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Nuclear reaction modeling for RIA ISOL target design

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Los Alamos scientists are collaborating with researchers at Argonne and Oak Ridge on the development of improved nuclear reaction physics for modeling radionuclide production in ISOL targets. This is being done in the context of the MCNPX simulation code, which is a merger of MCNP and the LAHET intranuclear cascade code, and simulates both nuclear reaction cross sections and radiation transport in the target. The CINDER code is also used to calculate the time-dependent nuclear decays for estimating induced radioactivities. We will give an overview of the reaction physics improvements we are addressing, including intranuclear cascade (INC) physics, where recent high-quality inverse-kinematics residue data from GSI have led to INC spallation and fission model improvements; and preequilibrium reactions important in modeling (p, xn) and $(p, xnyp)$ cross sections for the production of nuclides far from stability.

Reliable nuclear models, codes, and data libraries are required for modeling the target/blanket assembly of the Rare Isotope Accelerator (RIA) ISOL facility and other applications. To meet these needs, several nuclear reaction models and data libraries have been developed at Los Alamos National Laboratory in collaboration with researchers at Argonne and Oak Ridge and implemented further in the Monte Carlo transport code systems MCNPX [1,2] and LAHET [4–6] and in the transmutation inventory code CINDER'90 [8].

Between the recent work resulting in a significant improvement of the LANL capacity to predict spallation and fission cross sections at incident energies up to several GeV and reliably simulate target/blanket assemblies we want to mention first an upgrade of the MCNPX transport code [1] and a recent realization of its Version 2.1.5 [2], representing the efforts of many people over many years, including a Beta Test Team of about 500 users internationally. The MCNPX code development project involves the formal extension of MCNP4B [3] to all particles and all energies.

MCNPX incorporates all of the basic LAHET nuclear modules [4–7]. Physics modules from FLUKA [9] high-energy code are also included.

MCNPX also includes a number of features intended to facilitate applications in complex simulations. In addition to extensions of the familiar MCNP tallies to other particle types and to wider energy ranges, a geometry-independent mesh tally and a radiography simulation tally have recently been incorporated into the code. Coupled with transmutation codes such as CINDER'90 [8], the full neutronic history of a system may be obtained with MCNPX.

Three important recent developments in transport physics are being incorporated into MCNPX:

1) A suite of evaluated reaction cross section files collectively known as the LA150 Library has been developed [10,11] in support of accelerator-driven systems design and implemented into MCNPX [12]. These evaluations are in ENDF-6 format, and have recently been accepted into the U.S.-standard ENDF/B-VI Library as Release-6. For incident neutrons, they extend the previously-existing ENDF/B-VI information from 20 MeV up to 150 MeV. For incident protons, the files extend from 1–150 MeV. To date, evaluations have been completed for isotopes of the following structural, shielding, and target-blanket materials: H, Li, C, N, O, Al, Si, P, Ca, Fe, Ni, Cr, Cu, Nb, W, Hg, Pb, and Bi. The primary motivation for using these evaluated data is the accuracy improvements that one can expect to obtain in the below-150 MeV energy region. In most previous transport simulations, intranuclear-cascade methods have been used for neutrons above 20 MeV and for protons at all energies, even though the semiclassical assumptions inherent within such models do not hold at lower energies. By developing evaluated cross section libraries up to 150 MeV, using state-of-the-art nuclear reaction models in the GNASH code as well as experimental data, one can expect to have the most accurate possible representation of the nuclear cross sections. The nuclear models used are based on theoretical approaches that are appropriate for the energies in the few-MeV to 150 MeV range: the Hauser-Feshbach compound nucleus theory; preequilibrium calculations based on the Feshbach-Kerman-Koonin theory or the exciton model; direct reactions calculated from the optical model using collective excitation form factors; and elastic scattering from the optical model. The GNASH code was demonstrated to be one of the most accurate codes available for model calculations below 150 MeV in a Nuclear Energy Agency code intercomparison [13]. The optical model is used for predictions of the total, reaction, and elastic scattering cross sections, making use of nucleon potentials at higher energies developed by Madland [14], Chiba, and Koning. It is particularly useful for accurately representing the angular distributions in elastic scattering, allowing more accurate neutron transport simulations. (Many previous intranuclear cascade transport codes instead represent elastic scattering using a black-disc diffraction formula or use even simpler approaches, which poorly approximate reality below a few hundred MeV.)

