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Title: Analysis of the JINR p(660 MeV) I129, Np237, and Am241
Measurements with Eleven Different Models

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ANALYSIS OF THE JINR $p(660 \text{ MeV}) + {}^{129}\text{I}$, ${}^{237}\text{Np}$, AND ${}^{241}\text{Am}$ MEASUREMENTS WITH ELEVEN DIFFERENT MODELS

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Abstract

Isotopically enriched ${}^{129}\text{I}$ (85% ${}^{129}\text{I}$ and 15% ${}^{127}\text{I}$), ${}^{237}\text{Np}$, and ${}^{241}\text{Am}$ targets were irradiated with a beam of 660-MeV protons at the JINR DLNP Phasotron and cross sections of formation of 207 residual products (74 from ${}^{129}\text{I}$, 53 from ${}^{237}\text{Np}$, and 80 from ${}^{241}\text{Am}$) were determined using the γ -spectrometry method. Here, we analyze all these data using eleven different models, realized in eight codes: LAHET (Bertini, ISABEL, INCL+ABLA, and INCL+RAL options), CASCADE, CEM95, CEM2k, LAQGSM+GEM2, CEM2k+GEM2, LAQGSM+GEMINI, and CEM2k+GEMINI, in order to validate the tested models against the experimental data and to understand better the mechanisms for production of residual nuclei. The agreement of different models with the data varies quite a bit. We find that most of the codes are fairly reliable in predicting cross sections for nuclides not too far away in mass from the targets, but differ greatly in the deep spallation, fission, and fragmentation regions. None of the codes tested here except GEMINI allow fission of nuclei as light as iodine, therefore the best agreement with the ${}^{129}\text{I}$ data, especially in the $A=40-90$ region, is shown by the codes CEM2k and LAQGSM when they are merged with GEMINI. At the same time, GEMINI is not yet very reliable for an accurate description of actinides and the ${}^{237}\text{Np}$ and ${}^{241}\text{Am}$ data are reproduced better by LAHET (Bertini, ISABEL, or INCL+RAL/ABLA options), and by CEM2k and LAQGSM merged with GEM2 and not so well when using GEMINI. We conclude that none of the codes tested here are able to reproduce well all these data and all of them need to be further improved; development of a better universal evaporation/fission model should be of a high priority.

Introduction

Interest in the physics of transmutation (i.e., conversion into stable isotopes as a result of nuclear reactions) of actinides and fission products produced at nuclear power stations has increased significantly during the last decade. Estimations made by different groups [1, 2] show that the radiation risk of the spent nuclear fuel due to its possible leakage from deep underground storage systems after its transmutation is about the same as of the uranium ore after 1000 years of storage, that is significantly shorter than 5×10^6 years necessary to store the same spent fuel without transmutation to decrease its risk to a similar level.

Analysis of the radiation hazard of the spent fuel showed that after the extraction of the uranium-plutonium group of elements and such fission products like ^{90}Sr , ^{137}Cs , and ^{129}I , the highest hazard comes from ^{237}Np and ^{241}Am [3]. At that, ^{241}Am ($T_{1/2} = 432.2$ years) contributes the most to the radioactivity, while ^{237}Np ($T_{1/2} = 2.144 \times 10^6$ years) is dangerous because of its high concentration in the spent fuel and its high migration ability in the biosphere, that increases the probability of its penetration into human body through the food chain [4].

Investigation of ^{237}Np and ^{241}Am transmutation dynamics in the flow of thermal neutrons of different densities shows that the higher the density of neutrons, the smaller the number of different actinides noticeably contributing to the radioactivity of wastes [4]. To solve the problem of transmutation, high-current proton accelerators can be used to produce neutron fluxes of $\sim 10^{17} \text{ cm}^{-2}\text{s}^{-1}$ for transmutation purposes. In some recent publications, both transmutation of actinides by thermal neutron irradiation and their spallation and fission with the proton and ion beams are investigated [5].

Hadron-nucleus event generators are the basis for calculations of the Accelerator Driven System (ADS) setups, their targets, and the blanket effect. Such calculations are done using models of different accuracy. The best test for different models and codes used in such applications is to compare calculated and experimental yields of the residual nuclei from reactions of interest. From experimental point of view, determination of the independent cross-section for yields of short-lived nuclear products from mono-isotope targets is the most important for such comparisons [6]. Experimental cross-sections for residual nuclei in radioactive ^{129}I , ^{237}Np , and ^{241}Am targets are undeniably important for the projects of transmutation of nuclear wastes in a direct proton beam. Measurements of the yield of residual nuclei from ^{237}Np , ^{241}Am , and ^{129}I (85% ^{129}I and 15% ^{127}I) targets were recently performed at the JINR Phasotron with proton beams of 660 MeV [7, 8]. In the present work, we analyze these measurements with eleven models implemented in several event generators and transport codes used in different nuclear applications, to test these models against the experimental data and with a hope to understand better mechanisms of nuclear reactions and ways to improve the models and codes.

