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# Study of 660-MeV Proton-Induced Reactions on $^{129}\text{I}$

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**Abstract.** Isotopically enriched  $^{129}\text{I}$  (85%  $^{129}\text{I}$  and 15%  $^{127}\text{I}$ ) targets were irradiated with a beam of 660-MeV protons at the JINR DLNP Phasotron and cross sections of formation of 74 residual products were determined using the  $\gamma$ -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. 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. 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  $5 \times 10^6$  years necessary to store the same spent fuel without transmutation to decrease its risk to a similar level.

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 [3]. Experimental cross-sections for residual nuclei in radioactive  $^{129}\text{I}$  target [4] are undeniably important for the projects of transmutation of nuclear wastes in a direct proton beam. 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. Details on our measurement and on models used here to analyze the data may be found in Ref. [4].

Preliminary results of our measured product cross sections from  $^{129}\text{I}$  targets irradiated with 660 MeV protons were published earlier [5]. Here, we analyze final experimental cross sections for our 85%  $^{129}\text{I}$  and 15%  $^{127}\text{I}$  target irradiated by 660 MeV protons [7]. Using previous measurements on  $^{127}\text{I}$  targets generally performed at energies below 660 MeV analyzed by Molodo and Holzbach [6] and reducing them to our proton energy of 660 MeV by linear interpolation between energies 600 and 800 MeV, we estimate [7] experimental cross sections for the target  $^{129}\text{I}$  at 660 MeV.

## RESULTS AND DISCUSSION

Our  $^{129}\text{I}$  data are tabulated in [7] and we do not show them here. We analyzed all the measured data with the LAHET3 version [8] of the transport code LAHET [9] using the Bertini [10] and ISABEL [11] IntraNuclear-Cascade (INC) models merged with the Dresner evaporation model [12] and the Atchison fission model (RAL) [13], and using the Liege INC code by Cugnon *et al.* INCL [14] merged in LAHET3 with the ABLA [15] and with Dresner [12] (+ Atchison [13]) evaporation (+ fission) models, with the Dubna transport code CASCADE [16], with versions of the Cascade-Exciton Model (CEM) [17] as realized in the codes CEM95 [18] and CEM2k [19], with CEM2k merged [20] with the Generalized Evaporation/fission Model code GEM2 by Furihata [21], with the Los Alamos version of the Quark-Gluon String Model code LAQGSM [22] merged [20] with GEM2 [21], as well as with versions of the CEM2k and LAQGSM codes merged both [20] with the sequential-binary-decay code GEMINI by Charity [23]. 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 [8]-[23] and references therein.

As we have done previously (see, *e.g.*, [3, 24]), 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 [7], only 42 were selected to satisfy some