2) For the first time in MCNP or MCNPX, data for the generation of photo-induced neutrons and charged secon-

daries are available. MCNPX has been extended to use the newly available tabular data. Specifically, the distance to the next photon collision reflects the possibility of a photonuclear collision, and such collisions will produce a combination of neutrons, photons or light ions ($A \leq 4$) for further transport. Thus, photonuclear events are fully integrated within a simulation. The implementation has been subjected to verification and validation testing [15]. Limitations of this extension are the small number (12) of isotopes for which photonuclear cross sections were implemented into MCNPX, and the energy range of up to 150 MeV that covers the physics from the threshold through the giant resonance region before pion production phenomena become important. (In the very near future cross section sets for many more isotopes will be made available building on data provided by the IAEA.) To eliminate these limitations, a photonuclear physics option was recently implemented into MCNPX [16] by the extending of the CEM97 module [17,18] in MCNPX to describe photonuclear reactions.

3) Last but not least, the essential physics of the Cascade-Exciton Model (CEM) [20,21] with the recent developments and improvements as realized in the code CEM97 [17,18] has now been included in MCNPX. Recent developments of the CEM motivated by new data on isotope production measured recently in "reverse kinematics" at GSI for interactions of ^{208}Pb and ^{238}U at 1 GeV/nucleon and ^{197}Au at 800 MeV/nucleon with liquid ^{1}H leads us to CEM2k [19]. CEM2k is a next step in the improvement of the CEM; it differs from CEM97 [17,18] mainly in the details of the transitions from the cascade stage of a reaction to the preequilibrium one, and from the latter to equilibrium decay. This preliminary version of CEM2k has less preequilibrium emission than the earlier versions. CEM2k is still under development and will be incorporated into MCNPX in several months.

Two typical examples of predictive powers of the LANL codes for cross sections of radioactive and stable isotopes produced in p (1 GeV) + ^{208}Pb interactions are given in Figs. 1 and 2. We chose this reaction not only because Pb is of interest for RIA and for the Pb-Bi technology preferable for ATW facilities, but mainly because these data [22,23] are of the best quality and completeness among all other similar data on proton-induced spallation cross sections measured by now worldwide: The data by Wlazlo et al. [22] were measured recently at GSI in the inverse kinematics, i.e., from spallation of ^{208}Pb on proton at $1{\cdot}A$ GeV, down to 0.1 mb with a high accuracy, for all radioactive and stable isotopes produced, and the radioisotope data by Titarenko et al. [23] were measured recently at ITEP, Moscow by an international team, using the best technique, software, and monitoring reactions available at present for the direct γ-spectrometry, therefore, are believed to be of the highest reliability for the γ-spectrometry method.

One can see that both CEM2k and LAHET, either with ISABEL or Bertini options, describe well these new data. In Fig. 2, beside curves connecting open symbols with nuclide yields calculated by six different modern available codes together with the experimental data shown by the curve connecting the black symbols, are given as well the mean squared simulated-to-experimental data ratios, $< F >$, averaged over all nuclides used in comparison, for every of the six codes. This mean deviation factor, $< F >$, may be regarded as a measure of a general, mean agreement of all calculated cross sections with the data, averaged over all residual isotopes produced. One can see that CEM2k has the lowest values of $< F >$ among other codes and agrees the best with the data. LAHET along with the Obninsk code CASCADE/INPE, specially modified to describe such data [23], are also in a very good agreement with these data.