Results

The ^{237}Np and ^{241}Am experimental data are published in tabulated form in Ref. [7], while the ^{129}I data are tabulated in [8]. Details on the measurements may be found in [7, 8] and we do not discuss them here.

We analyzed all the measured data using eleven models, contained in eight transport codes and event generators. Namely, we calculated the reactions with the LAHET3 version [9] of the transport code LAHET [10] using the Bertini [11] and ISABEL [12] IntraNuclear-Cascade

(INC) models merged with the Dresner evaporation model [13] and the Atchison fission model (RAL) [14], and using the Liege INC code by Cugnon *et al.* INCL [15] merged in LAHET3 with the ABLA [16] and with Dresner [13] (+ Atchison [14]) evaporation (+ fission) models, with the Dubna transport code CASCADE [17], with versions of the Cascade-Exciton Model (CEM) [18] as realized in the codes CEM95 [19] and CEM2k [20], with CEM2k merged [21]-[23] with the Generalized Evaporation/fission Model code GEM2 by Furihata [24], with the Los Alamos version of the Quark-Gluon String Model code LAQGSM [25] merged [21]-[23] with GEM2 [24], as well as with versions of the CEM2k and LAQGSM codes merged both [21] with the sequential-binary-decay code GEMINI by Charity [26]. The limited size of the present work does not allow us to discuss these models here; description of the models may be found in the original publications [9]-[26] and references therein.

Let us start with discussing results for the ^{129}I target. As we have done previously (see, *e.g.*, [6, 27]), we choose here one qualitative and one quantitative criterion to judge how well our data are described by different models; namely, the ratio of calculated cross section for the production of a given isotope to its measured values $\sigma^{cal}/\sigma^{exp}$ as a function of the mass number of products (Fig. 1), and the mean simulated-to-experimental data ratio (Table 1)

$$\langle F \rangle = 10 \sqrt{\langle (\log[\sigma^{cal}/\sigma^{exp}])^2 \rangle}, \quad (1)$$

with its standard deviation :

$$S(\langle F \rangle) = 10 \sqrt{\langle (|\log(\sigma^{cal}/\sigma^{exp})| - \log(\langle F \rangle))^2 \rangle}. \quad (2)$$

For such a comparison, out of all the 74 measured cross sections [8], only 42 were selected to satisfy some rules based on appreciation of the physical principles realized in the models. For instance, if only a long-lived isomer or only the ground state of a nuclide was measured, such nuclides were excluded from the quantitative comparison, but if both were measured separately, their sum was compared with calculations. Such rules are essentially similar to those used by Titarenko *et al.* [6, 27].

To understand how different models describe nuclides produced in the spallation and fission or fragmentation regions, we divided all 42 measured nuclides included in our quantitative comparison into two groups, spallation ($A \geq 95$) and fission/fragmentation ($A < 95$). The left panel of Tab. 1 shows values of $\langle F \rangle$ and $S(\langle F \rangle)$ for all compared products (both spallation and fission/fragmentation), while the right panel of this table shows such results only for spallation; N is the total number of comparisons, $N_{30\%}$ is the number of comparisons in which calculated and measured values differ by not more than 30 %, while $N_{2.0}$ shows the number of comparisons where the difference was not more than a factor of two.

We note that the codes CEM95 [19] and CEM2k [20] consider only competition between evaporation and fission of excited compound nuclei and calculate the fission cross sections for a nuclear reaction on a heavy nucleus, but do not calculate the fission fragments, as they do not contain a fission model. The Bertini [11] and ISABEL [12] INC's are used in our calculations with the default options of LAHET3 for evaporation/fission models; they consider evaporation with the Dresner code [13] and a possible fission of heavy compound nuclei using the Atchison RAL fission model [14], but only if they are heavy enough ($Z > 71$), *i.e.*, they do not consider fission for such light targets as ^{129}I .

