Residual Nuclide Production By Proton-Induced Reactions On Uranium For Energies Between 20 MeV And 70 MeV

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Abstract. Within the HINDAS project, proton irradiation experiments were performed at the injector cyclotron of the Paul Scherrer Institute at Villigen/Switzerland in order to investigate the production of residual nuclides from natural uranium. The stacked-foil technique was used to cover proton energies between 20 MeV and 70 MeV. Copper targets were used for monitoring the proton beam using the reaction ^{65}Cu (p,n)^{65}Zn. Residual radionuclides were measured by off-line γ-spectrometry. Excitation functions were obtained for the production of ^{91}Y, ^{95}Zr, ^{95m}Nb, ^{99}Mo, ^{103}Ru, ^{112}Pd, ^{115}Cd, ^{124}Sb, ^{126}Sb, ^{127}Sb, ^{132}Te, ^{131}I, ^{134}Cs, ^{136}Cs, ^{137}Cs, ^{140}Ba, ^{141}Ce, ^{144}Ce, ^{147}Nd, and ^{238}Np. The experimental data are compared the sparse results of earlier measurements and with theoretical excitation functions calculated by the newly developed TALYS code. Good agreement between theory and experiment was obtained for product masses up to 115. For higher mass fission products and for ^{238}Np there are still systematic deviations between theory and experiments. These deviations are discussed as deficits of the fission model in the heavy part of the fission product distribution.

INTRODUCTION

For proton-induced reactions, the efforts of the HINDAS (*High and Intermediate Energy Nuclear Data for Accelerator-Driven Systems*) project [1] aimed to further developing and completing the cross section database which was established by our collaboration in recent years; [2] and references therein. During the HINDAS project the data base was extended to the heavy target elements Ta, W, Pb, and Bi [3-6]. As part of these efforts, also the production of residual nuclides by proton-induced reactions with natural uranium was investigated [7]. For the target element uranium just sparse and partially contradictive earlier data were available. The new, systematic experimental results for the energy region from 20 MeV to 70 MeV closed some gaps and allowed to decide about some of the discrepancies. They provided a basis to test the new TALYS code developed within the HINDAS project. The TALYS code includes the optical model, direct, pre-equilibrium, fission and statistical models and thereby gives a prediction for all the open reaction channels; see A. Koning et al. in this conference proceedings.

EXPERIMENTAL

Cross sections were determined using irradiation experiments with classical kinematics and subsequent γ-spectrometry. The irradiation experiments were performed at the injector cyclotron at PSI/Switzerland using the stacked-foil technique. Two stacks consisting of uranium foils in natural isotopic composition separated by high purity copper foils for flux monitoring were irradiated for about 7.74 h and 4.98 h, respectively, at initial proton energies of 70.92 MeV and 44.98 MeV. The 70 MeV stack also contained some aluminum targets for cross checking the fluence determinations.

All the uranium, copper, and aluminum targets were high-purity (99.999 %) materials supplied by Goodfellow Metals Ltd., UK. A measurement of the isotopic composition of the uranium targets yielded 0.006 % ^{234}U, (0.71 ± 0.004) % ^{235}U, <0.005% ^{236}U and (99.27 ± 0.01) % ^{238}U.

After irradiation the samples were transported to Hanover and each foil was subsequently measured up to 15 times covering decay times from hours up to 6 months after irradiation by means of γ-spectrometry. Up to six GeLi and HPGe detectors in different geometries were used to minimize systematic influences. The peak areas in the spectra were determined with the commercial software GammaW [8]. Detector efficiencies were determined with calibrated radionuclide sources and interpolations of the efficiency points were made using a fit-function proposed by Gray and Ahmad [9]. The initially produced activity of every identified radionuclide was computed taking into account decay corrections, γ-ray probabilities, detector efficiencies, and self-absorption of γ-rays in the targets. was chosen. All necessary nuclear data were taken from [10].

