Nuclide Production in $^{197}$Au, $^{208}$Pb, and $^{nat}$U Irradiated with 0.8-1 GeV Protons: Comparison with other Experiments and with Theoretical Predictions

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We used the ITEP proton synchrotron U10 to irradiate isotopically-enriched $^{208}$Pb and $^{nat}$U thin targets with 1.0 GeV protons and $^{197}$Au thin targets with 0.8 GeV protons. More than 400 cross sections of the nuclides produced were measured using the direct $\gamma$-spectrometry method with a high-resolution Ge detector. The measured $\gamma$-spectra were interactively processed by the GENIE2000 code. The $\gamma$-lines were identified, and the cross sections determined, by the ITEP-developed SIGMA code using the PCNUDAT database.

The measured cross sections are compared with similar data obtained at GSI for kinematically inverse reactions of 1 GeV/nucleon $^{208}$Pb, 1 GeV/nucleon $^{238}$U, and 0.8 GeV/nucleon $^{197}$Au interacting with a hydrogen target and with the ZSR data on $^{nat}$Pb and $^{197}$Au irradiated with 1 and 0.8 GeV protons, respectively. Our results are on average ~10 to 20% higher than the GSI data. The measured data are analyzed with the LANL codes CEM2k+GEM2 and LAQGSM+GEM2 and with the INCL intranuclear cascade code from Liege merged with the GSI evaporation/fission code ABLA. The obtained results may help to find possible sources of some observed discrepancies, thereby increasing the reliability of the data (both measured and simulated) required for different accelerator applications.

INTRODUCTION

Development of high-energy hadron-nucleus interaction models and codes requires they are validated against reliable experimental data. Among such data are cross sections for residual nuclide production measured at different laboratories, for instance, at the Gesellschaft fuer Schwerionenforschung (Darmstadt, GSI), Zentrum fur Strahlenschutz und Radioökologie (Universitaet Hannover, ZSR), and the Institute for Theoretical and Experimental Physics (Moscow, ITEP).

The ITEP and ZSR measurements of the product cross sections are based on proton irradiation of experimental samples and subsequent $\gamma$-spectrometric analysis (called hereafter the "direct kinematics" method)1-5). The GSI approach is fundamentally different and involves accelerated ion irradiation of a liquid hydrogen target with a subsequent recording of the mass and charges of the nuclei produced from the projectile-nuclei (the "inverse kinematics" method)6-8).

In view of the above, it is topical to compare the different experimental data sets among each other and with results by modern theoretical codes, such as CEM2k+GEM2, LAQGSM+GEM2, and INCL+ABLA that have been recently developed at Los Alamos and Liege.

COMPARISON BETWEEN THE "DIRECT" AND "INVERSE" KINEMATICS DATA

The features and results of the direct and inverse kinematics methods are described in detail in1-9). The results of three inverse kinematics experiments at GSI6-8) may be compared with the direct

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kinematics results, namely, the $^{197}\text{Au}$ and $^{208}\text{Pb}$ GSI measurements at 0.8 and 1.0 GeV/nucleon, may be compared with the ITEP and ZSR data, respectively, and the $^{238}\text{U}$ GSI measurements at 1 GeV/nucleon may be compared with the ITEP data. Comparison of results by these two different methods has to allow for the fact that the direct kinematics method provides mainly cumulative cross sections and just a small fraction of independent cross sections, whereas the inverse kinematics method determines only independent cross sections. Also, the inverse kinematics method does not separate metastable and ground states that can be measured by the direct kinematics method. These circumstances were taken into account in our previous works where our ITEP experimental $^{208}\text{Pb}$ and $^{197}\text{Au}$ results were compared with the GSI and ZSR data. In the present work, as an addition to we compare our data on 1.0 GeV proton-irradiated $^{nat}\text{U}$ with the GSI inverse kinematics measurements and also perform a detailed comparison of all three experimental data sets with theoretical results by simulation codes CEM2k+GEM2, LAQGSM+GEM2, and INCL+ABLA. Table 1 shows results of comparing the direct and inverse kinematics experimental data for all three reactions. The mean cross section ratios $\langle \sigma_{\exp A,i}/\sigma_{\exp B,i} \rangle$ and the mean squared deviation factors $<F>$ were chosen as quantitative criteria for this comparison. Fig. 1 shows another comparison of these experimental data sets, namely for the residual product mass distributions. Considered together, the mass distributions and the two quantitative criteria make it possible to judge with a higher confidence both about the level of systematic differences in the compared data (the cross section ratio) and about the differences proper (the mean squared deviation factor).