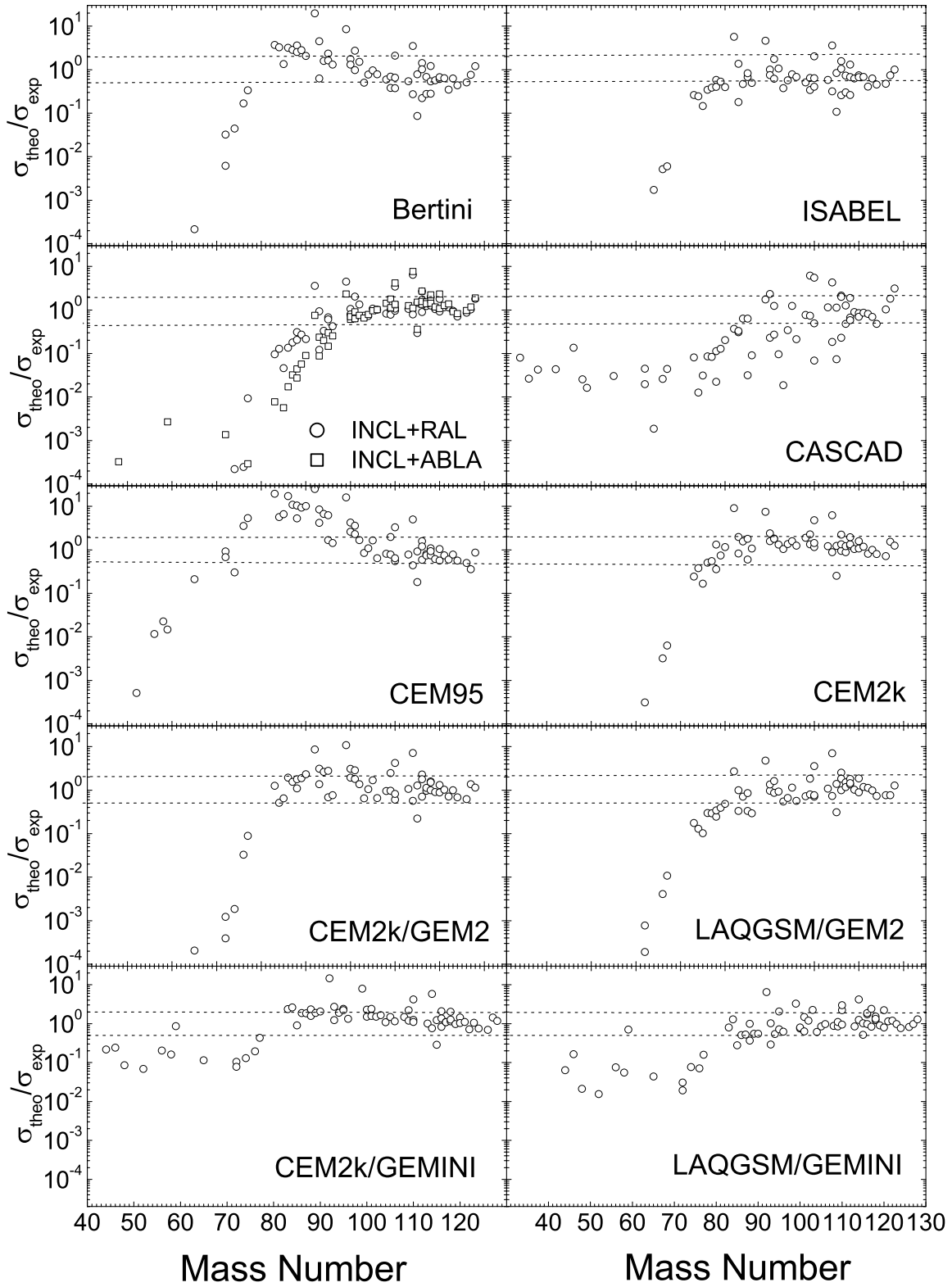


Figure 1: Ratio of theoretical to measured [8] cross sections of isotopes produced by a 660 MeV proton beam on a 15% ^{127}I + 85% ^{129}I target as a function of the product mass number.

CEM2k+GEM2 and LAQGSM+GEM2 consider fission using the GEM2 model [24] of only heavy nuclei, with $Z > 65$, *i.e.*, also not considering fission of this target. Similarly, INCL+ABLA [15, 16] and CASCADE [17] also do not consider fission of ^{129}I . Only the code GEMINI by Charity [26] merged with CEM2k and LAQGSM considers fission (via sequential binary decays) of practically all nuclei, and provides fission products from this reaction. This is why CEM2k+GEMINI and LAQGSM+GEMINI agree better than all the other models tested here with experimental data for this reaction, especially in the $A = 40\text{-}80$ mass region.

