co, Cu, Zn, Ga, Ge, As, Se, Rb, Y, Nb, Rh, Ag, Cd, In, Sb, Te, Cs, La, Pr, Nd, Eu, Gd, Ho, Tm, Lu, Ir, Pt, Au, Tl, Pb, Bi, Th and U. Due to their chemical similarity the sensitivities for the lanthanides do not vary much (7000 - 8000 counts/sec per ppb). Generally, for a large part of the periodic table the sensitivity values are within 30% of those for the lanthanides. However, the sensitivities of elements with unusually high and low values may differ by a factor of 10 to 100. In a 0.1% U solution trace element signals are suppressed by a factor of 2 and vary by less than 10% for the set of different trace elements [5]. In preliminary estimates of the mass distributions, a global value of 7000 counts/sec per ppb was assumed and a suppression factor of 2 in 0.1% target solutions was used for all masses.

The fractional contribution of different elements to a certain mass number is, at the time of measurement, sufficiently well known from the viewpoint of applying relative sensitivity corrections. Representative sensitivity values for nearly all the measured mass numbers were evaluated from the fractional contributions of the HETC-calculated components. Some missing sensitivity values were found by linear interpolation between those of the nearest known neighbouring elements and by considering the fact that low sensitivity is usually related to high ionisation potential. This treatment of element-dependent sensitivity represents a refinement on the above global values of 7000 counts/sec per ppb.

Detection limits were found to lie usually between 0.003 and 0.03 ppm depending on the background signals. Only those mass-yield concentrations were considered which gave a signal at least five times the standard deviation of the blank (i.e. non-irradiated) analyte.

3.2 Results

In the mass range 80 - 160 a large number of nuclides are detectable. As expected, the refinement procedure smoothed the experimental mass-yield curves. The majority of the fission-product isotope concentrations determined between mass numbers ~ 70 - 170 have individual standard deviations of less than a factor of 2, only a few deviations being as much as a factor of 3.

Mass yields between mass numbers 183 and 194 are almost exclusively composed of platinum metals. Because of their chemical inertness they can only be partly dissolved. Some species may even sublimate, e.g the volatile OsO_4 oxidation product. Conse-

