

LA-UR-DO - 3599

Approved for public release;  
distribution is unlimited.

*Title:* THRESHOLD REACTION RATES INSIDE AND ON THE SURFACE OF THICK W-NA TARGET IRRADIATED WITH 0.8 GeV PROTONS

*Author(s):* Yury E. Tiarenko, Oleg Shvedov, Vyacheslav F. Batyaev, Evgeny I. Karpikhin, Valery M. Zhivun, Aleksander B. Koldobsky, Ruslan D. Mulambetov, Dmitry V. Fischenko, Svetlana V. Kvasova  
Audrey M. Voloschenko, Stepan G. Mashnik,  
Richard E. Prael

*Submitted to:* SHIELDING ASPECTS OF ACCELERATORS, TARGETS AND IRRADIATION FACILITIES (SATIF-5)  
17-21 July, 2000

## Los Alamos NATIONAL LABORATORY

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.

## **DISCLAIMER**

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, make any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

## **DISCLAIMER**

**Portions of this document may be illegible  
in electronic image products. Images are  
produced from the best available original  
document.**

RECEIVED

DEC 18 2000

OSTI

**THRESHOLD REACTION RATES INSIDE AND ON THE SURFACE OF THICK W-NA  
TARGET IRRADIATED WITH 0.8 GeV PROTONS**

**Yury E. Titarenko, Oleg V. Shvedov, Vyacheslav F. Batyaev,  
Evgeny I. Karpikhin, Valery M. Zhivun, Aleksander B. Koldobsky,  
Ruslan D. Mulambetov, Dmitry V. Fishchenko, Svetlana V. Kvasova**

Institute of Theoretical and Experimental Physics, B. Cheremushkinskaya 25, 117259 Moscow, Russia

**Andrey .M. Voloschenko**  
Keldysh Institute of Applied Mathematics, Miusskaya Sq. 4, 125047 Moscow, Russia

**Stepan G. Mashnik, Richard E. Prael**  
Los Alamos National Laboratory, Los Alamos, NM 87545, USA

**Abstract**

The preliminary results are presented of the experimental determination of threshold reaction rates in experimental samples made of Al, Co, Bi, In, Au et al. placed both inside and on the surface of extended thick W-Na target irradiated with 0.8 GeV protons. The target consists of 26 alternating discs each 150 mm in diameter: 6 tungsten discs are 20 mm thick, 7 tungsten discs are 40 mm thick, and 13 sodium discs are 40 mm thick. The relative position of discs is matched with the aim of flattening the neutron field along the target surface. The comparison is made of the measured rates with results of their simulation using LAHET and KASKAD-S codes, and ENDF/B6, MENDL2, MENDL2P, SADKO-2, and ABBN-93 databases. The results are of interest both in terms of integral data collection and demonstration the up-to-the-date predictive power of codes applied in designing hybrid (ADS) systems that use tungsten targets cooled with sodium [1, 2].

**Foreword**

The quantitative information describing interaction of accelerated proton with a target is necessary in designing even demo ADS. Application of hadron-nuclei process simulation should be tested by special experiments in which irradiation conditions, target material composition and location approximate the design type to the limit. There are several projects, [1] for example, where tungsten cooled by sodium is considered to be used as target material. This served as the basis for conducting experiment with a micromodel of such target. Comparison of experimental data obtained on such micromodel with corresponding calculated values will give us valuable information both for modification of codes and databases and for assessment of calculation accuracy of the target part of the relevant ADS facility designs.

## Experiment plan

The target with alternating adherent tungsten and sodium discs was designed as such micromodel. The location of these discs which was specially chosen facilitates the maximum flattening of the neutron field along the target. Experimental samples made of Al, Co, In, Au, Bi,  $^{63}\text{Cu}$ ,  $^{65}\text{Cu}$ ,  $^{93}\text{Nb}$ ,  $^{64}\text{Zn}$ ,  $^{19}\text{F}$ ,  $^{12}\text{C}$ , Ta, Tm which were manufactured by punching the corresponding foils or by molding fine powders and represent experimental samples 10.5 mm in diameter and 0.1-0.3 mm thick were placed inside the target and on its wall. The layout of tungsten and sodium discs is shown in Fig. 1. All discs are 150 mm in diameter, disc thickness and sequence is listed in Table 1. The tungsten discs have special design providing insertion of special bars with round recesses for experimental samples to be placed inside the target. The discs are located on a special adjustment table which provides alignment of target and proton beam axes with accuracy in the order of 1 mm. The proton beam size examination has been performed by using in tentative exposures to radiation aluminum cutted foils and Polaroid film.

Table 1. Disc sequence, thickness, and experimental samples layout.

