STUDY OF RESIDUAL PRODUCT NUCLIDE YIELDS FROM 0.1, 0.2, 0.8, AND 2.6 GEV PROTON-IRRADIATED $^{nat}$Hg TARGETS

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Abstract

The direct $\gamma$-spectrometry method is used to measure more than 350 residual product nuclide yields from 0.1, 0.2, 0.8, and 2.6 GeV proton irradiated $^{nat}$HgO targets. The $\gamma$-spectrometer resolution is of 1.8 keV at the 1332 keV $\gamma$-line. The resultant $\gamma$-spectra are processed by the GENIE2000 code. The $\gamma$-lines are identified, and the cross-sections calculated, by the ITEP-designed SIGMA code using the PCNUDAT radioactive database. The $^{27}$Al(p,x)$^{22}$Na reaction is used as monitor. The experimental results are compared with calculations by the LAHET, CEM95, CEM2k, INUCL, CASCADE, and YIELDX codes.

Introduction

Mercury is planned to be used as a target material in all the present-day designs of the Spallation Neutron Source (SNS) facilities, thus necessitating that the proton-Hg interaction characteristics should be studied in a broad energy range from a few MeV to 2-3 GeV. Among the characteristics, the yields of residual product nuclei are of particular importance. They are independent nuclear constants to be used in practical calculations as well as to verify the codes for calculating the SNS facility design parameters, such as radioactivity (both current and residual), deterioration of resistance to corrosion, yields of gaseous products, poisoning, etc.
Table 1: Parameters of experimental samples and monitors, irradiation conditions, and monitor reaction cross sections

| Proton energy, GeV | Sample thickness, mg/cm² | Monitor thickness, mg/cm² | Irradiation time, min | Mean proton flux density, p/cm²/s | \(^{27}\text{Al}(p,x)^{22}\text{Na} \) cross section, mb |
|-------------------|--------------------------|---------------------------|-----------------------|----------------------------------|---------------------------------|
| 0.10              | 536.0                    | 138.2                     | 60                    | 2.5·10⁹                         | 19.1 ± 1.3                     |
| 0.20              | 537.4                    | 137.3                     | 45                    | 7.1·10⁹                         | 15.1 ± 0.9                     |
| 0.80              | 529.3                    | 139.1                     | 15                    | 1.5·10¹⁰                        | 15.5 ± 0.9                     |
| 2.6               | 536.3                    | 137.0                     | 60                    | 4.9·10¹⁰                        | 11.7 ± 0.9                     |

**Experiment**

The experimental samples are 10.5-mm diameter discs manufactured by pressing fine-dispersed \(^{nat}\)HgO powder. The weight contents of impurities in the samples do not exceed 0.16%, of which 0.01% Si, 0.03% Cl, 0.02% Ca, 0.04% Ti, 0.03% Fe, and 0.01% Ba. The total content of the rest 60 elements, found by the spark mass-spectrometry, is below 0.02%.

The measurements were made by the relative method, using the \(^{27}\text{Al}(p,x)^{22}\text{Na} \) reaction to monitor the process. The monitors are 10.5-mm Al foils with chemical impurities below 0.001%. Two independent proton beams from the ITEP U-2 synchrotron are used to irradiate the samples, namely, the low-energy (70-200 MeV) and high-energy (800-2600 MeV) beams.

Table 1 presents the characteristics of the experimental samples and monitors together with the main irradiation parameters.

The techniques for irradiating the samples and for processing the \(\gamma\)-spectra are presented in [4] together with formulas used to determine the fragment nuclide yields.

**Results**

More than 350 yields of residual nuclei (from \(^{22}\text{Na} \) to \(^{203}\text{Hg} \)) from 0.1, 0.2, 0.8, and 2.6 GeV proton-irradiated \(^{nat}\)Hg have been measured. Figs. 1-9 show the products that were measured at all the four energies.

The experimental data were compared with the LAHET, CEM95, CEM2k, INUCL, CASCADE, and YIELDX code-simulated yields. The comparison method and a short description of all codes together with further references may be found in [4]. It should be noted that all these codes do not calculate the independent and cumulative yields individually for the ground and metastable states of the produced radionuclides, whereas the yields of either ground or metastable states alone are often measured. Therefore, those particular measurement results were excluded from the comparison procedure. The only exclusion is the case of measuring both states, so the total yields can be compared with the simulation results. Table 2 and Figs. 1-4 present the results of a detailed nuclide-by-nuclide comparison.