The energy degradation in the stack was calculated with a PC program based on the work of Anderson and Ziegler [11]. The stacks were designed to meet the following requirements: 1. to provide overlapping energy regions by arranging several stacks to complement each other; 2. to determine the proton flux inside the stack via monitor reactions in the copper foils; 3. to avoid any recoil contamination or recoil loss of produced nuclides by covering each measured foil with thin catcher-foils of the same element; and 4. to check the accuracy of the calculated proton energies inside the stack by comparing the determined cross-sections in the copper foils with previously measured values.

Activities obtained for ^{65}Zn in the copper foils were used to calculate the proton fluences using the monitor cross sections given in ref. [2]. From the copper foils also 80 new cross sections for the production of ^{56}Co, ^{57}Co, ^{58}Co, ^{60}Co, and ^{59}Fe from natural copper were determined [7]. The results were in excellent agreement with our earlier measurements [2]. Together with the results obtained from the aluminum targets they confirmed our energy calculations and fluence determination and showed within the experimental uncertainties no evidence of secondary particles in the stacks. The proton fluences for the first uranium targets in the two experiments were $6.19 \cdot 10^{15}$ cm^{-2} and $2.90 \cdot 10^{15}$ cm^{-2}, respectively.

RESULTS AND DUSCUSSION

For proton-induced reactions on natural uranium, we determined a total of about 460 cross sections for the production of ^{91}Y, ^{95}Zr, ^{95m}Nb ^{99}Mo, ^{103}Ru, ^{112}Pd, ^{115}Cd, ^{124}Sb, ^{126}Sb, ^{127}Sb, ^{132}Te, ^{131}I, ^{134}Cs, ^{136}Cs, ^{137}Cs, ^{140}Ba, ^{141}Ce, ^{144}Ce, ^{147}Nd, and ^{238}Np from natural uranium for 23 individual energies energies between 21 MeV and 69 MeV. In accordance with earlier discussions [2, 3, 4], we estimate the systematical uncertainties from all known sources of uncertainty to be less than 8 % not including the uncertainty due to counting statistics.

Except for ^{238}Np, produced via ^{238}U(p,n)^{238}Np, all the radionuclides observed are fission products. Most of the cross sections determined are cumulative ones including the production from the decay of radioactive progenitors. Only the cross sections for ^{124}Sb, ^{134}Cs, ^{136}Cs, and ^{238}Np are independent ones.

To survey earlier work, a literature search was performed with EXFOR databases maintained by OECD NEA Data Bank (Paris) and National Nuclear Data Center (Brookhaven). The survey resulted in 34 references for ^{nat}U and 12 for ^{238}U. Most of the reports covered only a limited region of incident energy and/or product nuclides (e.g., some light nuclides, fission fragments, or spallation products). Moreover, 27 out of 46 found publications dated back to 1950-60ies. Almost all early experiments employed chemical separation as an intermediate step between the beam exposure and the radioactivity measurement. Furthermore, most detection techniques employed in the early studies did not include spectrometric techniques for cross section determinations.

In summary, the data base for residual nuclide production by proton-induced reactions on uranium is not comprehensive with respect to product or energy coverage and frequently the data quality is insufficient. Only for a few products, e.g. ^{99}Mo, ^{136}Cs, ^{147}Nd, and ^{238}Np, the complete excitation functions can be roughly estimated for the energies relevant for accelerator-driven techniques, i.e. from thresholds up to a few GeV. In our comparison of the new results with the TALYS calculations (Fig. 1-3) also the earlier work is included thus giving a taste of the presently available data base for uranium. A detailed discussion of the earlier data is given elsewhere [7].

Our new results cover just a relatively small energy range, but they allow deciding about some discrepancies in the earlier data, for some nuclides they are the first data at all, and, last not least, they allow for a systematic test of theoretical calculations

using the newly developed TALYS code. To the latter end, blind calculations were performed in a default mode of TALYS for proton energies up to 250 MeV. Some exemplary results are compared with our new data and the existing earlier ones in Fig. 1 to 3.

