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Cross Sections for the Production of Radionuclides by Proton-Induced Reactions on W, Ta, Pb and Bi from Thresholds up to 2.6 GeV

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Extending our earlier work on cross sections for the production of residual nuclides by proton-induced reactions, the production of radionuclides from the target elements Ta, W, Pb, and Bi was investigated from thresholds up to 2.6 GeV using accelerators at LNS/Saclay, PSI/Villigen, and TSL/Uppsala. Residual nuclides were measured by X- and γ-spectrometry. We obtained more than 5000 new individual cross sections for 402 reactions. Together with the yet not published cross sections for the target elements Rb, Mo, Rh, Ag, In, Te and La and with published results of special investigations of cosmogenic nuclides, our entire consistent data base now covers the target elements C, N, O, Mg, Al, Si, Ca, Ti, V, Mn, Fe, Co, Ni, Cu, Rb, Sr, Y, Zr, Nb, Mo, Rh, Ag, In, Te, Ba, La, Ta, W, Au, Pb and Bi. It contains data for about 1300 nuclear reactions and more than 24000 cross sections. Here, some phenomenological aspects of the residual nuclide production from heavy target elements are described and the capabilities of nuclear models to predict the respective cross sections are discussed.

KEYWORDS: p-induced reactions, residual nuclide production, heavy target elements, model calculations

I. Introduction

Integral excitation functions for the production of residual nuclides are basic quantities for the calculation of radioactive inventories of spallation targets in spallation neutron sources and in accelerator-driven devices for energy amplification or for transmutation of nuclear waste. In this context, we investigated the production of radionuclides by proton-induced reactions on lead,[1,2] partially within the EC Concerted Action "Physical Aspects of Lead as a Neutron Producing Target for Accelerator Transmutation Devices".[3] We extended this work by investigating the target elements Ta, W[4,5] and Bi.[6] Together with the earlier published data[7,8] they provide a database for 31 target elements.

Here, we concentrate on a systematic survey on the phenomenology of excitation functions for heavy target elements and report the results of some tests of nuclear models and codes describing medium-energy nuclear reactions. The phenomenology and energy dependence of excitation functions exhibits clearly distinguishable reaction modes like multifragmentation, three different types of medium-energy asymmetric and symmetric fission, deep spallation, preequilibrium reactions and classical compound nucleus reactions.

A comparison of the experimental data with theoretical ones calculated by the INC/E models incorporated into the HERMES[9] and LAHET[10] code systems and by preequilibrium codes such as AREL[11] and ALICE-IPPE[12] demonstrates the necessity for further improvements if medium-energy cross sections for applications shall be reliably predicted by theory. The new database can serve as a basis to scrutinize existing and to benchmark improved models and codes.

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II. Experimental

Integral cross sections for the production of residual radionuclides were determined by off-line γ- and X-ray-spectrometry of metal targets which were irradiated by protons at accelerators of the Laboratoire National Saturne (LNS) at Saclay (200 MeV – 2.6 GeV), of the The Svedberg Laboratory (TSL) at Uppsala (70 – 180 MeV) and of the Paul Scherrer Institute (PSI) at Villigen (< 70 MeV).

The experimental details have been given earlier.[1,7] We here just give a description of modifications compared to previous setups with respect to the measurements of flux densities and the evaluation of the activities.

For irradiations with energies above 60 MeV the flux monitoring was done offline via the ^{27}Al(p,3p3n)^{22}Na reaction. Because of its threshold energy of 22 MeV and the relatively steep slope below 60 MeV additionally flux monitoring was also done via the ^{nat}Cu(p,xn)^{65}Zn reaction. Both methods were used in our previous work[1,7] and gave consistent results in the overlapping energy range.