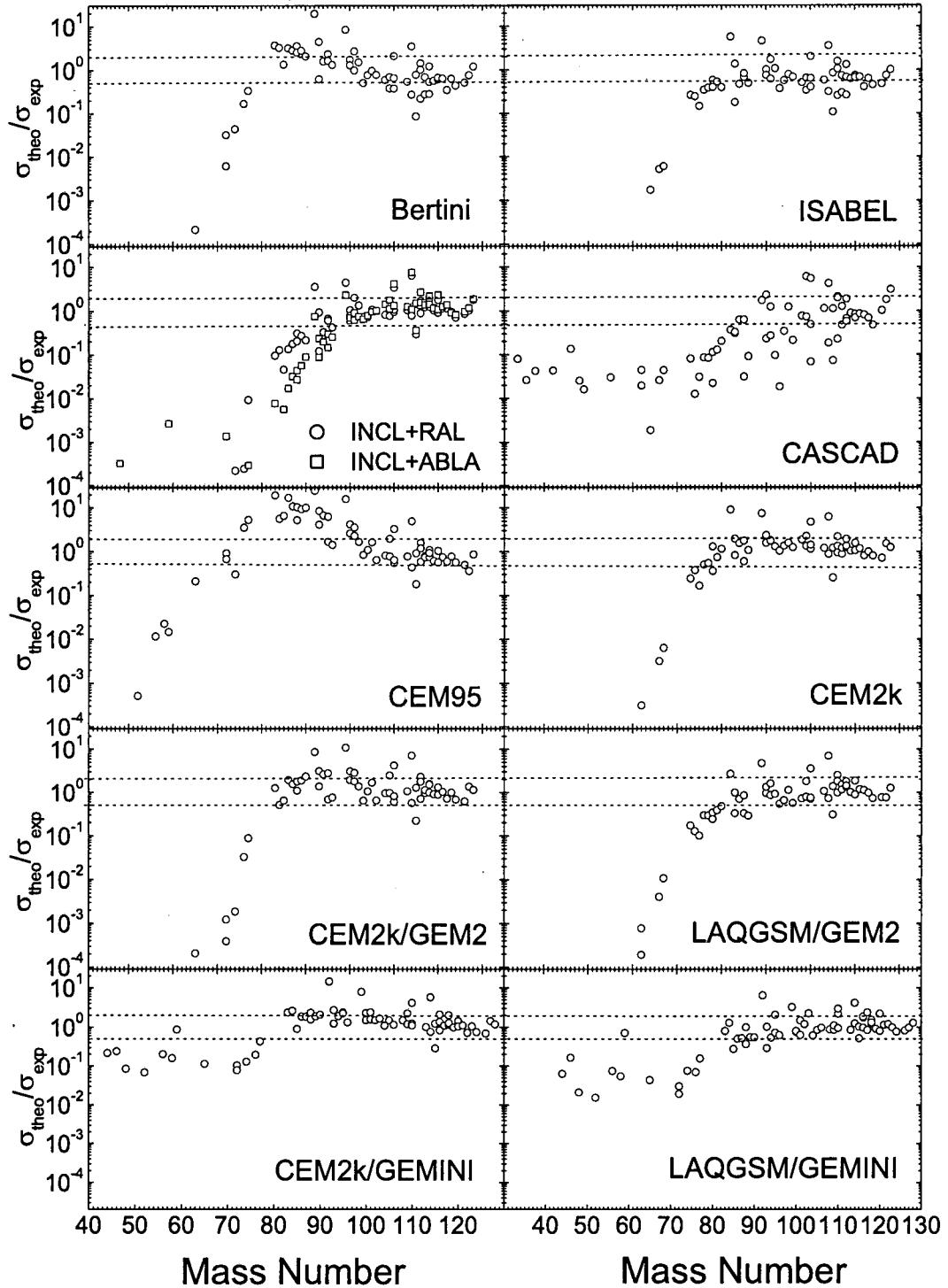
rules based on appreciation of the physical principles realized in the models. For instance, if only an isomer or only the ground state of a nuclide with a relatively long-lived isomer was measured, such nuclides were excluded from the quantitative comparison, but if both were measured separately, their sum was compared with calculations. Such rules are essentially similar to those used by Titarenko *et al.* [3, 24].

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 [18] and CEM2k [19] 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 [10] and ISABEL [11] INC's are used in our calculations with the default options of LAHET3 for evaporation/fission models; they consider evaporation with the Dresner code [12] and a possible fission of heavy compound nuclei using the Atchison RAL fission model [13], but only if they are heavy enough ( $Z > 71$ ), *i.e.*, they do not consider fission for such light targets as  $^{129}\text{I}$ .

CEM2k+GEM2 and LAQGSM+GEM2 consider fission using the GEM2 model [21] of only heavy nuclei, with  $Z > 65$ , *i.e.*, also not considering fission of this target. Similarly, INCL+ABLA [14, 15] and CASCADE [16] also do not consider fission of  $^{129}\text{I}$ . Only the code GEMINI by Charity [23] 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  $A = 40-80$  mass region.

Newer calculations [7] have shown that it is possible to extend the fission model of GEM2 so that it describes also fission of light nuclei, like  $^{129}\text{I}$ , 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. For this, it is necessary to fit the ratio of the level-density parameters for the fission and evaporation



**FIGURE 1.** Ratio of theoretical to measured [7] cross sections of isotopes produced by a 660 MeV proton beam on our 15%  $^{127}\text{I}$  + 85%  $^{129}\text{I}$  target as a function of the product mass number.