As examples for the low-mass products we use ^{95}Zr and ^{115}Cd (Fig. 1). For ^{95}Zr only one earlier measurement [12] existed for energies up to 300 MeV. The TALYS calculations are in excellent agreement with our measurements and with the only earlier data point. The agreement between theory and experiment seen in Fig. 1 is typical for all product nuclides with masses up to 115.

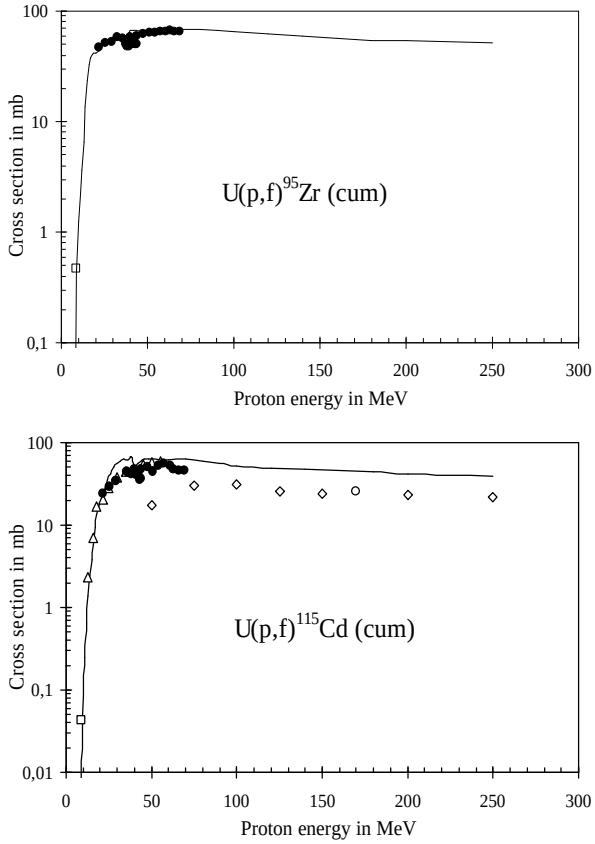

FIGURE 1. Comparison of experimental cross sections from this work (full circles), from Baba et al. [13] (triangles), Hicks et al. [14] (diamonds), Pappas [15] (open circles), and Yokoyama [12] (squares) with theoretical ones calculated by the TALYS code for the production of ^{95}Zr and ^{115}Cd in proton-induced fission of natural uranium.

For ^{115}Cd the experimental data base is better than for 95Zr and allow for a comparison over the entire energy range, but some discrepancies remain. It cannot be decided whether the differences between experiment and theory are due to failure of the theory or simply reflect the inconsistency of the experimental data base.

