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Cross Sections for the Production of Residual Nuclides at Medium-Energies Relevant for Accelerator-Driven Technologies

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Introduction

Integral cross sections for the production of residual nuclides by medium-energy nuclear reactions are of importance for many fields of basic and applied sciences ranging from astrophysics over space and environmental sciences, medicine, accelerator technology, space and aviation technology to *a*ccelerator *d*riven *t*ransmutation of *w*aste (ADTW) and *e*nergy *a*mplification (ADEA). The data needs of those applications cover cross sections for proton- and (more importantantly) neutron-induced reactions. The particular problem for accelerator driven technologies is that the data needs are extreme with respect to both, target element coverage and types of reaction data. Since it will be practically impossible to measure all relevant data, one will have to rely to a large degree on theoretical estimates.

Given the fact that the predictive power of present day models and codes does not satisfy the requirements, an initiative was taken by a number of European laboratories to improve this situation. To this end, a complete experimental data base was and is being established for a selected number of target elements depending on which newly developed models shall be scrutinized.

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This initiative started with a European Concerted Action “Lead for Transmutation” [1] and continued by the 5th Framework project HINDAS “High- and Intermediate-Energy Data for Accelerator-Driven Systems” [2, 3]. Here, we report on some aspects of work-package 3 of HINDAS, namely the production of residual nuclides by proton- and neutron-induced reactions.

For proton-induced reactions, these investigations aim to further develop and complete the cross section database which was established by our collaboration in recent years [4, 5, and references therein]. It is now extended to heavy target elements such as Ta [6], W [6 - 8], Pb [9], and Bi [6]. New, but still unpublished data are available for the target element iron. Work on the target element uranium for proton-energies up to 72 MeV is underway. All those data allow for stringent tests of nuclear models and codes when calculating cross sections for residual nuclide production from thresholds up to 2.6 GeV.

For neutron-induced reactions the situation is worse. Contrary to their importance, the availability of neutron cross sections for the production of residual nuclides above 30 MeV is marginal. A solution of this problem is to perform irradiation experiments with quasi-monoenergetic neutrons produced by the $^{7}Li(p,n)^{7}Be$ reaction.

Production of Residual Nuclides by Neutron-Induced Reactions up to 175 MeV

Within the HINDAS project, activation experiments are performed at TSL/Uppsala and UCL/ Louvain La Neuve in order to determine excitation functions for the production of residual radionuclides from a variety of target elements up to 175 MeV.

The neutron beam-line at TSL/Uppsala [10] is equipped with a special irradiation chamber (PARTY facility) allowing for parasitic activation experiments [11]. Irradiations can be performed with peak neutron energies up to 175 MeV. Neutron flux densities of $\sim 0.5 \cdot 10^{5}$ cm^{-2} s^{-1} μA^{-1} (mm Li)$^{-1}$ are obtained in the high energy peak of the neutron spectrum. 99.984 % ^{7}Li-targets with a thickness between 2 and 15 mm can be used. Proton currents of 10 μA are available for energies up to 100 MeV; they are about a factor of ten lower for higher energies.

The UCL cyclotron facility provides neutron beams in the energy range from about 25 to 70 MeV, and well-characterized neutron reference fields have been previously established [12]. Proton currents up to 10 μA are possible; with a 5 mm ^{nat}Li target the resulting neutron flux density in the high-energy peak of the neutron spectrum is $\sim 1 \cdot 10^{5}$ cm^{-2} s^{-1} at a distance of 5m from the target.

