

LA-UR-04- 8090

Approved for public release;  
distribution is unlimited.

*Title:* Excitation Functions of Products from 208,207,206,nat-Pb and 209-Bi(p,x) Reactions Measured in the 40-2600 MeV Energy Range and Predicted Theoretically

*Author(s):* Yu. E. Titarenko, V. F. Batyaev, V. M. Zhivun, R. D. Mulambetov, S. V. Mulambetova, S. L. Zaitsev, S. G. Mashnik, and R. E. Prael

*Submitted to:* Proc. Int. Conf. on Nuclear Data for Science and Technology (ND2004), September 26 - October 1, 2004, Santa Fe, New Mexico, USA



Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the University of California for the U.S. Department of Energy under contract W-7405-ENG-36. By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.

Form 836 (8/00)



# Excitation Functions of Products from $^{208,207,206,\text{nat}}\text{Pb}$ and $^{209}\text{Bi}$ (p,x) Reactions Measured in the 40-2600 MeV Energy Range and Predicted Theoretically

Yu.E. Titarenko, V.F. Batyaev, V.M. Zhivun, R.D. Mulambetov, S.V. Mulambetova, S.L. Zaitsev, S.G. Mashnik<sup>1</sup>, R.E. Prael<sup>1</sup>

*Institute for Theoretical and Experimental Physics (ITEP), 117218 Moscow, Russia  
<sup>1</sup> Los Alamos National Laboratory, Los Alamos, NM 87545, USA*

**Abstract.** More than 5000 independent and cumulative yields of radioactive residual product nuclides with lifetimes ranging from 13.2 min ( $^{187}\text{Re}$ ) to 31.55 years ( $^{207}\text{Bi}$ ) are measured in the  $^{208, 207, 206, \text{nat}}\text{Pb}$  and  $^{209}\text{Bi}$  thin targets irradiated by 0.04, 0.07, 0.10, 0.15, 0.25, 0.40, 0.60, 0.80, 1.20, 1.60, and 2.60 GeV external proton beams from the ITEP U10 accelerator. The independent and cumulative measured yields of residual products in thin lead and bismuth targets irradiated with 0.04-2.6 GeV protons are compared with results by the LAHET, CEM03, LAQGSM03, INCL+ABLA, CASCADE, CASCADO and LAHETO codes, in order to evaluate the predictive power of the codes in this energy region. We found that the predictive power of the tested codes is different but is satisfactory for most of the nuclides in the spallation region, though none of the codes agree well with the data in the whole mass region of product nuclides and all should be improved further. On the whole, the predictive power of all codes for the data in the fission and fragmentation product regions and, especially, at the borders between spallation and fission and between fission and fragmentation regions is much worse than in the spallation region; therefore, development of better evaporation/fission/fragmentation models is of first priority.

## INTRODUCTION

A number of current and planned nuclear projects, such as transmutation of nuclear wastes with Accelerator-Driven Systems (ADS) require a large amount of nuclear data. Since not all the required data can be measured, the reliable models and codes are to be used in the projects. The codes should be verified, validated, and benchmarked against the measurements that are as reliable as possible.

During 2002 - 2004, under the ISTC Project # 2002 [1], ITEP has realized an experimental program to measure the residual nuclide production cross sections in  $^{208, 207, 206}\text{Pb}$ ,  $^{209}\text{Bi}$  and  $^{209}\text{Pb}$  thin targets irradiated with protons of 0.04, 0.07, 0.10, 0.15, 0.25, 0.4, 0.6, 0.8, 1.2, 1.6 and 2.6 GeV. In the present work, we also analyze all our measured data with seven codes used in many applications in order to validate their predictive powers.