Several examples of excitation functions for the reaction p + Ta calculated with the code CEM97 [17] compared with available experimental data from the compilation by Iljinov et al. [24], known in the literature as NUCLEX, are shown in Fig. 3. Ta is of interest for RIA as it is commonly used as a target. Unfortunately, the experimental data for spallation cross sections from p + Ta are much more scarce, old, and not such reliable as the excellent data for Pb discussed above. One can see that the code CEM97 describes these excitation functions quite well, though it is difficult to make a satisfactory comparison in this case, as the data were obtained in different old measurements and are too sparse.

A brief description of several other recent enhancements in the MCNPX may be found in our recent paper [25]. Another forthcoming enhancement of the code will be its ability to more accurately model light ion induced reactions for RIA applications. In the future we will continue our collaboration with other RIA researches so that MCNPX can play a strong role in RIA ISOL target design.

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[1] H. G. Hughes, R. E. Prael, and R. C. Little, "MCNPX - The LAHET/MCNP Code Merger," Technical Report XTM-RN-97-012(U), Los Alamos National Laboratory, Los Alamos, 1997.

[2] Laurie S. Waters, Editor, "MCNPX^{TM} User's Manual, Version 2.1.5," Los Alamos National Laboratory Report TPO-E83-G-UG-X-00001, Los Alamos, 1999.

[3] J. F. Briesmeister, Ed., "MCNP^{TM} – A General Monte Carlo N-Particle Transport Code," Los Alamos National Laboratory Report LA-12625-M, Version 4B, Los Alamos, 1997; http://www-xdiv.lanl.gov/XCI/PROJECTS/MCNP/manual.html.

[4] R. E. Prael and H. Lichtenstein, "User Guide to LCS: The LAHET Code System," Los Alamos National Laboratory Report LA-UR-89-3014 (1989); http://www-xdiv.lanl.gov/XTM/lcs/lahet-doc.html.

[5] R. E. Prael, "Primary Beam Transport Methods in LAHET^{TM}," Los Alamos National Laboratory Report LA-UR-99-0667, Los Alamos, 1999.

[6] R. E. Prael and D. G. Madland, "The LAHET Code System with LAHET2.8," Los Alamos National Laboratory Report LA-UR-00-2140 (2000); http://www-xdiv.lanl.gov/XTM/lcs/lahet-doc.html.

[7] R. E. Prael, "A New Nuclear Structure Library for MCNPX^{TM} and LAHET3^{TM}," Proc. 4th Int. Topical Meeting on Nucl. Applications and Accelerator Technology (AccApp'00), Nov. 12-15, 2000, Washington, DC, American Nuclear Society, La Grange Park, IL, 2001, pp. 350-352.

[8] W. B. Wilson ans T. R. England, "Status of CINDER'90 Codes and Data," Proc. 4th Workshop on Simulating Accelerator Radiation Environments (SARE4), Sept. 14-16, 1998, Knoxville, TN, ed. T. A. Gabriel (ORNL, 1999), pp. 69-79.

[9] P. A. Aarino *et al.*, "FLUKA89," Consiel Europeene Organisation pour La Recherche Nucleaire informal report (January 2, 1990).

[10] M. B. Chadwick *et al.*, Nucl. Sci. Eng. **131**, 293 (1999).

[11] A. J. Koning, T. Fukahori, and A. Hasegawa, Technical Report No. NEA/WPEC-13, Nuclear Energy Agency, Paris.

[12] M. B. Chadwick, H. G. Hughes, R. C. Little, E. J. Pitcher, P. G. Young, "Nuclear Models and Data for Accelerator-Driven Systems," Proc. 4th Int. Topical Meeting on Nucl. Applications and Accelerator Technology (AccApp'00), Nov. 12-15, 2000, Washington, DC, American Nuclear Society, La Grange Park, IL, 2001, pp. 297-301.

[13] M. Blann, H. Gruppelaar, P. Nagel, and J. Rodens, *International Code Comparison for Intermediate Energy Nuclear Data* (Organization for Economic Cooperation and Development Nuclear Energy Agency, Paris, 1994), pp. 1–206.