**Table 1.** Results of quantitative comparison between the direct and inverse kinematics experimental data.

| Comparison          | $\langle \sigma_{\exp A,i}/\sigma_{\exp B,i} \rangle$ | $<F>$ |
|---------------------|-------------------------------------------------------|-------|
|                     | $^{197}\text{Au} + p$ 0.8GeV  | $^{208}\text{Pb} + p$ 1.0GeV | $^{nat}\text{U} + p$ 1.0GeV | $^{197}\text{Au} + p$ 0.8GeV  | $^{208}\text{Pb} + p$ 1.0GeV | $^{nat}\text{U} + p$ 1.0GeV |
| GSI/ITEP            | 0.82                                                   | 0.89  | 0.77       | 1.40                                                   | 1.30                                                   | 1.83                          |
| GSI/ZSR             | 0.83                                                   | 0.81  | -          | 1.99                                                   | 1.50                                                   | -                             |
| ITEP/ZSR            | 1.16                                                   | 0.93  | -          | 1.30                                                   | 1.24                                                   | -                             |

* $<\sigma_{\exp A,i}/\sigma_{\exp B,i}>, <F>$ were calculated as:
$<\sigma_{\exp A,i}/\sigma_{\exp B,i}> = 10^{**<R_i>}, <F> = 10^{**\sqrt<(R_i^2>)}$, where $R_i = \log_{10}(\sigma_{\exp A,i}/\sigma_{\exp B,i})$

The following conclusions can be drawn from results presented in Table 1:

- We see a stable correlation between the values of the cross section ratios obtained by the direct kinematics method at ITEP and ZSR on the one hand, and by the fundamentally different inverse kinematics method at GSI, on the other hand. Namely, the cross section values obtained by the inverse kinematics method for all three reactions studied here are on average ~10-20% lower compared with those obtained by the gamma-spectrometry method. Interesting correlations can be also traced for values of the factor $<F>$: We see that, on average, our ITEP data are closer with the ZSR measurements and agree a little worse with the GSI data, while the biggest disagreement is observed between the ZSR and GSI measurements.

- Fig. 1 shows a significant difference in the experimental mass distributions for Au in the region A~110-140, that is intermediate between the fission and spallation modes. The mass yields for A=113 and 127 (ZSR) and 121 (ITEP) measured by the direct kinematics method are significantly higher (up to an order of magnitude) in comparison to the values obtained by inverse kinematics method (GSI). We note that there is no ground to assume a methodological error in
the gamma-spectrometry measurements of these products from p+Au, as the yields of nuclides with the same masses from p+Pb measured by the same method agree well with the GSI data.

The observed discrepancies are not yet completely clear to us and should be overcome by further measurements.

THEORETICAL RESULTS

In the present work, we have simulated all studied measured cross sections with the Liege intranuclear cascade model INCL\textsuperscript{10} in conjunction with the GSI evaporation/fission model ABLA\textsuperscript{11} (INCL+ABLA) and using the improved Cascade-Exciton Model (CEM) code CEM2k\textsuperscript{12} and the Los Alamos version of the Quark-Gluon String Model code LAQGSM\textsuperscript{13} both merged\textsuperscript{14} with the Generalized-Evaporation Model code GEM2 of Furihata\textsuperscript{15}. Table 2 shows results of a quantitative experiment-to-simulation comparison, using the same criteria we compared different experimental sets in Table 1. Fig. 1 shows a qualitative comparison of calculation results with the data, presenting experimental and simulated mass distributions for all three reactions studied here.

Table 2. Quantitative comparison the ITEP, ZSR, and GSI experimental data with results by the LAQGSM+GEM2, CEM2k+GEM2, and INCL+ABLA codes.

| Data set | \(\frac{\sigma_{\text{calc}}}{\sigma_{\text{exp}}} \times 10^3\) | \(\frac{\sigma_{\text{calc}}}{\sigma_{\text{exp}}} \times 10^3\) | \(\frac{\sigma_{\text{calc}}}{\sigma_{\text{exp}}} \times 10^3\) | \(\frac{\sigma_{\text{calc}}}{\sigma_{\text{exp}}} \times 10^3\) | \(\frac{\sigma_{\text{calc}}}{\sigma_{\text{exp}}} \times 10^3\) |
|---|---|---|---|---|---|
| \(^{197}\text{Au} + p\ 0.8\text{GeV}\) | | | | | |
| ITEP | 0.69 | 0.85 | 0.63 | 1.83 | 1.63 | 2.07 |
| ZSR | 0.49 | 0.73 | 0.45 | 2.53 | 2.45 | 2.94 |
| GSI | 0.75 | 0.99 | 0.79 | 1.96 | 2.17 | 1.89 |
| \(^{208}\text{Pb} + p\ 1.0\text{GeV}\) | | | | | |
| ITEP | 0.66 | 0.87 | 0.63 | 1.86 | 1.90 | 2.07 |
| ZSR | 0.55 | 0.71 | 0.57 | 2.26 | 1.94 | 2.30 |
| GSI | 0.68 | 0.83 | 0.84 | 2.10 | 2.58 | 1.88 |
| \(^{235}\text{U} + p\ 1.0\text{GeV}\) | | | | | |
| ITEP | 0.58 | 0.64 | 0.63 | 2.45 | 2.50 | 2.04 |
| GSI | 0.89 | 0.99 | 0.84 | 2.11 | 2.67 | 1.90 |