## EXPERIMENT

The thin  $^{208, 207, 206, \text{nat}}\text{Pb}$  and  $^{209}\text{Bi}$  targets of 10.5 mm diameter (127 - 358 mg/cm<sup>2</sup> thickness) together with aluminum monitors of the same diameter (127 - 254 mg/cm<sup>2</sup> thickness) were irradiated using external beam of ITEP U10 proton synchrotron. The targets used were of the following isotopic composition:  $^{208}\text{Pb}$  (0.87%  $^{206}\text{Pb}$ , 1.93%  $^{207}\text{Pb}$ , 97.2%  $^{208}\text{Pb}$ );  $^{207}\text{Pb}$  (0.03%  $^{204}\text{Pb}$ , 2.61%  $^{206}\text{Pb}$ , 88.3%  $^{207}\text{Pb}$ , 9.06%  $^{208}\text{Pb}$ );  $^{206}\text{Pb}$  (94.0%  $^{206}\text{Pb}$ , 4.04%  $^{207}\text{Pb}$ , 1.96%  $^{208}\text{Pb}$ );  $^{209}\text{Pb}$  (1.4%  $^{204}\text{Pb}$ , 24.1%  $^{206}\text{Pb}$ , 22.1%  $^{207}\text{Pb}$ , 52.4%  $^{208}\text{Pb}$ );  $^{209}\text{Bi}$  > 99.9%. The  $^{27}\text{Al}(\text{p},\text{x})^{22}\text{Na}$  reaction was used for monitoring the proton flux. The proton fluencies were from  $3.1 \times 10^{13}$  to  $1.4 \times 10^{14}$  p/cm<sup>2</sup>. The produced radionuclides were detected using the direct gamma-spectrometry via the Ge detector with a 1.8 keV resolution at a 1332 keV  $^{60}\text{Co}$  gamma-line. Each of the irradiated targets was measured during 3 to 6 months. The gamma spectra were processed via the interactive mode of the GENIE200 program using preliminary results of an automatic mode processing. The results of gamma-spectra processing are the input to the SIGMA code, which identifies the measured

gamma-lines using the PCNUDAT nuclear decay database and determines the cross sections of the found radionuclides. The details of experimental techniques are described in [2].

As a result, more than 5500 nuclide production cross sections were measured in 55 experiments. The data themselves and their graphical interpretation will be presented in the final technical report of the ISTC Project #2002 and will be uploaded into the EXFOR data base.

## THEORETICAL MODELING

Seven codes were used for simulation of the measured cross sections: LAHET [5], CEM03 [6], LAQGSM+GEM2[7], INCL4+ABLA[8, 9], CASCADE [10], CASCADO and LAHETO [11].

The modeling was carried out at 25 energies from 0.03 to 3.5 GeV to produce smooth excitation functions (EF). To make the comparison to experimental data (ED) correct, the required cumulative yields were calculated on the base of simulated independent yields. The metastable products were not simulated. We compared simulated and experimental EF both qualitatively (plots) and quantitatively. For our qualitative comparison, 1036 figures with EF by the seven codes and ED obtained by us under the ISTC Project #2002 have been drawn. As an example, some of those figures are presented here in Figs. 1 and 2. For our quantitative comparison,

we chose the mean simulated-to-experiment squared deviation factor  $\langle F \rangle$ , as described in [2].

To understand how different codes agree with the data in different nuclide production regions, we divided conventionally all products into four groups: shallow spallation products ( $A > 170$ ), deep spallation products ( $140 < A < 170$ ), fission products ( $30 < A < 140$ ), and fragmentation products ( $A < 30$ ). Tab. 1 presents averaged mean deviation factors  $\langle F \rangle$  for all these four conventional regions separately. From the presented table and figures and very many other results not shown here due to limited size of this paper we can conclude:

1)  $A > 170$  (shallow spallation products): Most of the products from this region are predicted satisfactorily, with a mean deviation factor less than 2. Deviations above a factor of two are observed, as a rule, for independent yields (e.g., for  $^{192}\text{Ir}$ ), for (p,xn) reactions, and for near-threshold energies. Also, the deviations between ED and simulated EF's as well as between results by different codes increase at energies above 1 GeV. The near-target products ( $A$  above 200) are predicted variously at different proton energies: For instance, CEM03 predicts such products with  $\langle F \rangle \sim 1.5$  at energies below 1 GeV, but underestimates them significantly ( $\langle F \rangle \sim 6$ ) at energies above 1 GeV. On the contrary, LAHET and LAQGSM predict these products with  $\langle F \rangle \sim 1.5\text{-}2$  at energies above 0.1 GeV, but fail to do so well at lower energies ( $\langle F \rangle \sim 4\text{-}5$ ). The same is true for INCL+ABLA:  $\langle F \rangle \sim 1.3\text{-}1.5$  at  $E_p > 0.1$  GeV,  $\langle F \rangle \sim 6$  at  $E_p < 0.1$  GeV.

TABLE 1. Mean squared deviation factors  $\langle F \rangle$  for different ranges of products in three energy groups:  
 $<0.1 / 0.1\text{-}1.0 / >1.0$  GeV.