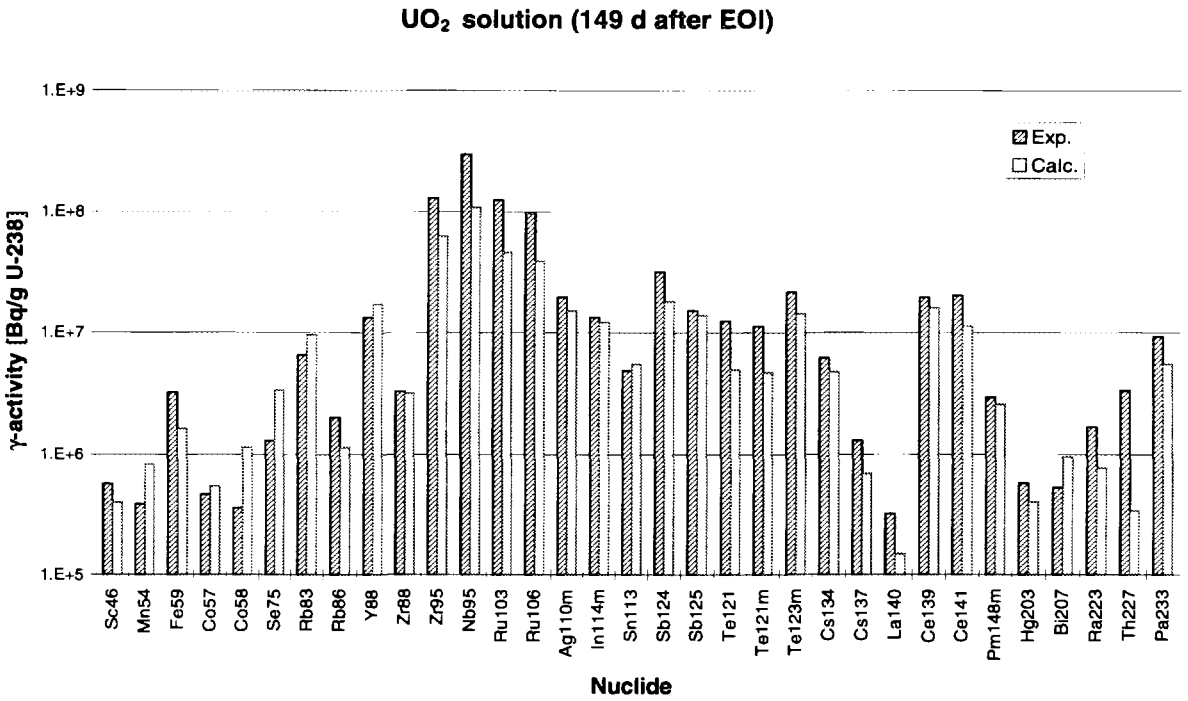

Fig. 2: Comparison of measured and calculated γ-activities of a U-238 solution sample

quently, the measured mass yields could be lower than their actual values. When calculating the concentration, the dominating presence of low-sensitivity elements around mass number 200 (Au, Hg) results in low count rates which have to be divided by low sensitivity values, thus introducing uncertainties which may be as large as a factor of 3 - 5. Similar arguments hold for mass numbers around 230 with their dominant Th contribution. Due to impurities or argon ion interferences, the mass range below mass numbers ~ 70 had to be omitted for both targets, and the range between 165 and 186 is reported only for Th.

4 RESULTS FROM γ-SPECTROSCOPY

The γ-spectroscopy method was applied to determine the experimental proton fluence from the Al-monitor, as well as to provide qualitative confirmation of the mass spectrometric results for the uranium and thorium targets.

The ^{27}Al target was geometrically identical to the actinide targets and, as regards the experimental setup, was exposed to the same fluence. Three measurements of the Na-22 γ-activity at different times gave a measured fluence value of $(4.19 \pm 0.07) \cdot 10^{19}$ p/cm^2 with 13.6 mb assumed as the reference cross-section