[14] D. G. Madland, in *Proc. of a Specialists Meeting on Preequilibrium Reactions*, Semmering, Austria, 1988, ed. B. Strohmaier (OECD Nuclear Energy Agency, Paris, France, 1988), No. NEANDC-245, pp. 103–116.

[15] M. C. White, "Development and Implementation of Photonuclear Cross-Section Data for Mutually Coupled Neutron-Photon Transport Calculations in the Monte Carlo N–Particle (MCNP) Radiation Transport Code," Los Alamos National Laboratory Report LA-13744-T (July 2000).

[16] Franz X. Gallmeier and Stepan G. Mashnik, "Photonuclear Physics Modules for MCNPX," Proc. 4th Int. Topical Meeting on Nucl. Applications and Accelerator Technology (AccApp'00), Nov. 12-15, 2000, Washington, DC, American Nuclear Society, La Grange Park, IL, 2001, pp. 342-349.

[17] S. G. Mashnik and A. J. Sierk, "Improved Cascade-Exciton Model of Nuclear Reactions," Proc. 4th Workshop on Simulating Accelerator Radiation Environments (SARE4), Sept. 14-16, 1998, Knoxville, TN, ed. T. A. Gabriel (Oak Ridge: ORNL, 1999), pp. 29-51; http://xxx.lanl.gov/ps/nucl-th/9812069.

[18] A. J. Sierk and S. G. Mashnik "Modeling Fission in the Cascade-Exciton Model," Proc. 4th Workshop on Simulating Accelerator Radiation Environments (SARE4), Sept. 14-16, 1998, Knoxville, TN, ed. T. A. Gabriel (Oak Ridge: ORNL, 1999), pp. 53-67; http://xxx.lanl.gov/ps/nucl-th/9812070.

[19] S. G. Mashnik and A. J. Sierk, "CEM2k – Recent Developments in CEM," Proc. 4th Int. Topical Meeting on Nucl. Applications and Accelerator Technology (AccApp'00), Nov. 12-15, 2000, Washington, DC, American Nuclear Society, La Grange Park, IL, 2001, pp. 328-141; http://xxx.lanl.gov/ps/nucl-th/0011064.

[20] K. K. Gudima, S. G. Mashnik, and V. D. Toneev, Nucl. Phys. **A 401** 329 (1983).

[21] S. G. Mashnik, "User Manual for the Code CEM95", JINR, Dubna, 1995; OECD Nuclear Energy Agency Data Bank, Paris, France, 1995; http://www.nea.fr/abs/html/iaea1247.html; RSIC-PSR-357, Oak Ridge, 1995.

[22] W. Wlazlo, T. Enqvist, P. Armbruster, J. Benlliure, M. Bernas, A. Boudard, S. Czájkowski, R. Legrain, S. Leray, B. Mustapha, M. Pravikoff, F. Rejmund, K.-H. Schmidt, C. Stéphan, J. Taieb, L. Tassan-Got, and C. Volant, Phys. Rev. Lett. **84**, 5736 (2000).

[23] Yu. E. Titarenko, O. V. Shvedov, V. F. Batyaev, E. I. Karpikhin, V. M. Zhivun, A. B. Koldobsky, R. D. Mulambetov, D. V. Fischenko, S. V. Kvasova, A. N. Sosnin S. G. Mashnik, R. E. Prael, A. J. Sierk, T. A. Gabriel, M. Saito, and H. Yasuda, "Cross sections for nuclide production in 1 GeV proton-irradiated "Study of Residual Product Nuclide Yields in GeV Proton-Irradiated ^{208}Pb," LANL Report LA-UR-00-4779 (2000); http://xxx.lanl.gov/ps/nucl-th/0011083; submitted to Phys. Rev. C.

[24] A. S. Iljinov, V. G. Semenov, M. P. Semenova, N. M. Sobolevsky, and L. V. Udovenko, *Production of Radionuclides at Intermediate Energies*, Springer Verlag, Landolt-Börnstein, New Series, subvolumes **I/13c** (1993) and **I/13d** (1994).