Table 1: Comparison of experimental and calculated results for all 42 selected product isotopes from ^{129}I (left panel) and for only 22 spallation products with $A \geq 95$ (right panel)

Model	All 42 selected isotope			22 spallation products with $A \geq 95$		
	$N/N_{30\%}/N_{2.0}$	$\langle F \rangle$	$S(\langle F \rangle)$	$N/N_{30\%}/N_{2.0}$	$\langle F \rangle$	$S(\langle F \rangle)$
Bertini+Dresner	36/ 6/22	3.72	3.00	22/ 6/19	1.67	1.34
ISABEL+Dresner	34/ 5/18	5.18	4.45	22/ 5/16	1.72	1.37
INCL+Dresner	33/14/21	3.86	3.16	22/14/21	1.42	1.28
INCL+ABLA	32/ 9/21	9.32	7.01	22/ 9/21	1.57	1.34
CASCADE	42/ 9/15	11.05	5.19	22/ 9/14	3.32	2.75
CEM95	40/10/20	5.40	3.52	22/ 9/18	1.78	1.44
CEM2k	33/13/26	2.89	2.74	22/11/20	1.48	1.27
LAQGSM+GEM2	33/13/22	3.16	2.68	22/13/21	1.50	1.34
CEM2k+GEM2	35/10/28	5.03	5.04	22/ 8/20	1.60	1.35
LAQGSM+GEMINI	42/12/29	4.28	3.58	22/17/21	1.31	1.21
CEM2k+GEMINI	42/12/27	2.74	2.15	22/ 9/20	1.46	1.25

Newer calculations [8] have shown that it is possible to extend the fission model of GEM2 so that it describes also fission of light nuclei, like ^{129}I , and gives with CEM2k+GEM2 and LAQGSM+GEM2 for this reaction (as well as for other reactions, on other targets) results very similar (even a little better) to the ones provided by GEMINI (compare the thin solid and dashed lines with the corresponding thick lines in Fig. 2; Note that the lines here show the calculated total yield of all products for every given mass number A , while the experimental points show the yield of only measured (not all) isotopes, therefore, the line should be, in general, higher than the experimental points). For this, it is necessary to fit the ratio of the level-density parameters for the fission and evaporation channels, a_f/a_n . We think that it is possible to extend in a similar way also the Atchison fission model [14] and the ABLA evaporation/fission model [16] to describe fission of Iodine also with the Bertini+Dresner/Atchison, ISABEL+Dresner/Atchison, INCL+Dresner/Atchison, and INCL+ABLA options of LAHET3; the same is true for the Dubna code CASCADE. Nevertheless, we are not too optimistic about the predictive power of such extended versions of these codes as they do not contain yet reliable models for fission barriers of light nuclei.

To make the situation even more intricate, we note that when we merge [28, 29] CEM2k+GEM2 and LAQGSM+GEM2 with the Statistical Multifragmentation Model (SMM) by Botvina *et al.* [30], it is possible to describe this reaction and get results very similar to the ones predicted by CEM2k+GEMINI and LAQGSM+GEMINI without extending the fission model of GEM2, *i.e.*, considering only INC, preequilibrium, evaporation, and multifragmentation processes, but not fission of ^{129}I (see the thick solid and dashed lines in Fig. 3). We will discuss these and

other similar results in more details in a future publication. Here, we note that it is impossible to make a correct choice between fission and fragmentation reaction mechanisms involved in our $p+^{129}\text{I}$ reaction by comparing theoretical results with our (or other similar) measurements of only product cross sections; addressing this question would require analysis of two- or multi-particle correlation measurements.

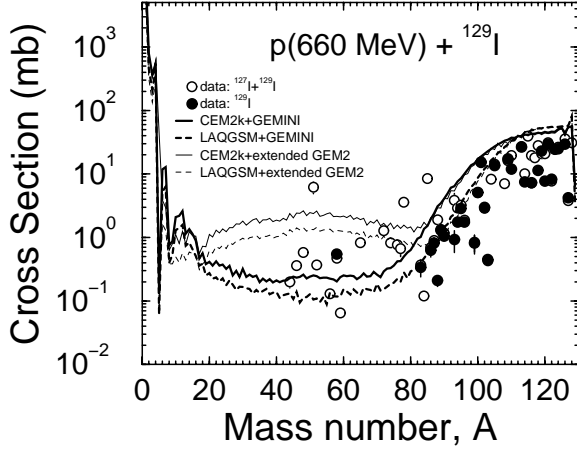


Figure 2: Comparison of experimental and theoretical cross sections for products from 660 MeV $p + \text{I}$. Open circles show data for our 15% $^{127}\text{I} + 85\%$ ^{129}I target, while filled circles show estimated experimental data for only ^{129}I [8]. Thick solid and dashed lines show results for ^{129}I by CEM2k and LAQGSM merged with GEMINI [26], while thin lines, corresponding results by an extended version of GEM2 to consider fission of light nuclei, as described in the text.