A total of 10 activation experiments were performed with proton energies of 36.4, 48.5, and 62.9 MeV at the UCL and of 69.1, 76.4, 98.5, 136.7, 148.4, 162.7 and 178.8 MeV at TSL. The target elements C, N, O, Mg, Al, Si, Fe, Co, Ni, Cu, Ag, Te, Pb, and U were irradiated with the highest beam currents available. Cylindrical target stacks with a diameter of 22 mm and a total length of 66 mm were irradiated in the PARTY facility. Typical beam-on-target times were 40 h at UCL and 80 h at TSL. At UCL, cylindrical target stacks with a diameter of 25 mm containing the different target foils were inserted in the center of a larger stack with a 100 mm × 100 mm cross section which had the same macroscopic cross section as the target stack. The total length of the target stacks was 81 mm. These stack arrangements were positioned at a distance of ~ 5 m from the neutron-producing target. At this position, measurements of the lateral profile of the collimated neutron beam with a beam profile monitor [13] showed a width of the beam (FWHM) of about 55 mm and confirmed that, on the one hand, the target stacks were irradiated homogeneously and, on the other hand, the entire neutron beam passed through the stack arrangement and was attenuated in the same way.

At UCL, the absolute neutron fluence was determined with via methods: a proton recoil telescope, a ^{238}U fission ionization chamber, and an NE213 scintillation detector. The spectral fluence was measured with the TOF method, using the latter two systems. A specially developed multi-wire proportional counter yielded the transverse intensity profile of the collimated neutron beam. All instruments employed for the measurement of the neutron fluence and the energy distributions were described in detail elsewhere [12]. The energy distributions of neutrons entering and leaving the stack were determined by TOF spectrometry at distances of about 11 m from the target without and with the stack in its irradiation position. For the two lower neutron energies, a scintillation detector and for the higher energy a ^{238}U fission ionization chamber was used as a spectrometer. Earlier investigations showed that the results obtained with these two devices are in good agreement [12]. Recent investigations [14] show that the spectral fluence below the threshold of the spectrometer of about 4 MeV is essentially constant down to zero neutron energy. At the position of the stack the neutron fluence in the high-energy peak was determined with a proton recoil telescope [15] (PRT). This device is based on the detection of recoil protons produced by elastic neutron scattering on hydrogen in a high-purity polyethylene (PE) radiator foil of 1.5 mm in thickness. Cross sections for n-p scattering were taken from the phase-shift analysis "VL40" which has been recommended as standard cross section data [16]. The uncertainties of 5% for these data dominate the total uncertainty of the fluence measurements.

At TSL, the fluence of neutrons passing through the activation stack is monitored using $^{238}U(n,f)$ and $^{209}Bi(n,f)$ reactions as standards [16]. Thin film breakdown counters [17] (TFBC) are used for fission fragment detection. The principle of TFBC operation is based on the phenomenon of electrical breakdown in a track created by a fragment passing through a thin SiO_2 layer. Typically, four neutron monitors were used. A pair of monitors with ^{238}U (99.999% isotopic purity) and/or ^{209}Bi (>99.995% purity) targets was placed upstream the stack, and a similar pair downstream the stack. Each monitor consists of a fissile target and a fission fragment detector mounted close to each other like a sandwich. The monitors operate in a time-of-flight (TOF) mode. However, the short flight path (about 2 m) and unfavorable time structure of the beam at TSL does not allow an explicit separation of the high-energy peak in the neutron spectrum from the low-energy tail. As a consequence, the use of TOF technique is limited at the PARTY facility to the separation of monitor reaction events from the ones due to an intrinsic detector background as well as due to spontaneous and neutron-induced fission of heavier nuclei contamination in the monitor samples.

To obtain the high-energy peak neutron fluence from the monitor count rate, one needs independent information on the fraction of fission events due to peak neutrons. The latter is provided by additional measurements of TOF fission spectra for the same monitor reactions, with similar detector arrangements, in the same beam line but at a longer flight path in the marble hall (10-13 m). There, a much better separation of the high-energy peak (accuracy ~10 %) in the neutron spectrum from the low-energy tail is possible. Assuming that the neutron spectrum at the PARTY location at an emission angel of about 1^{o} is equal to the neutron spectrum at 0^{o} allows extracting the number of neutrons in the high-energy peak irradiating the target stack.