Disc number	Material	Thickness, mm	Samples	
			Inside	Outside
W1	W	20	Al, Co	Al, In, Bi, Au, Ta
Na1	Na	3*40=120	-	-
W2	W	20	Al, Co	Al, In, Bi, Au, $^{169}\text{Tm}$ , $^{63}\text{Cu}$ , Ta, $^{65}\text{Cu}$ , $^{93}\text{Nb}$ , $^{64}\text{Zn}$ , $^{19}\text{F}$ , $^{12}\text{C}$ , Co
Na2	Na	2*40=80	-	-
W3	W	20	Al, Co	Al, In, Bi, Au, Ta
Na3	Na	2*40=80	-	-
W4	W	20	Al, Co	Al, In, Bi, Au, Ta
Na4	Na	40	-	-
W5	W	40	Al, Co	Al, In, Bi, Au, Ta
Na5	Na	40	-	-
W6	W	40	Al, Co	Al, In, Bi, Au, Ta
Na6	Na	40	-	-
W7	W	40	Al, Co	Al, In, Bi, Au, Ta
Na7	Na	40	-	-
W8	W	40	Al, Co	Al, In, Bi, Au, Ta
Na8	Na	40	-	-
W9	W	40+20=60	Al, Co	Al, In, Bi, Au, Ta
Na9	Na	40	-	-
W10	W	2*40=80	Al, Co	Al, In, Bi, Au, Ta
Total		W: 280	50 Al,	10 Al, 10 In, 10 Bi, 10 Au,
		Na: 520	10 Co	$^{169}\text{Tm}$ , $^{63}\text{Cu}$ , $^{65}\text{Cu}$ , $^{93}\text{Nb}$ , $^{64}\text{Zn}$ , $^{19}\text{F}$ , $^{12}\text{C}$ , Co
		900		123 samles <sup>1</sup>

W (97.5%), Ni (1.75%), Fe (0.75%), and less than 0.2 % of impurities are incorporated in tungsten discs. The average density of the tungsten discs is  $18.6\text{g/cm}^3$ . Sodium discs represent metallic so-

<sup>1</sup> There are samples (made of Al) beside those specified in Table 1 for measuring the proton beam density distribution across the front(al) surface and control of the neutron field uniformity along discs that form the target.

dium placed into a steel container with 0.4 mm thick walls. Impurities content in Na is less than 0.02%.

Target irradiation was performed with 0.8 GeV protons over a period of 10 hrs at the average intensity equal to  $4.8 \times 10^{10} \text{ p/cm}^2 \text{ pulse}$  using ITEP synchrotron. Pulse repetition rate is 15 pulses per minute. Change in the proton beam intensity over irradiation period is presented in Fig. 2. After short decay lag experimental samples were extracted from the target's surface and inside volume and packaged into labeled polyethylene packages. Subsequent gamma spectra measurements were taken using several spectrometers. The absolute values for different threshold reaction rates were determined using PCNUDAT decay database after gamma-spectra processing with GENIE2000 code (see Table 2).

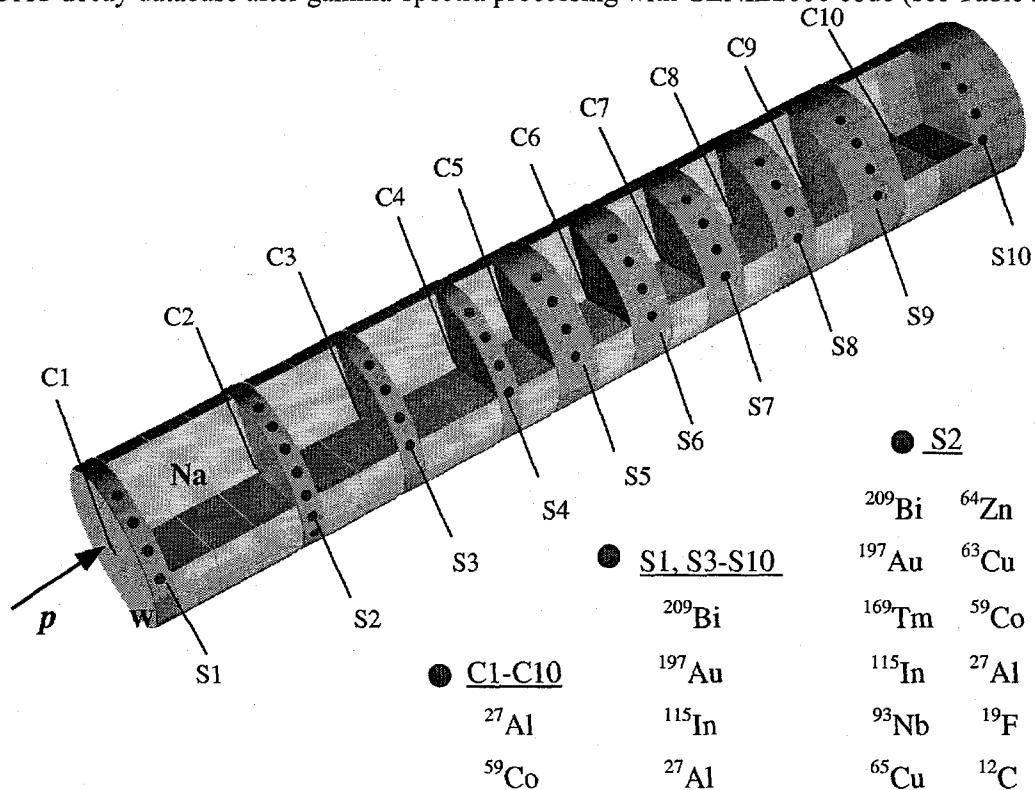


Fig. 1. Layout of W and Na discs and experimental samples.