In Table 2, \(N_T\) is the total number of the measured yields; \(N_G\) is the number of the measured yields selected to be used in comparison with calculations; \(N_S\) is the number of the products

\(^1\)The \(^{22}\text{Na} \) and \(^{24}\text{Na} \) yields have been determined disregarding the contributions from the Al monitor samples.
Table 2: Statistics of the experimental-to-simulated yield comparisons

| Code     | $E_p = 0.1$ GeV, $N_T = 48$, $N_G = 35$ | $E_p = 0.2$ GeV, $N_T = 66$, $N_G = 49$ |
|----------|--------------------------------------|----------------------------------------|
|          | $N_{C1.3}$ / $N_{C2.0}$ / $N_S$     | $N_{C1.3}$ / $N_{C2.0}$ / $N_S$     |
|          | $<F>$ | $S(<F>)$ | $<F>$ | $S(<F>)$ | $<F>$ | $S(<F>)$ |
| LAHET    | 13/21/30 | 2.24 | 1.92 | 22/36/48 | 1.99 | 1.69 |
| CEM95    | 6/15/28  | 2.29 | 1.61 | 20/31/38 | 1.79 | 1.56 |
| CEM2k    | 9/18/28  | 1.96 | 1.52 | 22/31/38 | 1.66 | 1.48 |
| INUCL    | 10/19/33 | 2.74 | 2.05 | 12/27/48 | 2.25 | 1.66 |
| CASCADE  | 16/24/33 | 2.36 | 1.98 | 21/35/48 | 2.33 | 1.95 |
| YIELDX   | –        | –   | –   | 14/35/49 | 2.10 | 1.66 |
|          | $E_p = 0.8$ GeV, $N_T = 106$, $N_G = 88$ | $E_p = 2.6$ GeV, $N_T = 142$, $N_G = 121$ |
|          | $N_{C1.3}$ / $N_{C2.0}$ / $N_S$     | $N_{C1.3}$ / $N_{C2.0}$ / $N_S$     |
|          | $<F>$ | $S(<F>)$ | $<F>$ | $S(<F>)$ | $<F>$ | $S(<F>)$ |
| LAHET    | 42/63/87 | 2.06 | 1.73 | 23/80/118 | 2.02 | 1.49 |
| CEM95    | 30/46/59 | 2.35 | 2.13 | 46/77/91  | 2.27 | 2.11 |
| CEM2k    | 26/51/63 | 1.65 | 1.43 | 32/77/101 | 2.73 | 2.29 |
| INUCL    | 26/42/82 | 2.74 | 1.95 | 37/77/115 | 2.55 | 2.03 |
| CASCADE  | 35/59/84 | 2.52 | 2.10 | 56/93/114 | 1.76 | 1.55 |
| YIELDX   | 32/62/88 | 2.21 | 1.82 | 26/65/120 | 2.52 | 1.78 |

whose yields were simulated by a particular code; $N_{C1.3}$ is the number of the comparison events when the simulation-experiment difference does not exceed 30%; $N_{C2.0}$ is the number of the comparison events when the simulation-experiment difference does not exceed a factor of 2.

Figs. 1-4 show the results of the nuclide-by-nuclide comparison of the experimental data with the LAHET, CEM95, CEM2k, CASCADE, INUCL, and the YIELDX code-simulated results. One can see that all codes (except YIELDX) adequately predict the $A > 170$ product yields for 100, 200, and 800 MeV protons and the $A > 120$ product yields for 2600 MeV protons. The yields simulated by all the codes in the remaining ranges of masses, i.e., in the fission and fragmentation regions, are very different from experiment, with the most significant differences observed in the $80 < A < 103$ range for 100 and 200 MeV protons, in the $48 < A < 130$ range for 800 MeV protons, and in the $28 < A < 100$ range for 2600 MeV protons. It should be noted that CEM95 and CEM2k do not calculate the process of fission itself, and do not provide fission fragments and a further possible evaporation of particles from them. When, during a Monte Carlo simulation of a compound stage of a reaction using the evaporation and fission widths these codes have to simulate a fission, they simply remember this event (that permits them to calculate fission cross section and fissility) and finish the calculation of this event without a real subsequent calculation of fission fragments. Therefore, the results from CEM95 and CEM2k shown here reflect the contribution to the total yields of the nuclides only from deep spallation processes of successive emission of particles from the target, but do not contain fission products. To be able to describe nuclide production in the fission region, these codes have to be extended by incorporating a model of high energy fission (e.g., in the transport code MCNPX, where these code are used, they are complemented by the RAL fission model).