Residual radionuclides with half-lives between 20 min and 5 years were measured by off-line γ-spectrometry. In spite of the long irradiation times and high beam currents applied, the measurement of the irradiated targets is a low-count-rate problem and requires close-to-detector geometries. This results in particular problems with respect to efficiency determination and to necessary corrections for systematic coincidence. In addition, γ-self-absorption in the targets has to be corrected for. Therefore, a new method for the determination of detector efficiencies was developed which takes into account all these effects simultaneously.

The γ-spectrometry of the activated target foils yields activities proportional to production rates P [s^{-1} g^{-1}]. A production rate P of a nuclide produced is given by:

$$P = \frac{N_L}{A_T} \int \sigma(E) \cdot J(E)\, \mathrm{d}E \quad \text{with} \quad J(E) = \frac{\mathrm{d}^2 \Phi(E)}{\mathrm{d}E\, \mathrm{d}t} \tag{1}$$

with N_L being Avogadro's number and A_T the atomic mass of target element. $\sigma(E)$ are the neutron cross sections, $J(E)$ the spectral neutron flux densities and $\Phi(E)$ the neutron fluence. The integral is taken over all neutron energies E.

Cross sections cannot be directly calculated from these response integrals since the neutrons used are just "quasi-monoenergetic" with only about 30 to 50% of the neutrons in the high-energy peak with a width of a few MeV. The neutron cross sections $\sigma(E)$ have to be extracted from production rates P_i (i = 1,..., n) determined in a series of n irradiation experiments with different neutron energies by unfolding. To derive the final cross sections one requires a "guess" excitation function of the respective nuclear reaction, and the spectral fluences in different experiments are needed. This guess excitation function contains all eventually existing experimental neutron cross sections (mostly for $E < 20$ MeV, if at all). For energies or target elements for which no experimental data are available, theoretical neutron excitation functions are calculated as guess functions up to the highest neutron energies applied in the activation experiments by the ALICE-IPPE code [18]. The calculated excitation functions are normalized to the existing experimental cross sections, if available.

Information on the energy dependence of the neutron spectra in the targets was obtained by modeling the neutron spectra by Monte Carlo techniques using the LAHET/MCNP code system [19, 20]. These transport calculations started either from the experimentally determined neutron spectra (at UCL) or from the systematics of experimentally measured neutron emission spectra of the ^{7}Li(p,n)-reaction [21] (at TSL). The calculations described the transport of the neutrons into the target stacks and into the individual targets as well the production and transport of secondary particles which inside the massive target stacks cannot be neglected. The normalization of the spectra was done on the basis of either direct measurements of the spectra of the impinging neutrons (at UCL) or the neutron fluences measured by the TFBC-monitors (at TSL). Measurements at UCL with and without the target stacks in the beam allowed to determine the attenuation of the neutrons in the stacks and to validate respective calculations of this attenuation using the LAHET/MCNP [19, 20] codes (Fig. 1).

First reports of the performance of the PARTY facility and some results for short- and medium-lived γ-emitting product nuclides are published [11, 22]. From the target elements activated, excitation functions for a total of about 120 reactions will be obtained.