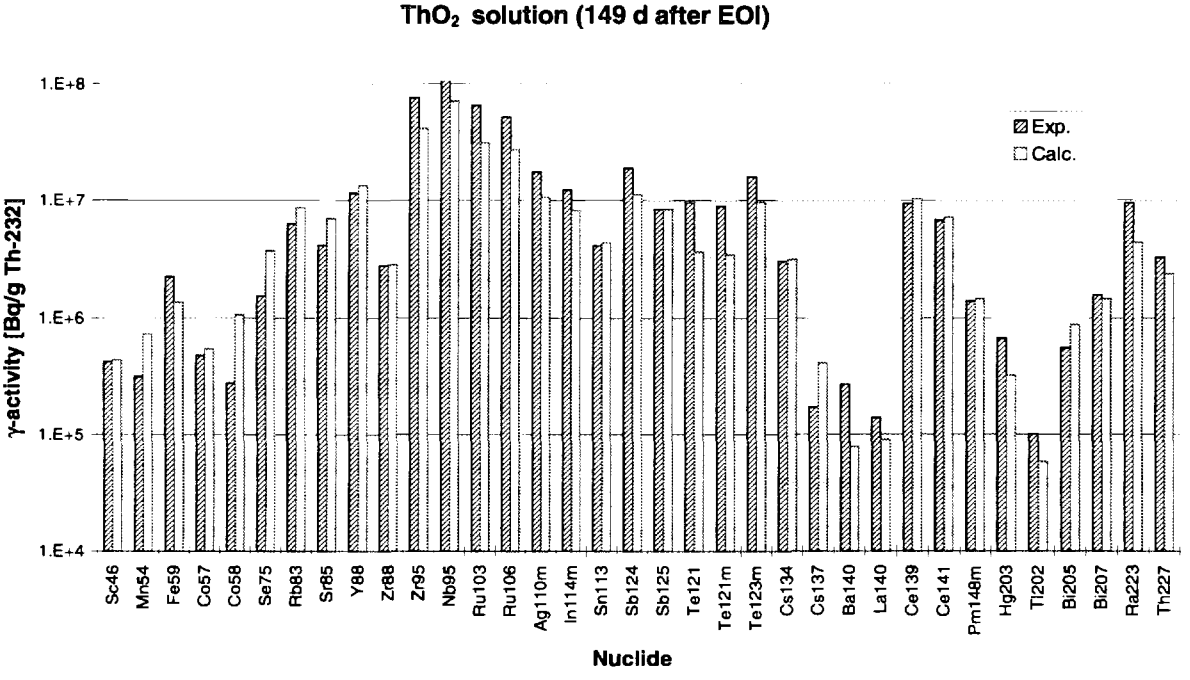

Fig. 3: Comparison of measured and calculated γ-activities of a Th-232 solution sample

for the $^{27}Al(p,x)^{22}Na$ reaction [6]. The indicated uncertainty corresponds simply to the achieved accuracy for the γ-measurement, the error in the assumed cross-section value not having been considered.

Three γ-spectra of solid samples and two of samples in solution were measured at different times and analysed for each actinide target, using procedures similar to those described [1]. Fig. 2 and Fig. 3 show calculation/experiment comparisons for U-238 and Th-232 solution spectra, both taken 149 days after the end of irradiation (EOI). The results are broadly consistent with the predictions. They have been used to provide a qualitative comparison of isobaric production cross-sections by employing time-dependent corrections based on the HETC/RAL and ORIHET[4] calculations [1]. They have not, however, been used for the determination of the total fission cross-sections, in contrast to the mass spectrometric results with their much lower dependence on theoretical correction factors.

time. Thus, in this mass range, the experimental isobaric production cross-sections can be directly calculated from mass-spectrometric concentrations by using the measured proton fluence. For mass numbers > 203, the isobaric production cross-sections at the time of irradiation were extrapolated from their values at measuring time by multiplying them by time-dependent extrapolation factors obtained from model calculations.