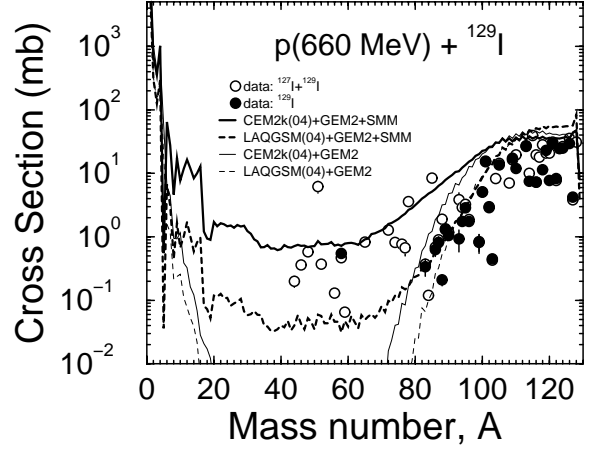


Figure 3: The same experimental data as in Fig. 2 but compared with results for ^{129}I by CEM2k+GEM2 and LAQGSM+GEM2 (thin solid and dashed lines) and by these models extended [28, 29] to consider also multifragmentation [30] of compound nuclei when their excitation energy E^* is above 2 MeV/A, as a competitive channel to evaporation of particles after the preequilibrium stage of reactions (thick solid and dashed lines); no fission is considered here.

Results for the ^{237}Np and ^{241}Am targets are shown in Figs. 4 and 5 and Tabs. 2 and 3, respectively. The situation for these actinides is quite different from what we have above for ^{129}I . First, almost all isotopes measured from these targets [7] are fission products. From all 53 measured isotopes from ^{237}Np , only 37 are selected for the quantitative comparison shown in Table 2, with 32 of them being fission products; and from all 80 measured isotopes from ^{241}Am , only 45 are selected for the comparison in Table 3, and 44 of them are fission products. All codes tested here describe fission of actinides (we do not compare results by CEM95 and CEM2k with the Am and Np data, as these codes do not calculate fission fragment production), but the agreement of the calculations with these measured data [7] is much worse than we have for ^{129}I , for all codes. From Tabs. 2 and 3, we see that none of the codes have a mean deviation factor $\langle F \rangle$ less than a factor of two, whether we compare both fission and spallation products (left panels in Tabs. 2 and 3), or only the fission products (right panels). A little better agreement with these data in comparison with other codes are provided by the old models Bertini+Dresner/RAL and ISABEL+Dresner/RAL from the LAHET transport code. GEMINI, that works so well for ^{129}I , does not describe the Am and Np fission products well. This is not a surprise, as GEMINI was developed to describe well sub-actinides; it needs to be further