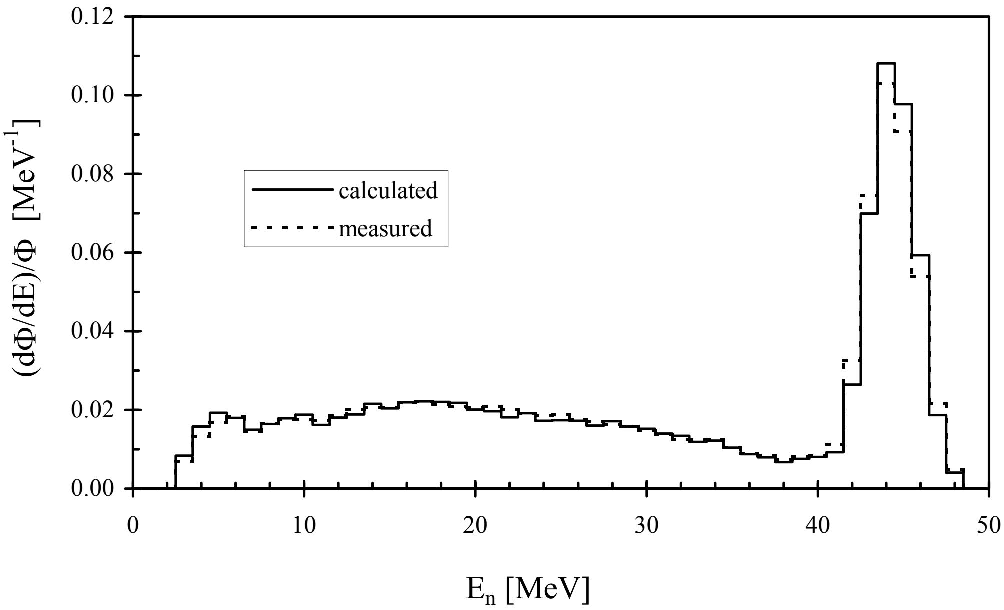

Fig. 1: Comparison of measured (broken line) and calculated (full line) neutron spectra behind the stack of the UCL experiment with E_p = 36.4 MeV. The spectra are normalized to a fluence 1 cm^{-2} of neutrons with E_n > 2.5 MeV.

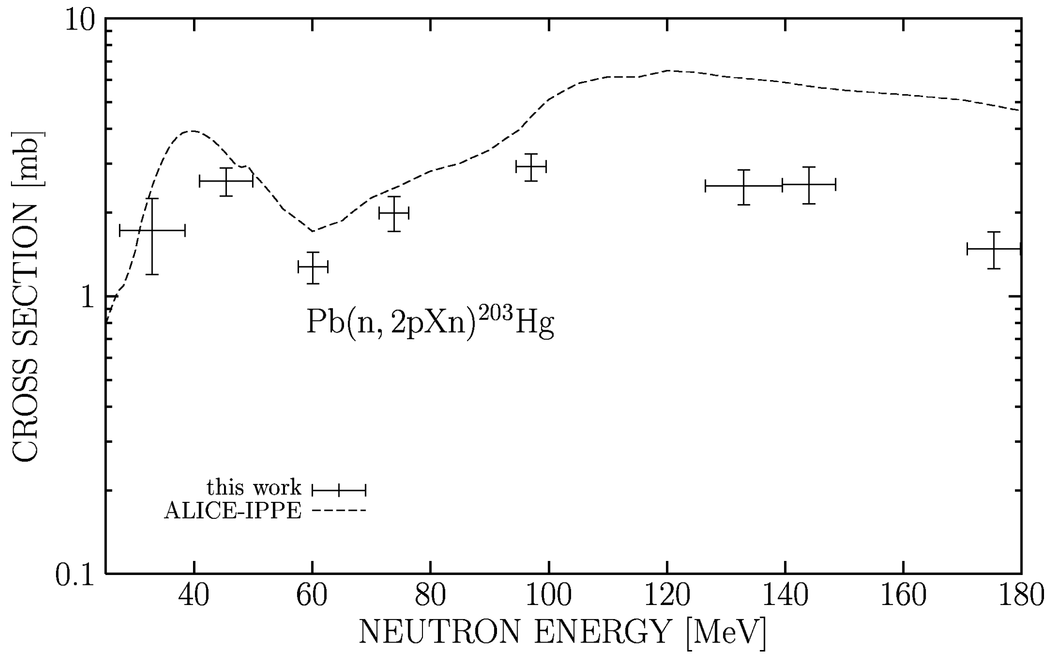

Fig. 2: Guess excitation function and final cross sections for the reaction $^{nat}Pb(n,2pxn)^{203}Hg$. The uncertainties in energy given represent the half-width of the peak in the neutron spectrum.

In Fig. 2, we present as an example the final cross sections and the initial guess excitation function for the $^{nat}Pb(n,2pxn)^{203}Hg$ reaction. Presently, work is going on to make use of the total information contained in the experimental production rates for a physically meaningful interpolation of the excitation functions between the data points and to further reduce the uncertainties of the cross sections.

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