From the quantitative and qualitative comparisons presented in Table 2 and Fig. 1 one can see that, on the average, all three codes provide a reasonable good agreement with all three experimental data sets and none of the tested codes shows a decisive advantage over other codes for the reactions studied here.

At the same time, partial comparison of results for a given reaction or for production of specific nuclei demonstrates different degrees of agreement of tested codes with experimental data from different sets:

- The LAQGSM+GEM2 and INCL+ABLA codes give very similar calculated-to-experimental mean cross section ratios when compared with data from each of three groups (the mean difference is around \(\sim 40\%\), see Table 2). At the same time, the CEM2k+GEM2 code gives, as a rule, much lower values of systematic deviations (\(\sim 10-20\%\) on the average) that are comparable with the respective values for the "experimental" comparisons. On the average, looking both at mean
cross section ratios and mean square deviation factors, the CEM2k+GEM2 code seems to agree better with all experimental data sets, though the calculated cross sections are on the average below the experimental data in all cases;

- On the average, we see that the INCL+ABLA code agrees better with the GSI data and not so well with the ITEP and ZSR measurements, and just an opposite situation for CEM2k+GEM2 and LAQGSM+GEM2.

- The observed systematic different agreement of different codes with different data sets suggests that we should not limit ourselves to comparisons with experimental data from only one type of measurements when developing and benchmarking nuclear reaction models and codes.

Fig. 1. Experimental and theoretical mass distributions of product nuclides from 0.8 GeV p + Au and 1.0 GeV p + Pb and U (The $^{208}$Pb GSI data are from $^{16}$).

ACKNOWLEDGMENT

The authors are indebted to Drs. S. Leray and A. Boudard of CEA-Saclay for providing us with the latest version of the code INCL+ABLA and to Prof. K.-H. Schmidt and Dr. T. Enqvist of GSI-Darmstadt for providing us numerical data of the GSI measurements and fruitful discussions.
This work was partly carried out under the ISTC Projects # 839 and 2002 and was partly supported by the U.S. Department of Energy, Moldovan-U. S. Bilateral Grants Program, CRDF Project MP2-3045, and by the NASA Astrophysical Theory Program grant, Project #NRA-01-01-ATP-066.

REFERENCES

1. YU. TITARENKO, O. SHVEDOV, M. IGUMNOV, et al. Experimental and Theoretical Study of the Yields of Radionuclides Produced in $^{209}$Bi Thin Target Irradiated by 1500 MeV and 130MeV Protons. Nucl. Instr. and Meth., A 414, 73 (1998).

2. YU. TITARENKO, O. SHVEDOV, V. BATYAEV, et al. Cross sections for nuclide production in 1 GeV proton irradiated $^{208}$Pb. LANL Report LA-UR-00-4779 (2000); Phys. Rev. C65, 064610 (2002).

3. YU. E. TITARENKO, V. F. BATYAEV, V. M. ZHIVUN et al., Cross Sections for Nuclide Production in 1 GeV Proton-Irradiated 208Pb and 0.8 GeV Proton-Irradiated 197Au, E-print: nucl-ex/0305024; to be published in Proc. AccApp'03 Embedded Topical Meeting "Accelerator Applications in a Nuclear Renaissance," San Diego, California, USA, June 1-5, 2003.

4. M. GLORIS, R. MICHEL, F. SUDBROK, et al. Proton-Induced Production of Residual Radionuclides in Lead at Intermediate Energies, Nucl. Instr. and Meth., A463, 593 (2001); EXFOR file O0500.

5. R. MICHEL, R. BODEMANN, H.BUSEMANN, et al. Cross sections for the production of residual nuclides by low- and medium-energy protons from the target elements C, N, O, Mg, Al, Si, Ca, Ti, V, Mn, Fe, Co, Ni, Cu, Sr, Y, Zr, Nb, Ba and Au. Nucl. Instr. and Meth., B129, 153 (1997).

6. T. ENQVIST, W. WLAZLO, P. ARMBRUSTER et al., Isotopic Yields and Kinetic Energies of Primary Residues in 1 A GeV 208Pb + p Reactions, Nucl. Phys. A686, 481 (2001); see also: Primary-Residue Production Cross Sections and Kinetic Energy in 1 A GeV 208Pb on Deuteron Reactions, Nucl. Phys. A703, 435 (2002).