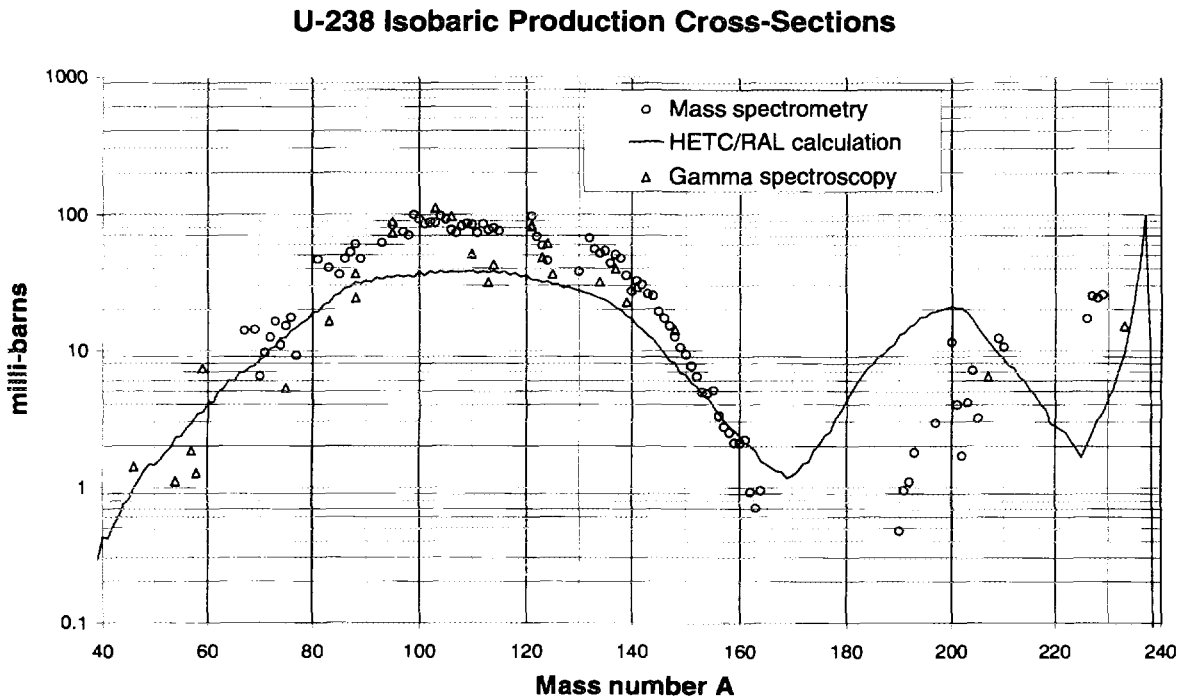

Fig. 4: Comparison of U-238 isobaric production cross-sections deduced from mass and γ-spectroscopic measurements with theoretical values from HETC/RAL

5 DEDUCED PRODUCTION AND FISSION CROSS-SECTIONS

5.1 Isobaric Production Cross-Sections

In the β-decay range for mass numbers < 204, the cumulative isobaric mass yields do not depend on

As indicated earlier, individual nuclide mass yield values from measured γ-activities can be extrapolated theoretically to obtain isobaric production cross-sections. These results, however, are more indirect than those from mass spectrometric measurements. They are, nevertheless, useful as confirmatory evidence.

Fig. 4 and Fig. 5 compare the currently obtained experimental isobaric production cross-sections for U-238 and Th-232, respectively, with theoretical values from HETC/RAL calculations.

5.2 Fission Cross-Sections of Uranium and Thorium

The integral of the isobaric production cross-sections in the fission-product hump (A < ~ 170) is twice the fission cross-section of a target nuclide. This fact allows an estimate of the total fission cross-sections for both actinide targets and a comparison with earlier measurements.

Numerical integration of the isobaric production cross-sections deduced from mass spectrometry was per-