A newly developed procedure was used to improve the evaluation and calculation of cross sections. Now the decision whether a nuclide is considered to be present is based on checking the self-consistency of all influencing parameters. By supporting the interpretation of basic quantities used in the calculations with a graphical display systematic errors are minimized.[13]

The analysis of the experimental uncertainties were performed in the same way as described in detail previously.[1]

The redesign of the stack containers used at Laboratoire National Saturne (Saclay/France) drastically reduced the influence of secondary particles. In contrast to our earlier work[7] we minimized the total mass in each stack by irradiation of up to 30 small individual stacks with distances of 5–10 cm be-

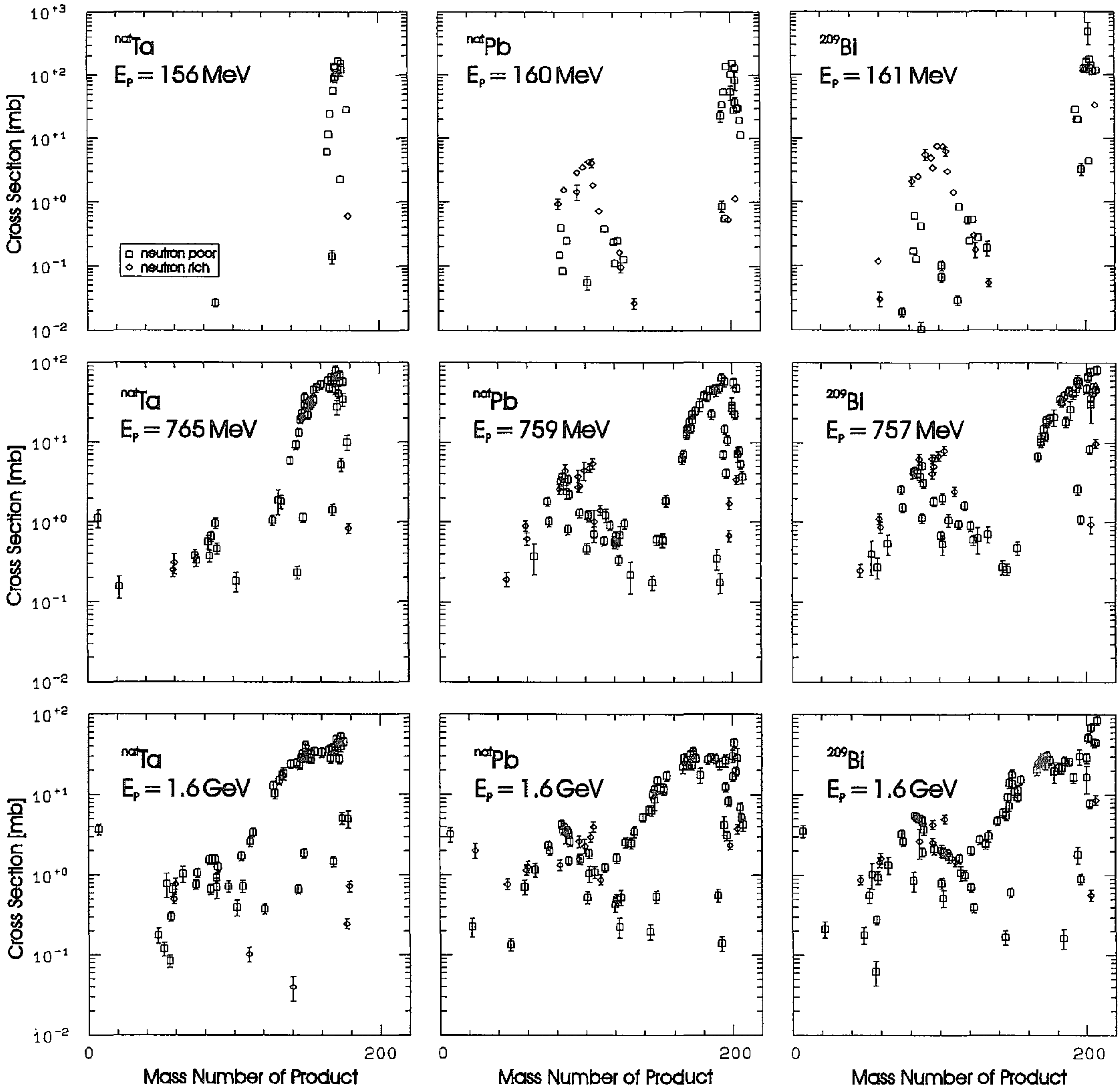

Fig. 1 Experimental production cross sections of proton-induced residuals from this work as a function of product mass number in Ta, Pb and Bi for proton energies of 160, 750 and 1600 MeV. Open squares correspond to neutron poor radionuclides and open diamonds indicate neutron rich radionuclides. With respect to the new data for Tungsten see Miah *et al.*, these proceedings.[5)]

tween.[1)]

