Study of Proton Induced Reactions in a Radioactive $^{129}$I Target at $E_p=660$ MeV

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Abstract: Two NaI (85$^{\%}$ $^{129}$I and 15$^{\%}$ $^{127}$I) targets were exposed to a beam of 660-MeV protons. Cross sections for formation of 76 residual nuclei were obtained by the induced activity method. The results are compared with other experimental data on $^{127}$I and theorectical calculations by eleven models contained in the codes LAHET3 (using the Bertini+Dresner, ISABEL+Dresner, INCL+Dresner, and INCL+ALBA options), CASCADE, CEM95, CEM2k, LAQGSM+GEM2, CEM2k+GEM2, LAQGSM+GEMINI, and CEM2k+GEMINI. Most of the models describe spallation products with masses close to the target reasonably well while the reliability of the codes differs greatly in the deep spallation and fission/fragmentation regions. The difficulties in describing products with $A=40-80$ by all of the codes tested here except for CEM2k+GEMINI and LAQGSM+GEMINI is related to the neglect of fission (and fragmentation) processes for targets as light as $^{129}$I.

Introduction

This work is a continuation of [1], where results of determining cross sections for the yields of residual nuclei from the $^{237}$Np and $^{241}$Am targets exposed to an extracted 660-MeV proton beam of the JINR DNLP Phasotron were reported. The first experiments to study transmutation of $^{129}$I ($T_{1/2} = 1.57 \times 10^7$ y) and some transuranic isotopes were carried out in 1996 with heavy-ion beams of the JINR LHE Synchrophasotron using relativistic protons of 3.7 GeV [2]. Preliminary results of our measured product cross sections from $^{129}$I targets irradiated with 660 MeV protons were published earlier [3]. Here, we present final experimental cross sections for our 85$^{\%}$ $^{129}$I and 15$^{\%}$ $^{127}$I target irradiated by 660 MeV protons. Using previous measurements on $^{127}$I targets generally performed at energies below 660 MeV analyzed by Molodo and Holzbach [4] and reducing them to our proton energy of 660 MeV by linear interpolation between energies 600 and 800 MeV, we estimate here experimental cross sections for the target $^{129}$I at 660 MeV. We analyze our measurements with eleven models incorporated into several transport codes used recently in different applications. Details on our measurement and on models used here to analyze the data may be found in Ref. [5]

Results and Discussion

Table 1 presents our data: Column 1 lists the isotopes (ground or metastable states) for which we obtained reliable production cross sections, columns 2 and 3 show their half-lives ($T_{1/2}$), decay and cross section types, respectively, column 4 presents the measured cross sections for our 85$^{\%}$ $^{129}$I and 15$^{\%}$ $^{127}$I target, column 5 shows cross sections for $^{127}$I from [4], and column 6 shows our deduced experimental cross sections for $^{129}$I.

Table 1: Experimental results

| Residual | $T_{1/2}$ | Decay | $\sigma_{\text{exp}, \text{mb}}$ | $\sigma_{\text{exp}, \text{mb}}$ ($^{127}$I) | $\sigma_{\text{exp}, \text{mb}}$ ($^{129}$I) |
|----------|----------|-------|-------------------------------|-------------------------------|-------------------------------|
| $^{44}$mSc | 2.44 d | I(\text{IT,EC}) | 0.20(4) | —— | —— |
| $^{46}$Sc | 83.83 d | C(\text{\beta-}) | 0.36(4) | —— | —— |
| $^{48}$V | 15.97 d | C(\text{\beta+,EC}) | 0.58(6) | —— | —— |