Code	Product mass ( $A$ ), Proton energy group ( $E_p$ , GeV)											
	$A > 170$				$140 < A < 170$				$30 < A < 140$			
	$<0.1$	0.1- 1.0	$>1.0$	$<0.1$	$0.1-$ 1.0	$>1.0$	$<0.1$	$0.1-$ 1.0	$>1.0$	$<0.1$	0.1- 1.0	$>1.0$
ISABEL	4.64	1.68	1.70	-	2.05	1.88	7.94	3.17	2.43	-	341	26.8
BERTINI	4.20	1.60	1.70	-	2.48	1.88	5.84	2.98	2.43	-	420	26.8
INCL4+ABLA	5.10	1.95	1.42	-	8.66	3.36	1.94	1.93	2.48	-	-	49.3
CASCADE	2.95	1.63	1.83	-	2.60	1.93	168	10.8	3.66	-	-	144
CASCADE-2004	4.04	1.86	1.72	-	2.22	1.53	116	6.53	3.84	-	-	63.9
LAQGSM+GEM2	3.25	1.99	1.93	-	6.60	1.72	2.37	2.92	2.59	-	42.0	15.1
CEM03	2.23	1.65	2.50	-	2.96	1.68	1.40	1.91	2.63	-	14.4	7.70
CASCADO	3.05	2.10	1.78	-	5.27	1.45	1.38	2.52	3.29	-	176	58.8
LAHETO	6.21	1.66	-	-	2.33	-	3.09	1.99	-	-	-	-

2)  $140 < A < 170$  (deep spallation products). With decreasing the mass of the products (excitation energy after the intranuclear cascade stage of a reaction increases), the predictive power of all the

codes also decreases. The degradation of the predictive power of different codes varies. For example, for LAHET,  $\langle F \rangle$  increases up to only 2.2; for LAQGSM,  $\langle F \rangle$  increases up to 2.5; for YIELDX,

$\langle F \rangle$  increases up to  $\sim 3$ ; and in the case of INCL+ABLA,  $\langle F \rangle$  increases up to 6. The INCL+ABLA underestimates significantly the deep spallation products, thus overestimating their threshold energies. Note also that the thresholds of some reactions predicted by different codes may vary

LAHET(isabel-solid, bertini-dashed)  
CASCADE(improved-solid, obsolete-dashed)  
INCL4+ABLA  
CEM03  
CASCADO  
LAQGSM+GEM2