[4] an adaption of the Oak Ridge isotope generation and depletion code ORIGEN

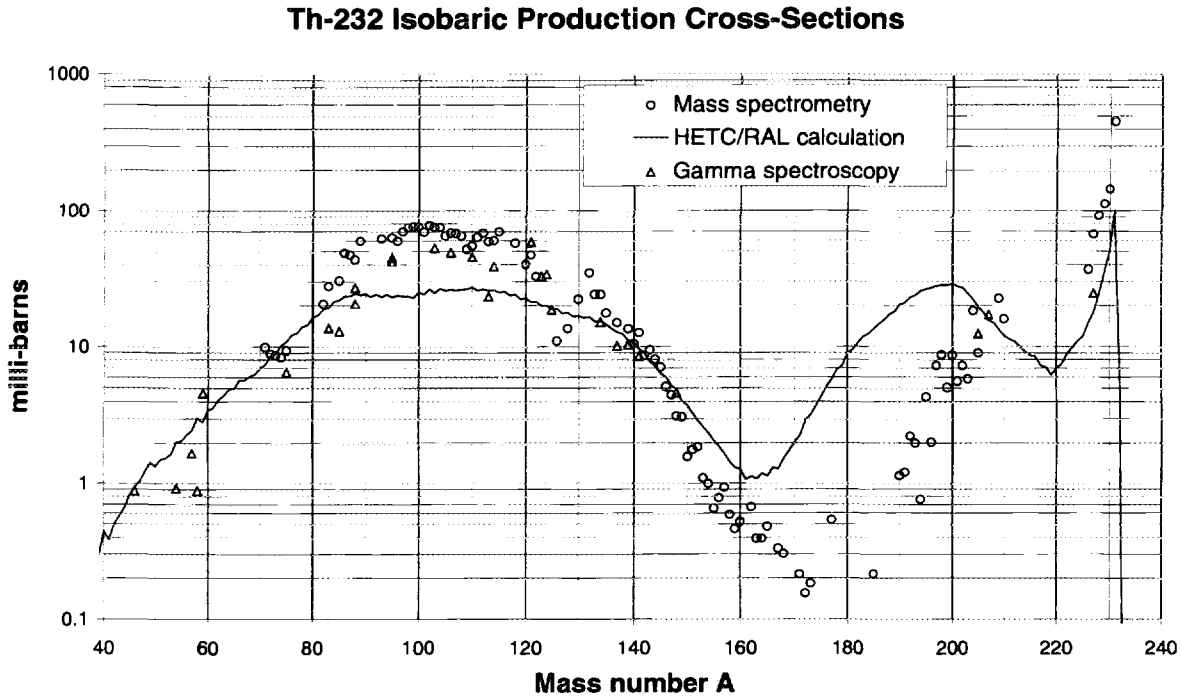

Fig. 5: Comparison of Th-232 isobaric production cross-sections deduced from mass and γ-spectroscopic measurements with theoretical values from HETC/RAL

formed by first least-squares fitting a Gaussian bell-shaped curve to each set of observations and then integrating this curve. Fig. 6 shows the shape of the fitted curves on a linear scale. The resulting total fission cross-sections are as high as 2.27 b for U-238 and 1.51 b for Th-232.

2.6 times larger than predicted for U-238 and Th-232, respectively. The mass range below mass number ~ 70 is uncertain and will be the subject of further investigations.

The ascent to the hump of spallation products (A > ~ 170) appears to be shifted towards higher

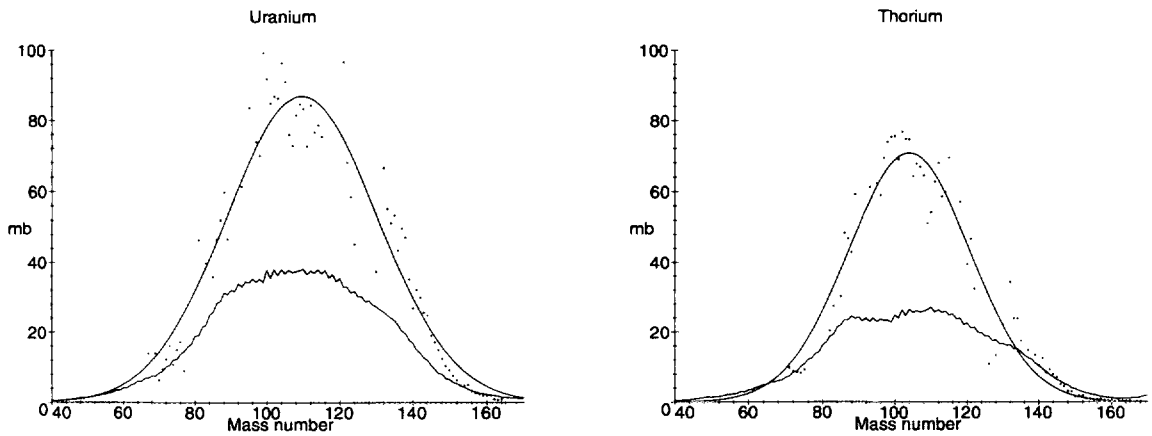

Fig. 6: Comparison between Gaussian fits to the experimental production cross-sections for individual fission products (upper curves) and results from HETC/RAL calculations (lower curves)

6 CALCULATION/EXPERIMENT COMPARISONS

In the isobaric production cross-section distributions deduced from mass spectra (Fig. 4 and Fig. 5), the general shape and position of the maximum value for the fission product hump are broadly consistent with HETC/RAL predictions. The experimental maxima (taken as maxima of the least-squares fitted bell-shaped curves) are, however, approximately 2.3 and masses by about 20 mass units and the dip between the humps of fission and spallation/evaporation products is shifted to the right and is deeper than predicted for both targets. There is, however, considerable uncertainty in the experimental points in this mass range as explained in the "mass-spectrometric analysis" section. We also note that results for uranium are totally absent between mass numbers 166-186, due to impurities in the unirradiated sample.