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| Element | Spin | I(β+),EC | I(β+,EC) | I(β−),EC | I(β−,IT) |
|---------|------|-----------|-----------|-----------|-----------|
| $^{52}$Mn | 5.29 d | C(EC,β+) | 0.37(5) | —— | —— |
| $^{56}$Co | 78.8 d | C(EC,β+) | 0.11(4) | —— | —— |
| $^{58}$Co | 70.92 d | I(EC,β+) | 0.47(15) | 0.015(5) | 0.55(20) |
| $^{59}$Fe | 44.5 d | C(β−) | 0.065(7) | —— | —— |
| $^{65}$Zn | 244.1 d | C(EC,β+) | 0.88(9) | —— | —— |
| $^{72}$As | 26 h | I(β+,EC) | 0.93(9) | —— | —— |
| $^{72}$Se | 8.4 d | C(EC) | 0.59(15) | —— | —— |
| $^{74}$As | 17.78 d | I(EC,β+,β−) | 0.85(9) | —— | —— |
| $^{76}$Br | 16.2 h | C(β+,EC) | 0.77(9) | —— | —— |
| $^{77}$Br | 2.38 d | C(EC,β+) | 0.64(15) | —— | —— |
| $^{83}$Rb | 86.2 d | C(EC) | 0.40(8) | 0.53(5) | 0.38(10) |
| $^{84}$Rb | 32.87 d | I(EC,β+,β−) | 0.12(4) | —— | —— |
| $^{85}$Sr | 64.84 d | C(EC) | 1.49(16) | —— | —— |
| $^{86}$Y | 14.74 h | C(β+,EC) | 0.69(25) | 1.04(10) | 0.63(30) |
| $^{87}$Y | 3.35 d | C(EC,β+) | 1.15(11) | 1.35(15) | 1.11(15) |
| $^{88}$Y | 106.6 d | I(EC,β+) | 0.36(10) | 2.21(25) | —— |
| $^{88}$Zr | 83.4 d | C(EC) | 1.4(4) | —— | —— |
| $^{89}$Zr | 3.27 d | C(EC,β+) | 1.51(15) | 1.54(30) | 1.50(17) |
| $^{90}$Nb | 14.60 h | C(β+,EC) | 1.18(13) | 1.93(20) | 1.05(13) |
| $^{92}$Nb | 10.15 d | I(EC,β+) | 0.11(4) | —— | —— |
| $^{93}$Mo | 6.85 h | I(IT,EC) | 0.94(30) | 1.00(20) | 0.93(35) |
| $^{93}$Tc | 2.75 h | C(EC,β+) | 2.14(24) | —— | —— |
| $^{94}$Tc | 4.88 h | I(EC,β+) | 1.79(17) | 2.01(35) | 1.74(12) |
| $^{94m}$Tc | 52 m | I(β+,EC) | 0.44(8) | —— | —— |
| $^{95}$Nb | 34.98 d | C(β−) | 0.36(5) | 0.14(2) | 0.40(6) |
| $^{96}$Tc | 20.0 h | C(EC) | 3.00(34) | 5.2(7) | 2.6(4) |
| $^{96}$Rh | 4.28 d | I(EC) | 2.50(25) | 2.57(50) | 2.49(30) |
| $^{99}$Rh | 16.0 d | I(EC,β+) | 0.84(25) | 0.95(10) | 0.82(27) |
| $^{100}$Pd | 20.8 h | I(EC,β+) | 4.21(50) | 4.81(60) | 4.10(50) |
| $^{100}$Rh | 3.63 d | C(EC) | 2.85(33) | 4.1(4) | 2.6(4) |
| $^{101}$Rh | 4.34 d | C(EC,β+) | 9.21(34) | 10.2(10) | 9.03(40) |
| $^{102}$Rh | 2.9h | I(EC,β+) | 2.9(9) | —— | —— |
| $^{103}$Ru | 39.25 d | C(β−) | 0.43(5) | 0.30(3) | 0.45(6) |
| $^{104}$Ag | 69.2 m | I(EC,β+) | 8.3(8) | —— | —— |
| $^{105}$Ag | 41.29 d | C(EC) | 14.3(17) | 18.3(20) | 13.6(20) |
| $^{106}$Ag | 8.46 d | I(EC) | 7.5(7) | 6.9(8) | 7.6(8) |
| $^{108}$In | 58 m | I(β+,EC) | 7.1(7) | —— | —— |
| $^{109}$In | 4.20 h | C(EC,β+) | 15.1(20) | 17.7(24) | 15.9(30) |
| $^{109}$Sn | 18.0 m | C(EC,β+) | 3.02(30) | —— | —— |
| $^{110}$Ag | 249.9 d | I(β−,IT) | 1.50(18) | 0.89(8) | 1.61(15) |
| $^{110}$In | 4.9 h | I(EC) | 11.7(12) | 19.6(20) | 10.3(12) |
| $^{110m}$In | 69.1 m | C(EC) | 6.4(8) | —— | —— |
| $^{113}$Sn | 115.1 d | C(EC) | 27.2(30) | 30.7(30) | 26.6(35) |
| $^{114m}$In | 49.51 d | I(IT,EC) | 7.1(7) | 4.8(5) | 7.5(7) |
| $^{114}$Sb | 3.49 m | C(EC,β+) | 2.92(35) | —— | —— |
| $^{115m}$In | 4.49 d | I(IT,β−) | 15.6(35) | —— | —— |
| $^{115}$Sb | 32.1 m | C(EC,β+) | 20.0(30) | —— | —— |
| $^{116}$In | 54.15 m | I(β−) | 2.72(42) | —— | —— |
| $^{116}$Sb | 15.8 m | I(β+,EC) | 2.0(4) | —— | —— |
| $^{116m}$Sb | 60.3 m | I(EC,β+) | 11.6(14) | —— | —— |
| $^{116}$Te | 2.49 h | C(EC,β+) | 9.9(10) | —— | —— |
| $^{117}$In | 43.8 m | I(β−) | 1.6(3) | —— | —— |
We analyzed our measurements with the LAHET3 version [6] of the transport code LAHET [7] using the Bertini [8] and ISABEL [9] intranuclear-cascade models (INC) merged with the Dresner evaporation model [10] and the Atchison fission model (RAL) [11], and using the code INCL by Cugnon et al. [12] merged in LAHET3 with the ABLA [13] and with Dresner [10] (+ Atchison [11]) evaporation (+ fission) models, with the Dubna transport code CASCADE [14], with versions of the Cascade-Exciton Model (CEM) [15] as realized in the codes CEM95 [16] and CEM2k [17], with CEM2k merged [18-20] with the Generalized Evaporation/fission Model code GEM2 by Furiihata [21], as well as with the Los Alamos version of the Quark-Gluon String Model code LAQGSM [22] merged [18-20] with GEM2 [21], and with versions of the CEM2k and LAQGSM codes both merged [18] with the sequential-binary-decay code GEMINI by Charity [23]. Most of these models are widely used to study reaction on heavier nuclei (see, e.g., [21] [23] and references therein).