[25] H. G. Hughes, M. B. Chadwick, R. K. Corzine, H. W. Egdorf, F. X. Gallmeier, R. C. Little, R. E. MacFarlane, S. G. Mashnik, E. J. Pitcher, R. E. Prael, A. J. Sierk, E. C. Snow, L. S. Waters, M. C. White, and P. G. Young, "Status of the MCNPX Transport Code," Los Lalmos National Report LA-UR-004942 (2000); Proc. Int. Conf. on Advanced Monte Carlo for Radiation Physics, Particle Transport Simulation and Application (MC 2000), 23-26 October, 2000, Lisbon, Portugal.

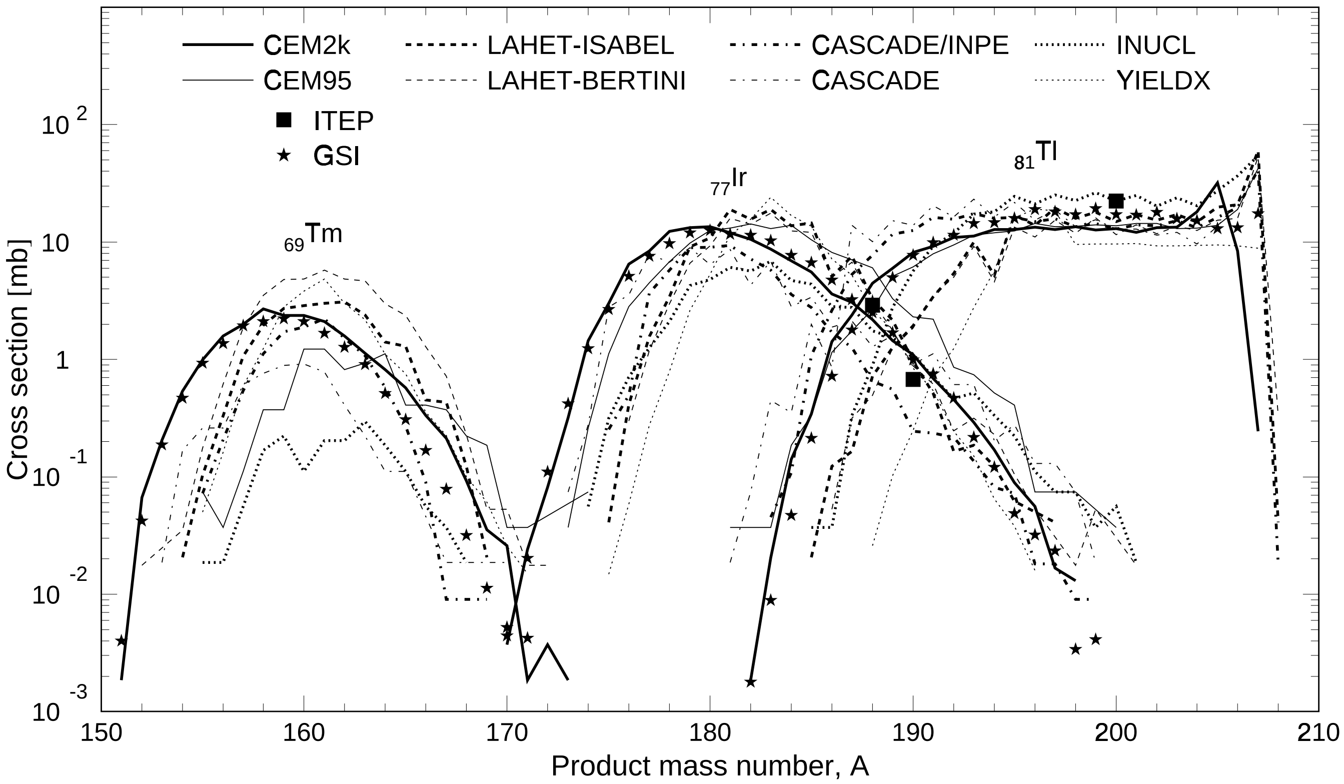

FIG. 1. Isotopic mass distribution for independent products of Tm, Ir, and Tl isotopes. Black squares are ITEP measurements [23], while filled stars show GSI data obtained in reverse kinematics [22]. Results from different codes are marked as indicated.