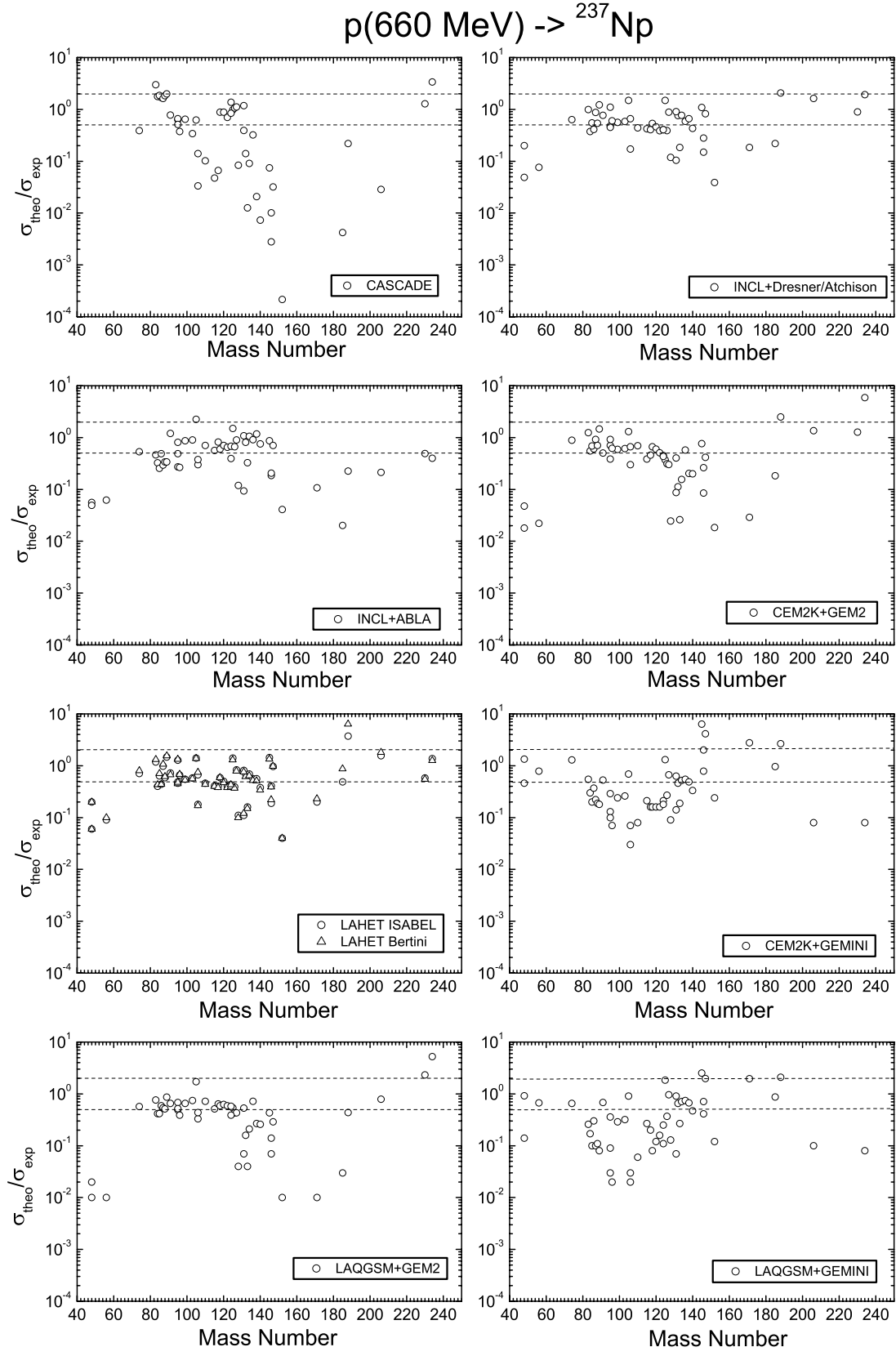


Figure 4: Ratio of theoretical to measured [7] cross sections of isotopes produced by a 660 MeV proton beam on a ${}^{237}\text{Np}$ target as a function of the product mass number.