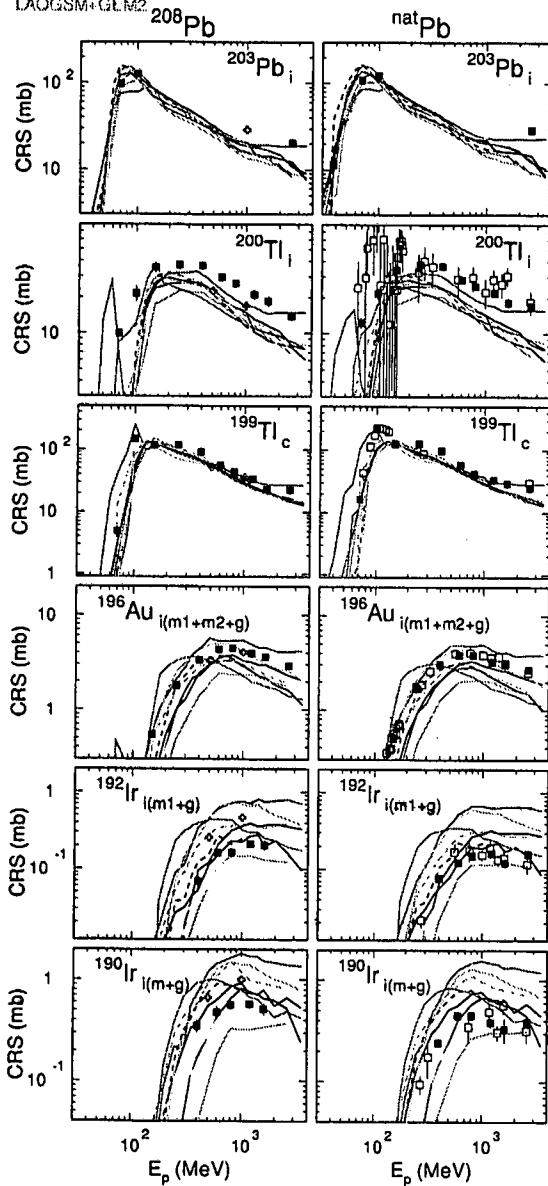


Fig. 1. Experimental and simulated excitation functions of  $^{203}\text{Pb}$ ,  $^{200}\text{Tl}$ ,  $^{199}\text{Tl}$ ,  $^{196}\text{Au}$ ,  $^{192}\text{Ir}$ , and  $^{190}\text{Ir}$  produced in  $^{208}\text{Pb}$  (left) and  $^{nat}\text{Pb}$  (right). (■ – this work, □ – [4], □ – [3]) (LAHET –black (ISABEL – solid, BERTINI – dashed), CEM03 – magenta, INCL4+ABLA – red, CASCADE – green, LAQGSM+GEM2 – green-blue, CASCADO – blue).

by up to hundreds of MeV. For example, the threshold for the production of  $^{146}\text{Eu}$  predicted by different codes varies from 600 to 1200 MeV. On the whole, it seems to us that LAHET predicts most adequately most of the measured reaction thresholds in comparison with other codes tested here.

LAHET(isabel-solid, bertini-dashed)  
CASCADE(improved-solid, obsolete-dashed)  
INCL4+ABLA  
CEM03  
CASCADO  
LAQGSM+GEM2

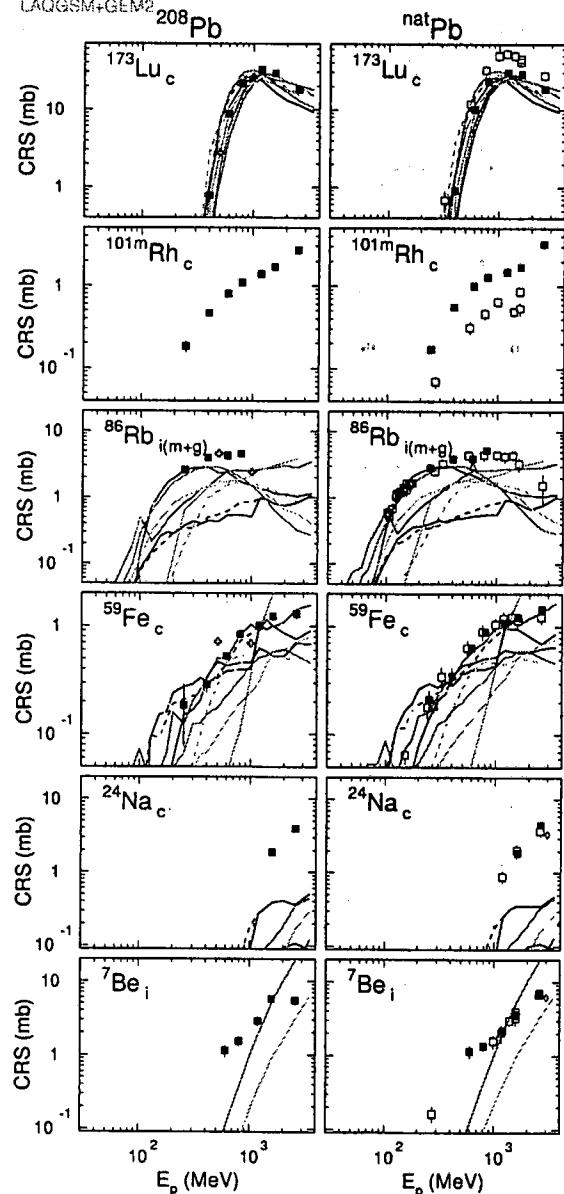


Fig. 2. The same as Fig. 3 but for  $^{173}\text{Lu}$ ,  $^{101m}\text{Rh}$ ,  $^{86}\text{Rb}$ ,  $^{59}\text{Fe}$ ,  $^{24}\text{Na}$ , and  $^7\text{Be}$ .