**TABLE 1.** Comparison of experimental and calculated results for all 42 selected product isotopes from  $^{129}\text{I}$  (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

channels,  $a_f/a_n$ . We think that it is possible to extend in a similar way also the Atchison fission model [13] and the ABLA evaporation/fission model [15] 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 [25, 26] CEM2k+GEM2 and LAQGSM+GEM2 with the Statistical Multifragmentation Model (SMM) by Botvina *et al.* [27], 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  $^{129}\text{I}$ . 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.

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## REFERENCES

1. H. Küsters, *Atomwirtschaft*, **35**, 287-292 (1990).
2. W. Gudowski, *Nucl. Phys.*, **A654**, 436-457 (1999).
3. Yu. E. Titarenko, *et al.*, *Phys. Rev.*, **C65**, 064610 (2002).
4. V. S. Pronskikh, *et al.*, *submitted to Phys. Rev. C* (2004).
5. J. Adam, *et al.*, *J. Nucl. Sci. and Technol., Suppl.*, **2**, 272-275 (2002).
6. G. Modolo and M. Holzbach, Preprint Jül-3257 (1996).
7. V. S. Pronskikh, *et al.*, in Proc. *Workshop on Nuclear Data for the Transmutation of Nuclear Waste*, ISBN 3-00-012276-1, GSI-Darmstadt, Germany, 2003.
8. R. E. Prael, LANL Report LA-UR-01-1655 (2001).
9. R. E. Prael and H. Lichtenstein, LANL Report LA-UR-89-3014 (1989).
10. H. W. Bertini, *Phys. Rev.*, **131**, 1801-1871 (1963).
11. Y. Yariv and Z. Fraenkel, *Phys. Rev.*, **C24**, 488-494 (1981).
12. L. Dresner, Oak Ridge National Laboratory Report ORNL-TM-196, (unpublished) (1962).
13. F. Atchison, in Proc. *Meeting on Targets for Neutron Beam Spallation Source*, Jülich, 1979, pp. 17-46.
14. A. Boudard, *et al.*, *Phys. Rev.*, **C66**, 044615 (2002).
15. J.-J. Gaimard and K.-H. Schmidt, *Nucl. Phys.*, **A531**, 709-745 (1991).
16. V. S. Barashenkov, *Comp. Phys. Comm.*, **126**, 28-31 (2000).
17. K. K. Gudima, S. G. Mashnik, and V. D. Toneev, *Nucl. Phys.*, **A401**, 329-361 (1983).
18. S. G. Mashnik, *User Manual for the Code CEM95*, JINR, Dubna, USSR; RSIC-PSR-357, Oak Ridge, USA (1995).
19. S. G. Mashnik and A. J. Sierk, in Proc. *AccApp* (Washington, DC, 2000), AIP, 2001, pp. 328-341.
20. S. G. Mashnik, K. K. Gudima, and A. J. Sierk, in Proc. *SATIF-6* (SLAC, USA, 2002), NEA/OECD, Paris, France, 2004, pp. 337-366.
21. S. Furihata, *Nucl. Instr. Meth.*, **B171**, 252-258 (2000).
22. K. K. Gudima, S. G. Mashnik, A. J. and Sierk, LANL Report LA-UR-01-6804, Los Alamos (2001).
23. R. J. Charity, *et al.*, *Nucl. Phys.*, **A483**, 371-405 (1988).
24. Yu. E. Titarenko, *et al.*, in Proc. *TRAMU Workshop*, ISBN 3-00-012276-1, GSI-Darmstadt, Germany, 2003.
25. S. G. Mashnik, K. K. Gudima, A. J. Sierk, and R. E. Prael, LANL Research Note X-5-RN (U) 04-08 (2004).
26. S. G. Mashnik, K. K. Gudima, R. E. Prael, A. J. Sierk, in Proc. *ICRS-10/RPS2004*, Madeira, Portugal, 2004, in press.
27. A. S. Botvina, A. S. Iljinov, and I. N. Mishustin, *Nucl. Phys.*, **A507**, 649-674 (1990).