p(660 MeV) \rightarrow ^{241}Am

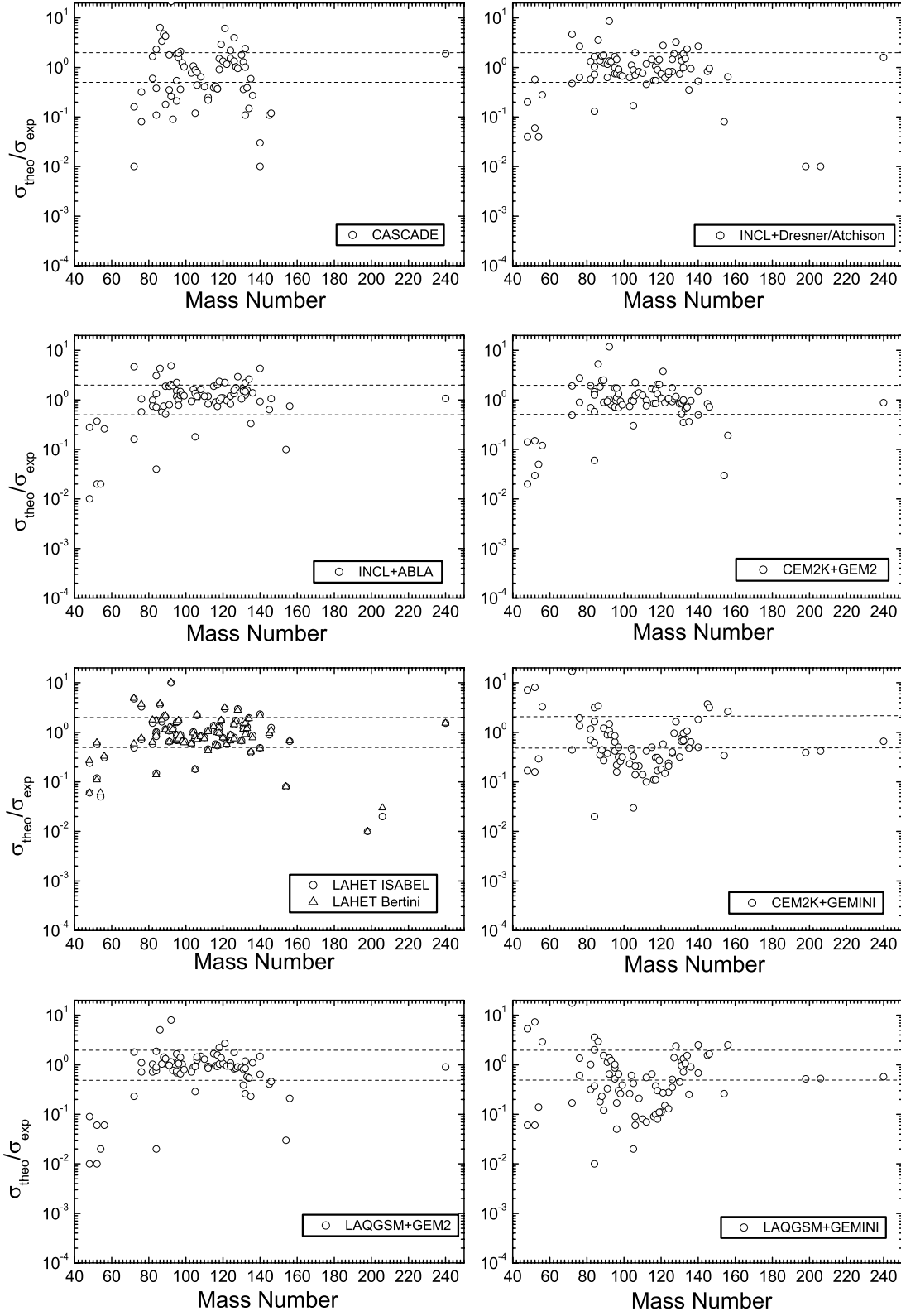


Figure 5: Ratio of theoretical to measured [7] cross sections of isotopes produced by a 660 MeV proton beam on a ^{241}Am target as a function of the product mass number.

improved to become a reliable tool also for actinides. The newly developed code CEM2k and LAQGSM merged with GEM2 and INCL+ABLA, that work so well for many other reactions, also fail to reproduce these Np and Am data, indicating that these codes still have deficiencies and need further improvement.

The qualitative comparisons shown in Figs. 4 and 5 are even more drastic: We see deviations between some calculated and measured cross sections as large as two orders of magnitude, and even higher.

Table 2: Comparison for all 37 selected isotopes (spallation and fission) from ^{237}Np (left panel) and for only 32 selected isotopes (fission products) with $A < 175$ (right panel)

Model	All 37 selected isotope			32 fission products with $A < 175$		
	$N/N_{30\%}/N_{2.0}$	$\langle F \rangle$	$S(\langle F \rangle)$	$N/N_{30\%}/N_{2.0}$	$\langle F \rangle$	$S(\langle F \rangle)$
CASCADE	32/ 5/16	11.29	7.75	27/ 4/15	9.75	7.40
INCL+Dresner/RAL	37/ 8/19	3.35	2.39	32/ 7/16	3.51	2.49
INCL+ABLA	37/ 9/14	4.52	2.92	32/ 9/14	4.09	2.78
Bertini+Dresner/RAL	37/ 6/20	3.18	2.26	32/ 4/16	3.29	2.30
ISABEL+Dresner/RAL	37/ 5/19	3.18	2.26	32/ 5/16	3.34	2.37
CEM2k+GEM2	37/ 6/18	5.43	3.47	32/ 5/16	5.80	3.70
CEM2k+GEMINI	36/ 4/11	3.87	2.05	32/ 3/10	3.57	1.88
LAQGSM+GEM2	37/ 2/15	7.10	4.29	32/ 1/14	7.28	4.47
LAQGSM+GEMINI	36/ 5/13	4.82	2.69	32/ 4/12	4.69	2.64

Table 3: Comparison for all 45 selected isotopes (spallation and fission) from ^{241}Am (left panel) and for only 44 selected isotopes (fission products) with $A < 175$ (right panel)