As we have done previously (see, e.g., [21] [25]), we choose here two quantitative criteria 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 (Tables 2 and 3)

$$\langle F \rangle = 10^{\sqrt{\langle \log(\sigma_{cal}/\sigma_{exp}) \rangle}} ,$$

with its standard deviation:

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

For such a comparison, out of all the 76 cross sections measured in this work, only 48 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 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. [24] [25].

To understand how different models describe nuclides produced in the spallation and fission or fragmentation regions, we divided all 48 measured nuclides included in the comparison into two groups, spallation ($A \geq 95$) and fission/fragmentation ($A < 95$). Table 2 shows values of $\langle F \rangle$ and $S(\langle F \rangle)$ for all compared products (both spallation and fission/fragmentation), while Tab. 3 shows such results only for spallation; $N$ is the total number of comparisons, $N_{30\%}$—the number of comparisons in which calculated and measured values differ by not more than 30 %, $N_{2,0}$—where the difference was not more than a factor of two.

We note that the codes CEM95 [16] and CEM2k [17] 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 [8] and ISABEL [9] INC's are used in our calculations with the default options of LAHET3 for evaporation/fission models; they consider evaporation with the Dresner code [10] and a possible fission of heavy compound nuclei using the Atchison RAL fission model [11], but only if they are heavy enough ($Z > 71$), i.e., do not consider fission for such light targets.

### Table 1: Experimental results (continued)

| No. | Isotope | T_{1/2} | Type | Cross Section | Error | Calculated sigma |
|-----|---------|---------|------|---------------|-------|-----------------|
| 117Te | 1.03 h | C(EC,β+) | 15.4(15) | —— | —— |
| 118mSb | 5.00 h | I(EC,β+) | 11.1(12) | 8.8(12) | 11.5(17) |
| 118Te | 6.00 d | I(EC) | 13.9(14) | —— | —— |
| 118I | 13.7 m | C(β+,EC) | 3.3(4) | —— | —— |
| 119Te | 16.05 h | C(EC,β+) | 11.5(12) | 15.8(16) | 10.7(15) |
| 119mTe | 4.69 d | I(EC,β+) | 16.1(15) | 15.5(20) | 16.3(20) |
| 120mSb | 5.76 d | I(EC) | 6.3(6) | 5.6(6) | 6.4(7) |
| 120I | 1.35 h | C(β+,EC) | 10.2(12) | —— | —— |
| 120mI | 53.0 m | I(β+,EC) | 2.9(3) | —— | —— |
| 121Te | 16.8 d | C(EC) | 17.9(18) | 24.0(40) | 16.8(20) |
| 121mTe | 154.0 d | I(IT,EC) | 13.3(16) | 16.9(17) | 12.7(13) |
| 122Sb | 2.70 d | C(β−,EC,β+) | 7.7(9) | 4.3(5) | 8.3(10) |
| 123I | 13.2 h | C(EC) | 25.0(24) | 26.1(30) | 24.6(30) |
| 124I | 4.18 d | I(EC,β+) | 26.3(30) | 32.3(40) | 25.3(35) |
| 126Sb | 12.4 d | I(β−) | 0.54(5) | —— | —— |
| 126I | 13.02 d | I(IT) | 35(5) | 66.0(80) | 29.5(20) |
| 127Xe | 36.46 d | C(EC) | 3.8(4) | 1.42(20) | 4.2(5) |
| 128I | 25.0 m | I(β−,EC) | 31(5) | —— | —— |
as $^{129}$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 our target. Similarly, INCL+ABLA [12, 13] also does not consider fission for 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 our reactions. 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 [5] 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 our reactions 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 channels, $a_f/a_n$. We think that it is possible to extend in a similar way also the Atchison fission model [11] and the ABLA evaporation/fission model [13] to describe fission of Iodine also with the Bertini+Dresner/Atchison, ISABEL+Dresner/Atchison, INCL+Dresner/Atchison, and INCL+ABLA