With the new stack arrangement no further corrections concerning the influence of secondary particles have been necessary. The results of different irradiations with overlapping energy ranges particulary near threshold with steep slopes in the excitation function are in excellent agreement with each other. This demonstrates the accuracy of the calculated proton energies as well as of the energy dependence of the cross sections determined.

III. Experimental Results

For the target elements W, Ta, Pb and Bi we obtained more than 5000 new individual cross sections for 402 proton-induced reactions from thresholds up to 2.6 GeV. The measured cross sections vary between a few μbarn and several barns. The results have been independently determined at Cologne and Hanover and are in excellent agreement with each other over the whole energy range. Strong emphasis has been laid upon strict quality assurance procedures. The results from all irradiations at several accelerators form con-

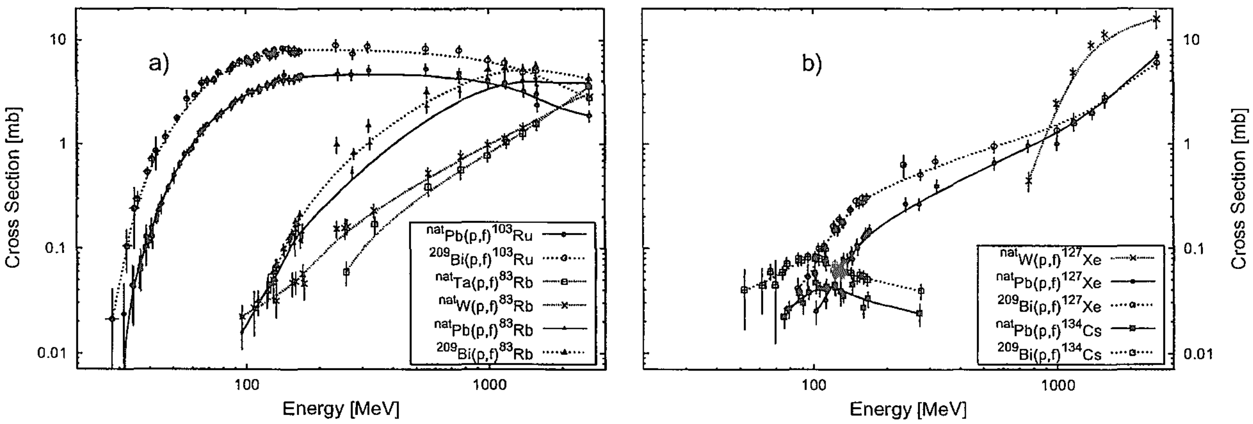

Fig. 2 Excitation functions from this work for the production of fission products ^{103}Ru from Pb and Bi and ^{83}Rb from Ta, W, Pb and Bi (a) and ^{127}Xe from W, Pb and Bi and ^{134}Cs from Pb and Bi (b). The lines are eyeguides only.

sistent excitation functions despite extremely different experimental conditions.

Figure 1 gives an overview of the experimental cross sections as a function of the product masses at energies of 160, 750 and 1600 MeV for the target elements Ta, Pb and Bi. One clearly realizes the growing importance with increasing energy of reaction modes such as spallation, medium-energy fission and deep-spallation and/or multi-fragmentation.

IV. Model Calculations

Theoretical excitation functions for proton energies between 100 MeV and 3 GeV were calculated using the INC/E code systems HERMES[9] and LAHET,[10] for proton energies below 200 MeV with the preequilibrium codes AREL[11] and ALICE-IPPE.[12] The details of the calculations with the INC/E models have already been given,[1] a comprehensive analysis of the parameter dependence will be published later. The calculations with ALICE-IPPE were performed for all mass-parameters and with both level density formulas. The preequilibrium-compound nuclear model ALICE-IPPE includes a preequilibrium-emission of complex particles like deuterons or alphas which improved the theoretical results for energies below 200 MeV drastically.