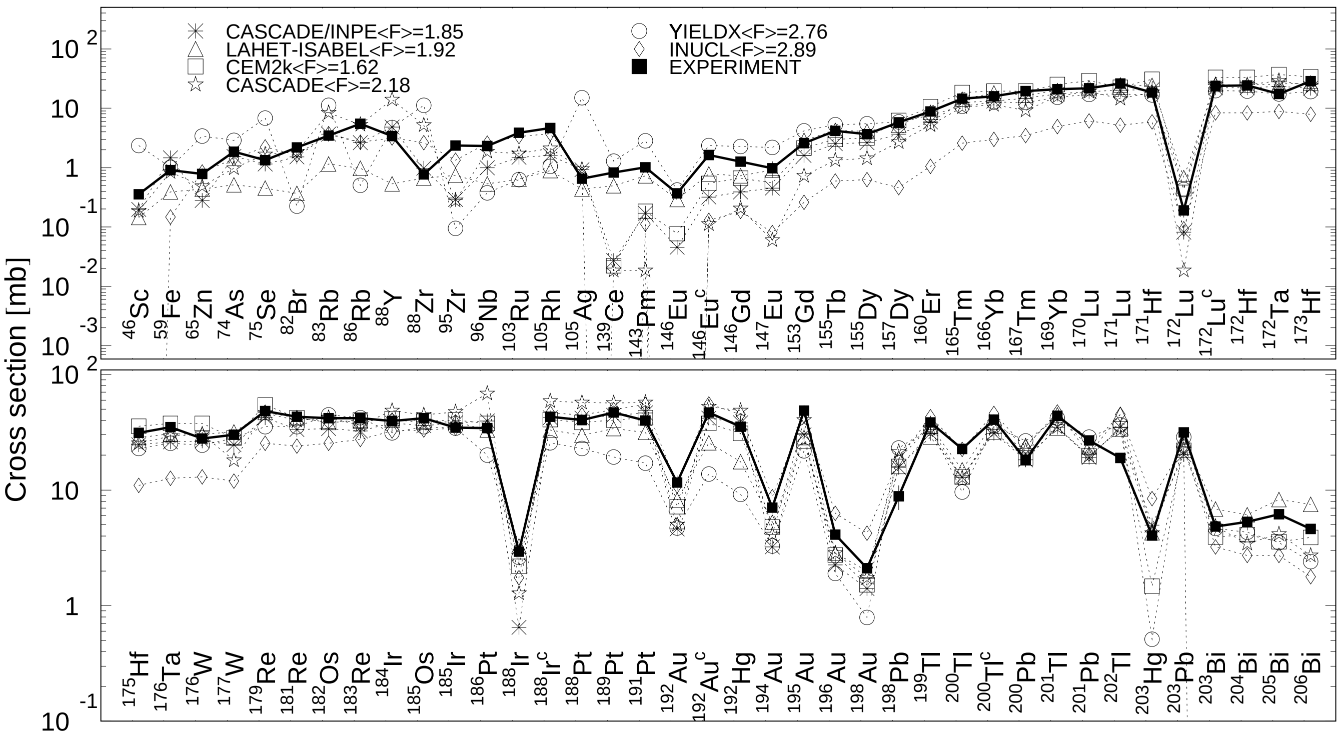

FIG. 2. Detailed comparison between experimental [23] and simulated yields of radioactive reaction products. The cumulative yields are labeled with a "c" when the respective independent yields are also shown.