Figure 1: Comparison of experimental and theoretical cross sections for our 15% $^{127}$I + 85% $^{129}$I target.
Table 2: Comparison for all 48 (spallation and fission/fragmentation) selected isotopes

| Model                  | $N/N_{30\%}/N_{2.0}$ | $\langle F \rangle$ | $S(\langle F \rangle)$ |
|------------------------|-----------------------|---------------------|-------------------------|
| Bertini+Dresner        | 43/6/23               | 6.18                | 5.28                    |
| ISABEL+Dresner         | 40/6/18               | 5.70                | 4.59                    |
| INCL+Dresner           | 36/13/21              | 4.10                | 3.12                    |
| INCL+ABLA              | 36/10/19              | 9.95                | 6.41                    |
| CASCADE                | 48/9/16               | 11.36               | 5.20                    |
| CEM95                  | 46/9/22               | 5.15                | 3.27                    |
| CEM2k                  | 40/15/29              | 6.50                | 6.62                    |
| LAQGSM+GEM2            | 42/12/23              | 14.37               | 12.78                   |
| CEM2k+GEM2             | 43/11/31              | 11.08               | 10.80                   |
| LAQGSM+GEMINI          | 48/17/32              | 4.21                | 3.35                    |
| CEM2k+GEMINI           | 48/12/29              | 2.78                | 2.11                    |

Table 3: Comparison for only 26 selected spallation isotopes with $A \geq 95$

| Model                  | $N/N_{30\%}/N_{2.0}$ | $\langle F \rangle$ | $S(\langle F \rangle)$ |
|------------------------|-----------------------|---------------------|-------------------------|
| Bertini+Dresner        | 26/6/20               | 1.86                | 1.47                    |
| ISABEL+Dresner         | 26/6/17               | 1.91                | 1.48                    |
| INCL+Dresner           | 26/13/21              | 2.07                | 1.89                    |
| INCL+ABLA              | 26/10/19              | 2.46                | 2.07                    |
| CASCADE                | 26/9/15               | 3.79                | 3.02                    |
| CEM95                  | 26/8/20               | 1.93                | 1.51                    |
| CEM2k                  | 26/12/22              | 1.72                | 1.51                    |
| LAQGSM+GEM2            | 26/12/21              | 1.84                | 1.61                    |
| CEM2k+GEM2             | 26/9/22               | 1.84                | 1.55                    |
| LAQGSM+GEMINI          | 26/16/23              | 1.55                | 1.40                    |
| CEM2k+GEMINI           | 26/9/20               | 1.75                | 1.49                    |

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 yet contain reliable models for fission barriers of light nuclei.

To make the situation even more intricate, we note that when we merge \cite{26} CEM2k+GEM2 and LAQGSM+GEM2 with the Statistical Multifragmentation Model by Botvina et al. \cite{27}, it is possible to describe these reactions 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, pre-equilibrium, evaporation, and multifragmentation processes, but not fission of $^{129}$I. We will discuss these results 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 $p+^{129}$I reaction by comparing theoretical results with only our (or other similar) data; addressing this question would require analysis of two- or multi-particle correlation measurements.

From Fig. 1 and Tabs. 2 and 3 we see that the agreement of different models with our 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 target, but differ greatly in the deep spallation, fission, and/or fragmentation regions. We conclude that none of the codes tested here is able to reproduce well all of our data and all of them need to be further improved; development of a better universal evaporation/fission model should be of a high priority.

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