V. Discussion

The new results for the target elements W, Ta, and Bi and their theoretical analysis confirm the conclusions published earlier.[1,14] The theoretical calculations show severe deficits for spallation products, fail completely in case of fission products and do not at all describe the production of fragmentation products ($A < 20$). The new database covering a broad range of target elements and the complete energy range between thresholds and 2.6 GeV offers a unique chance to test improved models and codes.

Presently, we restrict our discussion to the phenomenology of the excitation functions. In **Figure 2**, we demonstrate the different types of medium-energy fission observed in our data. Generally, symmetric fission prevails. For Pb and Bi, neutron-rich radionuclides such as ^{103}Ru are produced with thresholds well below 100 MeV. Their excitation functions show a broad plateau above 100 MeV and decrease for energies above 600 MeV. We interpret this as fission at relatively low excitation energies after a short intranuclear cascade. The absolute cross sections differ by a factor of two for Pb and Bi. Neutron-poor radionuclides such as ^{83}Rb have significantly higher thresholds and their excitation functions increase monotonically with increasing proton-energies. A strong dependence on the target element is also seen in this case. We explain these excitation functions by a symmetric fission process after a relatively long intranuclear cascade and subsequent neutron evaporation followed by fission.

Besides symmetric fission, also asymmetric modes are clearly distinguished (**Figure 2b**). The neutron-rich fission product ^{131}Cs is observed only at relatively low proton-energies for the target elements Pb and Bi with a maximum of the excitation functions at about 100 MeV. For higher energies, this fission mode rapidly disappears. The neutron-poor fission product ^{127}Xe from Pb and Bi clearly shows the transition with decreasing proton-energies from production by spallation to that by fission. For the target element tungsten only production of ^{127}Xe by spallation is observed.

Besides fission and spallation to product masses larger than 0.5 target mass, deep-spallation to products with masses ($20 < A < 65$) is observed. As shown exemplarily in **Figure 3**, also these product nuclides exhibit excitation functions increasing monotonically with proton-energy, the absolute cross sections decreasing with the differences between target and product masses.

From the systematics of mass distributions according to **Figure 1**, from the energy dependencies as revealed by **Figure 3**, and from direct measurements of fission products[15] we conclude that the production of these nuclides can only be explained by deep-spallation.

Fragmentation products ($A < 20$) are clearly seen in the mass distributions and in the excitation functions. They are characterized by increasing cross sections with decreasing product mass numbers and by excitation functions increasing

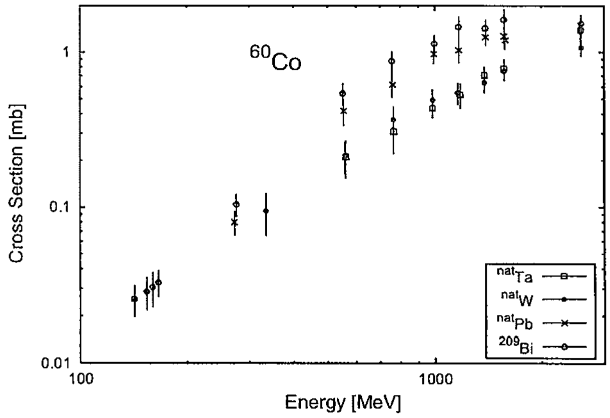

Fig. 3 Experimental cross sections from this work for the production of ^{60}Co from Ta, W, Pb and Bi.

monotonically with proton-energies. Their production cross sections are nearly independent of target masses.

VI. Conclusions

In extension of the recently published cross sections data for the proton-induced production of residual radionuclide in lead we present new data for the target elements W, Ta and Bi. They are not only essential for the calculation of radioactive inventories in accelerator driven systems with heavy targets, but also provide the experimental basis for the evaluation of new benchmarks for computer simulations of intermediate nuclear reactions. To completely describe the quality of those calculations one needs cross sections for energies from thresholds up to several GeV. Up to now the experimental investigations of intermediate energy radionuclide formation in heavy targets cannot be replaced by existing model calculations.

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