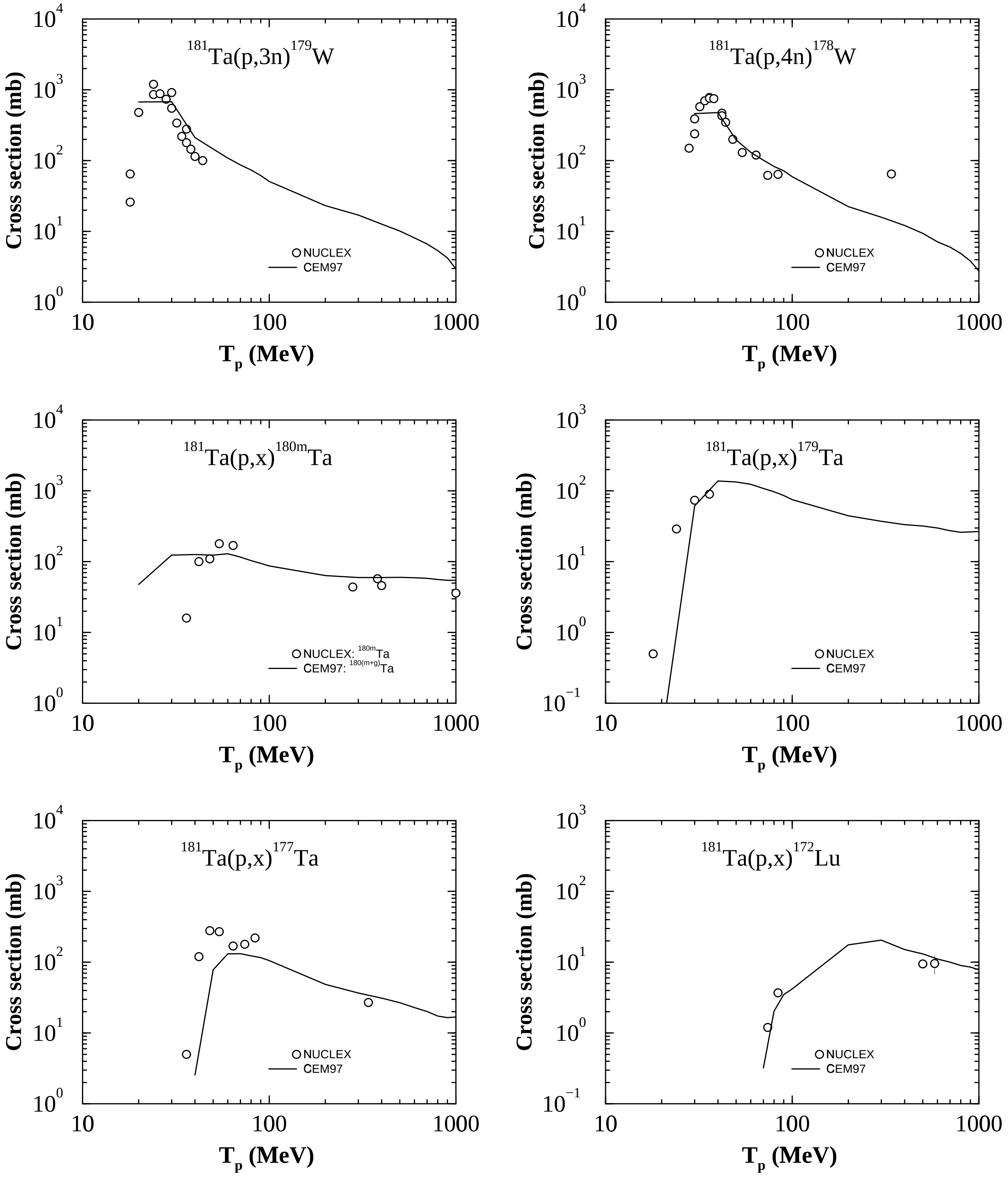

FIG. 3. Samples of data from the compilation NUCLEX [24] and calculated with the code CEM97 [17,18] excitation functions for the reaction p + Ta.