Figs. 5-8 show the simulated mass yield of the reaction products. The experimental cumulative yields, which are often equal to the mass yields within measurement errors, are also shown for comparison. The following conclusions may be drawn from the comparison between the experimental and simulated mass yields:

• in the case of 0.1 GeV protons and $A > 190$, all codes predict the mass curve shape quite
Figure 1: Nuclide-by-nuclide comparison between the experimental and simulated results for 0.1 GeV protons. The cumulative yields are labeled with a “c” when the respective independent yields are also shown.

adequately,

- in the case of 0.2 GeV protons and A > 180, all code-simulated yields are in a good agreement with the data,
- in the case of 0.8 GeV protons, the best agreement with experiment is obtained by YIELDX, for A > 130 and by INUCL, for A < 130,
- in the case of 2.6 GeV protons and A > 100, the CEM95 and CASCADE results agree with the data, while the LAHET calculations are underestimated and the YIELDX and INUCL yields represent the mass curve shape erroneously. None of the codes can describe well the experimental curve shape at A < 100.

Fig. 9 illustrates the dependence of a part of measured yields on the proton energy, i.e., excitation functions. These data will be analyzed further after a final release of all the experimental results.

Conclusion

Our experiment-to-simulation comparison has shown that, on the whole, the simulation codes can but poorly predict the experimental results. In some cases, the differences reach an
order of magnitude or even more. The last version of the improved cascade-exciton model code, CEM2k [5], shows the best agreement with the data in the spallation region at all energies except 2.6 GeV, where the model overestimates the expected experimental fission cross section of about 75 mb [6] by a factor of 4. This overestimation of the fission cross section causes an underestimation of the yield of nuclei which are most likely to fission at the evaporation stage of a reaction, after the cascade and preequilibrium stages, i.e., for $170 < A < 185$. (Similar disagreement with the data one can see as well for LAHET and CEM95, that is also related with an overestimation of the fission cross section at 2.6 GeV). The code CEM2k is still under development, its problem with the overestimation of fission cross sections at energies above 1 GeV has yet to be solved, and it has to be complemented with a model of fission fragment production as mentioned above, to be able to describe as well fission products.

This means that the nuclear data must be accumulated persistently in the above-mentioned ranges of energies and masses, to help improve hadron-nucleus interaction models released in codes used in applications.

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Figure 3: Nuclide-by-nuclide comparison between the experimental and simulated results for 0.8 GeV protons. The cumulative yields are labeled with a “c” when the respective independent yields are also shown.

References

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Figure 4: Nuclide-by-nuclide comparison between the experimental and simulated results for 2.6 GeV protons. The cumulative yields are labeled with a “c" when the respective independent yields are also shown.
Figure 5: The simulated and experimental mass yields at 0.1 GeV.

Figure 6: The simulated and experimental mass yields at 0.2 GeV.
Figure 7: The simulated and experimental mass yields at 0.8 GeV.

Figure 8: The simulated and experimental mass yields at 2.6 GeV.
Figure 9: Some experimental and simulated yields versus the proton energy.
[4] Yu. E. Titarenko et al., “Experimental and Computer Simulation Study of the Radionuclides Produced in Thin $^{209}$Bi Targets by 130 MeV and 1.50 GeV Proton-Induced Reactions,” Nucl. Instr. Meth. A 414, 73 (1998); Yu. E. Titarenko et al., “Study of Residual Product Nuclide Yields in 1.0 GeV Proton Irradiated $^{208}$Pb and 2.6 GeV Proton Irradiated natW Thin Targets,” Proc. this (SATIF-5) meeting.

[5] The code CEM2k is briefly surveyed in S. G. Mashnik, L. S. Waters, and T. A. Gabriel, “Models and Codes for Intermediate Energy Nuclear Reactions.” Proc. 5th Int. Workshop on Simulating Accelerator Radiation Environments (SARE5), July 17-18, 2000, OECD Headquarters, Paris, France, and will be described by S. G. Mashnik and A. J. Sierk in a future paper.

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Yields in nat-Hg irradiated with 0.1-2.6GeV protons

Proton energy [MeV] - Cross section [mbarn]

- $^{82}$Br
- $^{95}$Zr
- $^{95}$Nb
- $^{103}$Ru
- $^{101m}$Rh
- $^{188}$Ir
- $^{188}$Pt
- $^{189}$Pt
- $^{191}$Pt
- $^{192}$Au
- $^{193}$Au
- $^{194}$Au
- $^{196}$Au
- $^{198}$Au
- $^{200m}$Au
- $^{192}$Hg
- $^{193m}$Hg
- $^{195m}$Hg
- $^{197m}$Hg
- $^{199m}$Hg
- $^{203}$Hg
- $^{196m}$Tl
- $^{200}$Tl
- $^{202}$Tl

Cross section values are given in units of barns (mbarn).