7. J. BENLLIURE, P. ARMBRUSTER, M. BERNAS et al., Isotopic Production Cross Sections of Fission Residues in $^{197}$Au-on-Proton Collisions at 800 A MeV, Nucl. Phys. A683, 513 (2001); F. REJMUND, B. MUSTAPHA, P. ARMBRUSTER et al., Measurement of Isotopic Cross Sections of Spallation Residues in 800 A MeV $^{197}$Au + p Collisions, Nucl. Phys. A683, 540 (2001).

8. J. TAIEB, K.-H. SCHMIDT, L. TISSAN-GOT et al., Evaporation Residues Produced in the Spallation Reaction $^{238}$U+p at 1 A GeV, Nucl. Phys. A724, 413 (2003); Fission-Residues Produced in the Spallation Reaction 238U +p at 1 A GeV, Nucl. Phys. A725, 213 (2003).

9. Yu.E. TITARENKO, et al., Experimental and Theoretical Study of the Yields of Residual Product Nuclei in Thin Targets Irradiated by 100 - 2600 MeV Protons. ISTC #839 project final technical report; http://www-nds.iaea.org/indc_sel.html, NDC(CCP)-434; EXFOR file O0986.

10. BOUDARD, J. CUGNON, S. LERAY, et al., Intranuclear cascade model for a comprehensive description of spallation reaction data, Phys. Rev. C66, 044615 (2002)
11. J.-J. GAIMARD and K.-H. SCHMIDT, A Reexamination of the Abrasion-Ablation Model for the Description of the Nuclear Fragmentation Reaction, Nucl. Phys. A531, 709 (1991); A. R. JUNGHANS, M. de JONG, H.-G. CLERC et al., Projectile-Fragment Yields as a Probe for the Collective Enhancement in the Nuclear Level Density, Nucl. Phys. A629, 635 (1998).

12. S. G. MASHNIK and A. J. SIERK, CEM2k - Recent Developments in CEM, Proc. of the AccApp'00 Embedded Topical Meeting "Nuclear Applications of Accelerator Technology", Washington, DC, USA, November 12-16, 2000, American Nuclear Society, La Grange Park, IL, 2001, pp. 328-141; E-print: nucl-th/0011064; Recent Developments of the Cascade-Exciton Model of Nuclear Reactions, Proc. Int. Conf. on Nucl. Data for Sci. and Techn. (ND2001), Tsukuba, Ibaraki, Japan, October 7-12, 2001, Journ. of Nucl. Sci. and Techn., Suppl. 2, 720 (2002); E-print: nucl-th/0208074.

13. K. K. GUDIMA, S. G. MASHNIK, and A. J. SIERK, User Manual for the code LAQGSM, Los Alamos National Laboratory Report LA-UR-01-6804; http://lib-www.lanl.gov/lanpubs/00818645.pdf.

14. S. G. MASHNIK, K. K. GUDIMA, and A. J. SIERK, Merging the CEM2k and LAQGSM Codes with GEM2 to Describe Fission and Light-Fragment Production, E-print: nucl-th/0304012; to be published in Proc. 6th Int. Workshop on Shielding Aspects of Accelerators, Targets and Irradiation Facilities (SATIF-6), Stanford Linear Accelerator Center, CA 94025, USA, April 10-12, 2002; S. G. MASHNIK, A. J. SIERK, and K. K. GUDIMA, Complex Particle and Light Fragment Emission in the Cascade-Exciton Model of Nuclear Reactions, Invited talk presented at the 12th Biennial Topical Meeting of the Radiation Protection and Shielding Division (RPSD) of the American Nuclear Society, Santa Fe, NM, USA, April 14-17, 2002, E-print: nucl-th/0208048; M. BAZNAT, K. GUDIMA, and S. MASHNIK, Proton-Induced Fission Cross Section Calculation with the LANL Codes CEM2k+GEM2 and LAQGSM+GEM2, E-print: nucl-th/0307014, to be published in Proc. AccApp'03 Embedded Topical Meeting "Accelerator Applications in a Nuclear Renaissance," San Diego, California, USA, June 1-5, 2003.

15. S. FURIHATA, Statistical analysis of light fragment production from medium energy proton-induced reactions, Nucl. Instr. Meth. B171, 252 (2000); The Gem Code Version 2 Users Manual, Mitsubishi Research Institute, Inc., Tokyo, Japan (2001); Development of a Generalized Evaporation Model and Study of Residual Nuclei Production, Ph.D. thesis, Tohoku University (2003).

16. T. ENQVIST, private communication, 2003, and\textsuperscript{6).}