Model	All 45 selected isotope			44 fission products with $A < 175$		
	$N/N_{30\%}/N_{2.0}$	$\langle F \rangle$	$S(\langle F \rangle)$	$N/N_{30\%}/N_{2.0}$	$\langle F \rangle$	$S(\langle F \rangle)$
CASCADE	41/ 7/16	6.43	4.10	40/ 7/15	6.57	4.15
INCL+Dresner/RAL	45/14/37	2.43	2.13	44/14/36	2.44	2.15
INCL+ABLA	45/17/36	2.93	2.68	44/16/35	2.96	2.69
Bertini+Dresner/RAL	45/19/35	2.26	1.98	44/19/34	2.28	1.99
ISABEL+Dresner/RAL	45/19/35	2.28	1.99	44/19/34	2.30	2.01
CEM2k+GEM2	45/22/33	2.73	2.48	44/21/32	2.76	2.50
CEM2k+GEMINI	45/ 7/18	3.14	2.01	44/ 7/17	3.18	2.02
LAQGSM+GEM2	45/22/34	3.34	3.12	44/21/33	3.38	3.14
LAQGSM+GEMINI	45/ 8/23	3.64	2.51	44/ 8/22	3.69	2.53

Let us mention that such a big disagreement between calculations by the codes tested here and these experimental data does not mean that the codes are in general too bad and they can not predict well any data. The measurements [7, 8] were done by the gamma-spectrometry method which allows us to measure only a small part of all products from any reaction. In addition, most of the measured cross sections are cumulative, while all codes provide only independent cross sections that need to be summed up with the corresponding branching ratios to be able to compare with the measured cumulative yields. That makes such comparisons more difficult; many products are measured either only in the ground states or only as long-lived

isomers, and such data can not be compared with our calculations (see details about this in [6, 27]). It is much easier and convenient to test codes against recent measurements done at GSI in Darmstadt, Germany using inverse kinematics for interactions of ^{56}Fe , ^{197}Au , ^{208}Pb , and ^{238}U at 1 GeV/nucleon and several lower energies with liquid ^1H , as well as with many heavier targets up to ^{208}Pb . References to most of the GSI measurements and many tabulated experimental cross sections may be found on the Web page of Prof. Schmidt [31]. The GSI measurements provide a very rich set of cross sections for production of practically all possible isotopes from such reactions in a “pure” form, *i.e.*, individual cross sections from a specific given bombarding isotope (or target isotope, when considering reactions in the usual kinematics, $p + A$). Such cross sections are much easier to compare to models than the “camouflaged” data from γ -spectrometry measurements, like the ones analyzed here, and many of the codes tested here describe very well most of the GSI measurements, with a mean deviation factor usually not higher than a factor of two (see, *e.g.*, [6, 15, 32, 33] and references therein). With these circumstances in mind, our present comparison of the I, Np, and Am measurements [7, 8] with calculations by different models does not pretend either to be comprehensive or to pick up “the best” tested code. Rather, our goal is to investigate problems some models may still have in reproducing well some specific measurements, with a hope that this would help the authors of codes to improve their models. The results of the present work show that all codes tested here still have some big problems in a correct description of many of the ^{129}I , ^{237}Np , and ^{241}Am data and all models should be further improved.

Conclusions

We have analyzed the recent JINR (Dubna, Russia) measurements on nuclide production cross sections from interaction of 660 MeV proton beams with radioactive targets of enriched ^{129}I (85% ^{129}I and 15% ^{127}I), ^{237}Np , and ^{241}Am [7, 8] with eleven different models, realized in eight transport codes and event-generators: LAHET (Bertini, ISABEL, INCL+ABLA, and INCL+RAL options), CASCADE, CEM95, CEM2k, LAQGSM+GEM2, CEM2k+GEM2, LAQGSM+GEMINI, and CEM2k+GEMINI. We found out that all these models have problems in a correct description of many of these cross sections, though some of these models describe very well most of the recent measurements done at GSI using inverse kinematics, as well as many other reactions. None of the tested here models is able to reproduce well all the JINR data and all of them should be further improved. Development of a better universal evaporation/fission model should be of a highest priority.

In the case of the ^{129}I target, products with mass numbers $A = 40 - 70$ were measured [8]. Such isotopes can not be described by the models tested here as spallation products, and may be considered as produced either via fission or multifragmentation. The CEM2k and LAQGSM codes merged with the sequential binary decay model GEMINI can reproduce such isotopes as fission products. On the other hand, CEM2k and LAQGSM merged with the Statistical Multifragmentation Model (SMM) can also reproduce these isotopes via multifragmentation, *i.e.*, considering only intranuclear cascade, preequilibrium, multifragmentation, and evaporation processes, without fission of ^{129}I . Similar results were obtained with these codes for $p + ^{56}\text{Fe}$ reactions at 1.5, 1.0, 0.75, 0.5, and 0.3 GeV measured at GSI in inverse kinematics. We conclude that it is impossible to make a correct choice between fission and fragmentation reaction mechanisms analyzing only measurements on product cross sections, at least for interactions of intermediate-energy nucleons with medium-mass targets; addressing this question would require analysis of two- or multi-particle correlation measurements.

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