3) **Fission products (FP)** present about a third of all measured and analyzed here nuclides, and are described by the codes worse than the spallation products. The deviation between ED and simulated EF's as well as between different calculations themselves are much bigger than for the spallation products. LAHET and CEM03 show the best stability of the predictive power for fission products with  $\langle F \rangle$  from 1.5 to 3. LAQGSM shows somewhat bigger deviation from ED ( $\langle F \rangle$  up to 4), however, the agreement is better in the  $80 < A < 110$  region, with  $\langle F \rangle$  around 2. A peculiar agreement is demonstrated by the code INCL+ABLA:  $\langle F \rangle$  is too high (up to 6) in the  $120 < A < 140$  region where FP's overlap with deep spallation products, however, its agreement becomes the best ( $\langle F \rangle$  from 1.5 to 2.0) in comparison with other codes for fission products with  $A < 120$ . YIELDX and CASCADE show the worst agreement on FP's ( $\langle F \rangle$  up to  $\sim 20$ ). Note that most of simulated EF's are below ED in the fission region, i.e. the fission mode seems to be underestimated by the codes. The agreement of calculations with the fission data varies with the proton energy. For example, INCL+ABLA underestimates FP's at energies from  $\sim 0.1$  to  $\sim 1$  GeV, shows a good agreement at  $\sim 1$  GeV, and overestimates them at higher energies. CEM03 predicts most of FP at relatively low energies ( $< \sim 0.5$  GeV) much better than at higher energies.

4) **The fragmentation products** are significantly underestimated by all codes tested here. Only a few fragmentation products were measured and can be compared here with calculation results. These measured fragment yields are underestimated by an order of magnitude and more. As a whole, YIELDX results for these fragments are most closed to ED. However,  $^7\text{Be}$ , in particular, is best predicted by CEM03 and LAQGSM.

Finally, we like to mention that as the gamma-spectrometry method used to obtain all experimental data analyzed here allows measuring only part of the products from a nuclear reaction, our comparison cannot pretend to be universal and to choose the best from the tested codes. Rather, it points on some separate problems each code still has, helping the authors of the codes to further improve them.

## CONCLUSIONS

The predictive power of the tested codes is different but was found to be satisfactory for most of the nuclides in the spallation region, though none of the benchmarked codes agrees well with all data in the whole mass region of product nuclides and all

codes should be improved further. On the whole, the predictive power of all codes for the data in the fission product region is worse than in the spallation region; the agreement is even worse in the fragmentation region and on the border between spallation and fission regions. Therefore, development of better evaporation/fission/fragmentation models is of first priority.

## ACKNOWLEDGMENT

The work has been performed under the ISTC Project #2002 supported by the European Community. Also, the work was partially supported by the U. S. Department of Energy, the NASA ATP01 Grant NRA-01-ATP-066, and the CRDF Project MP2-3045.

## REFERENCES

1. ISTC Project #2002, Proton-Pb and Proton-Bi Reaction Yields. <http://tech-db.istc.ru/istc/db/projects.nsf/all-projects/2002>.
2. Titarenko Yu. E. et al. Experimental and Theoretical Study of the Yields of Residual Product Nuclei in Thin Targets Irradiated by 100 – 2600 MeV Protons. IAEA, Nuclear Data Section, INDC(CCP)-434, September, 2002, <http://www-nds.iaea.org/reports/indc-ccp-434.pdf>; Titarenko Yu.E. et al. *Phys. Rev. C* **65**, 064610 (2002); *Nucl. Instr. Meth. A* **414**, 73-99 (1998).
3. Gloris M. et al., *Nucl. Instr. Meth.*, **A463**, 593 (2001); EXFOR file O0500.
4. Enqvist T. et al., *Nucl.Phys. A* **686**, 481 (2001); *Nucl. Phys. A* **703**, 435(2002) (0.5 GeV – priv. comm.).
5. Prael R.E., Lichtenstein H., User Guide to LCS: The LAHET Code System. LA-UR-89-3014, Los Alamos (1989).
6. Mashnik S.G. et al., Improved Intranuclear Cascade Models for the Codes CEM2k and LAQGSM, *Proc. ND2004*, *in press*.
7. Mashnik S. G., Gudima K. K., Sierk A. J., Merging the CEM2k and LAQGSM Codes with GEM2 to Describe Fission and Light-Fragment Production. Proc. SATIF-6, OECD, Paris, pp. 337-366 (2004); Eprint: nucl-th/0304012. April 10-12, 2002, Stanford Linear Accelerator Center, CA 94025, USA.
8. Boudard A. et al., *Phys. Rev. C* **66**, 044615 (2002).
9. Gaimard J.-J., Schmidt K.-H. *Nucl. Phys. A* **531**, 709 (1991); Jungans A. R. et al. *Nucl. Phys. A* **629**, 635 (1998).
10. Barashenkov V.S., *Comp. Phys. Comm.* **126**, 28-31 (2000).
11. Ignatyuk A. V. et al., Analysis of Spallation and Fission Fragment Residues for Lead Isotopes Irradiated by Protons at 0.15, 1.0 and 2.6 GeV, Contribution #456 in *Proc. ND2004*, *in press*.