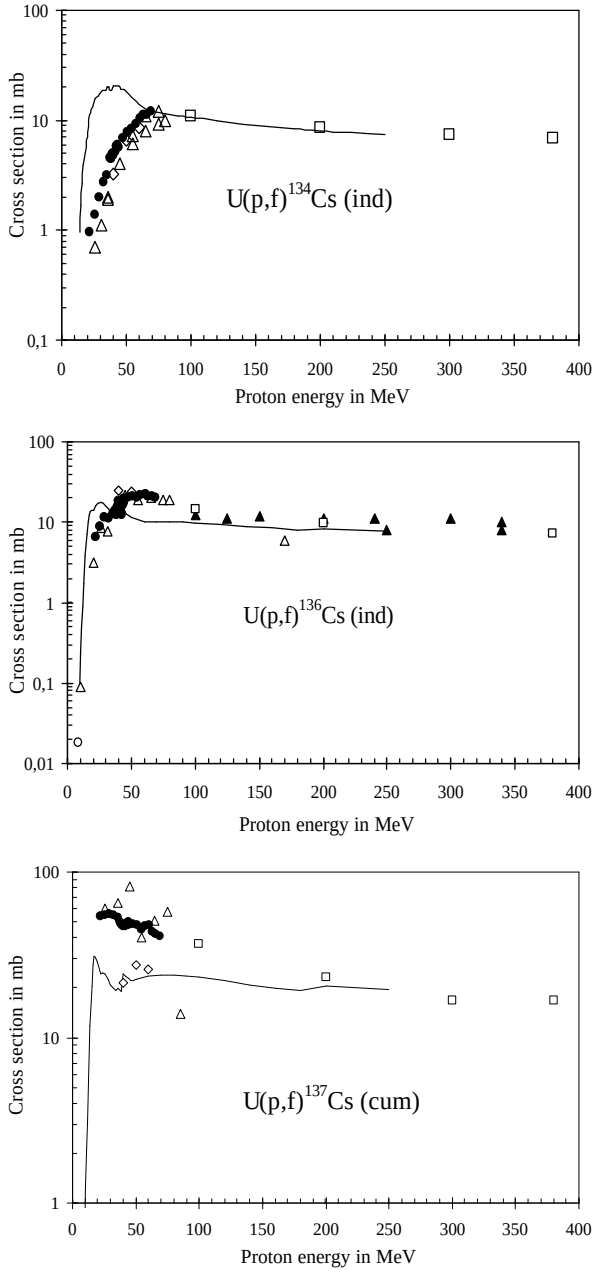

FIGURE 2. Comparison of experimental cross sections from this work (full circles), from Davies et al. [16] (triangles), Friedlander [17] (open squares), Hicks et al. [14] (full triangles), Tracy et al. [18] (diamonds), and Yokoyama [12] with theoretical ones calculated by the TALYS code for the production of Cs-isotopes in proton-induced fission of natural uranium.

For heavier fission products and for ^{238}Np the comparison between experimental and theoretical data shows several systematic discrepancies. In Fig. 2, this is exemplified by the production of cesium isotopes with masses 134, 136 and 137. For all three nuclides, a number of earlier measurements exist for energies below 300 MeV which allow estimating their excitation functions fairly well. Above 100 MeV the TALYS calculations tend to agree well with the experimental data. But at lower energies deviations between theory and experiment are seen which systematically change with product mass number (Fig. 2). The theory does not reproduce the shapes of the excitation functions at low energies. Also, the maxima of the experimental excitation functions are wrongly calculated with respect to their energies and shapes. These discrepancies are typical for all fission products with masse above 115 up to A = 147 (Fig. 3) and also for ^{238}Np a systematic overestimate by theory is observed. These deviations point to deficits of the fission model [20] in the heavy part of the fission product distribution. The systematic of the new experimental data provide a basis to describe these deficits and to improve the fission models.

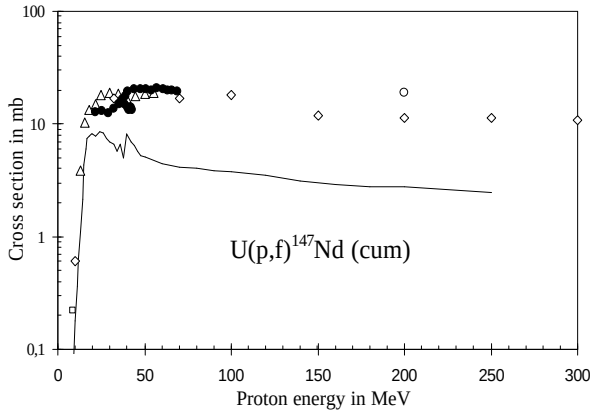

Figure 3. Comparison of experimental cross sections from this work (full circles) and from Baba et al. [13] (triangles), Friedlander [17] (open circles), Stevenson et al. [19] (diamonds), and Yokoyama [12] (open squares) with theoretical ones calculated by the TALYS code for the production of ^{147}Nd in proton-induced fission of natural uranium.

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