The total experimental fission cross-sections σ_f deduced from integration of the fission-product hump (2.27 b and 1.51 b for U-238 and Th-232, respectively) are about twice as large as results from the earlier measurements of Kon'shin et al. [7]. The HETC/RAL predictions of 1.13 b and 0.789 b for U-238 and Th-232, respectively, correspond quite well to their measurements.

As an instructive comparison, a very approximate estimate of spallation cross-sections is obtained by integration of the sparse spallation hump. This gives values of ~ 0.5 b and ~ 1.4 b as compared to 0.708 b and 1.014 b from HETC/RAL for U-238 and Th-232, respectively. These correspond to total inelastic cross-sections σ_a of ~ 2.8 b and ~ 2.9 b as compared to 1.85 b and 1.82 b from HETC/RAL for U-238 and Th-232, respectively.

Approximate experimental ratios of fission to total inelastic interaction cross-sections, σ_f/σ_a ("fissility"), are as high as 0.81 and 0.52 as compared to HETC/RAL predictions of 0.61 and 0.44 for U-238 and Th-232, respectively. For both actinides the experimental ratios are about 30% larger than evaluated by using an empirical relation suggested by N.A. Perfilov [8].

Work is currently in progress to confirm the large experimental cross-sections. A recent investigation based on measured data during and after the irradiation experiment (integrated total beam current, horizontal and vertical beam profiles at various stages of the irradiation, γ-scan along target axis, etc.) has shown that significant geometric and beam-profile effects can be excluded. Similarly, the estimated uncertainty in the fluence evaluation (combination of the uncertainties of the γ-measurements and the $^{27}Al(p,x)^{22}Na$ production cross-section) is < 15% and cannot account for the reported differences by a factor of ~ 2. We are seeking more information from additional mass-spectrometric measurements in selected ranges (< 70, 180 - 210, > 226) and from TXRF measurements planned for early 1997.

7 CONCLUSIONS

A dedicated ATHENA irradiation has provided appropriate conditions for applying various experimental methods to the analysis of irradiated uranium and thorium thin targets. The use of an irradiation head with three targets enabled direct monitoring of the proton fluence with aluminium to provide a standard. The deduction of absolute production cross-sections is thus possible.

Inductively coupled plasma mass spectrometry has, for the first time, successfully been applied for obtaining isobaric fission and spallation product yields. Certain mass ranges were difficult to measure (< 70, 180 - 210, > 226) and need further investigation. Isobaric production cross-section values evaluated from the ICP-MS measurements have been found to be broadly consistent with those deduced from γ-spectra. The latter, however, are more dependent on model assumptions than were the former.

The shape of the experimental isobaric production cross-sections broadly corresponds to the curves calculated by HETC/RAL. The fission hump, however, was found to be larger than predicted, and the spallation hump appeared shifted towards higher mass numbers. The predicted differences between U-238 and Th-232 in the fission-product region are reflected in the experimental results. The same may be said for the spallation hump region, but experimental uncertainties are larger in this case.

Total fission cross-sections deduced from integration of the fission humps, for both U-238 and Th-232, are about twice as large as predicted values and results obtained in earlier experiments. A significant underestimate of the experimental fluence, however, due to geometric and beam-profile effects can be ruled out, and further investigations are underway.

The purpose of the irradiation experiment was to provide experimental mass-yield data for the validation of high-energy fission models in nucleon-meson transport codes. This aim is fulfilled for the range of fission products (~70 < A < ~170). In the range of spallation products (A > ~170) and for low mass numbers (A < ~70), however, the uncertainties are large and more measurements are necessary. In addition, the tendency to larger production cross-sections in the fission hump should be confirmed by further irradiation experiments including other actinides.

8 ACKNOWLEDGEMENTS

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