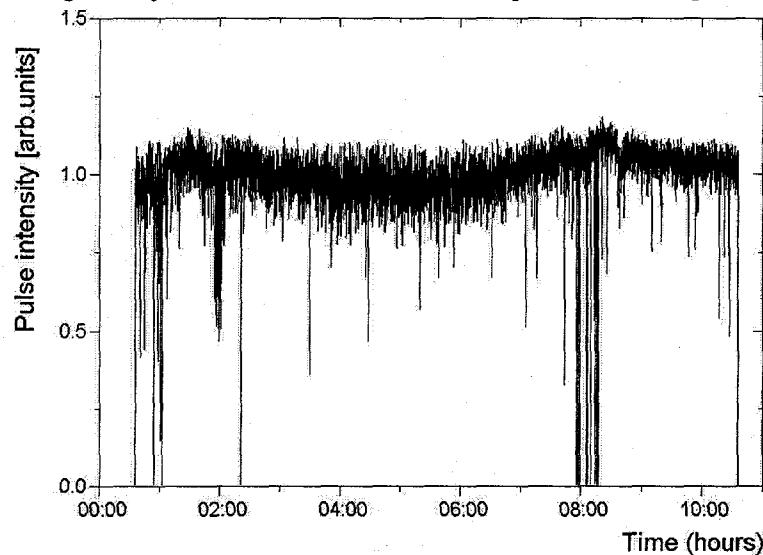


Fig. 2. Changes in the proton beam intensity over the period of irradiation.

Table 2. Thresholds and main parameters of products for reaction rates being measured.

Reaction	$E_{th}(\text{MeV})$	Product	$T_{1/2}$	$E_r(\text{keV}) (Y_r(\%))$
$^{115}\text{In}(n,n')$	0.335	$^{115m}\text{In}$	4.49h	336.2(45.8)
$^{27}\text{Al}(n,p)$	1.89	$^{27}\text{Mg}$	9.46m	843.7(71.8)
$^{64}\text{Zn}(n,p)$	2.1	$^{64}\text{Cu}$	12.70h	511.00(35.8)
$^{59}\text{Co}(n,p)$	2.6	$^{59}\text{Fe}$	44.50d	1099.3(56.5)
$^{27}\text{Al}(n,a)$	3.26	$^{24}\text{Na}$	15.02h	1368.5(100)
$^{197}\text{Au}(n,2n)$	8.5	$^{196}\text{Au}$	6.18d	355.7(86.9)
$^{115}\text{In}(n,p)$	9.0	$^{115}\text{Cd}$	2.23d	527.9(27.5)
$^{115}\text{In}(n,2n)$	9.0	$^{114}\text{In}$	49.51d	191.6(16)
$^{65}\text{Cu}(n,2n)$	10.1	$^{64}\text{Cu}$	12.70h	511.00(35.8)
$^{63}\text{Cu}(n,3n)$	~15	$^{61}\text{Cu}$	3.408h	656.0(10.7)
$^{59}\text{Co}(n,2n)$	11	$^{58}\text{Co}$	70.92d	810.8(99.4)
$^{19}\text{F}(n,2n)$	11.1	$^{18}\text{F}$	1.83h	511(194)
$^{64}\text{Zn}(n,2n)$	12.4	$^{63}\text{Zn}$	38.1m	669.6(8.40)
$^{12}\text{C}(n,2n)$	~20	$^{11}\text{C}$	20.38m	511(162)
$^{209}\text{Bi}(n,4n)$	22.6	$^{206}\text{Bi}$	6.24d	803.1(98.9)
$^{197}\text{Au}(n,4n)$	23	$^{194}\text{Au}$	39.5m	328.4(63)
$^{169}\text{Tm}(n,4n)$	25.5	$^{166}\text{Tm}$	7.7h	778.82(19.9)
$^{209}\text{Bi}(n,5n)$	29.6	$^{205}\text{Bi}$	15.31d	1764.3(32.5)
$^{93}\text{Nb}(n,4n)$	31	$^{90}\text{Nb}$	14.6h	1129.2(92.7)
$^{115}\text{In}(n,5n)$	35	$^{111}\text{In}$	2.83d	245.4(94)
$^{209}\text{Bi}(n,6n)$	38	$^{204}\text{Bi}$	11.22h	899.2(98.5)
$^{209}\text{Bi}(n,7n)$	45.3	$^{203}\text{Bi}$	11.76h	820.2(29.6)

### Simulation of reaction rates

To simulate reaction rates measured the LAHET Code System (LCS) [3] was used which involves LAHET Code for simulating hadron-nuclei interactions, HMCNP code for simulating neutron transport at energies below 20 MeV, and PHT code for simulating hard photon transport. As the result of this both neutron and proton spectra were calculated in all points where experimental samples are located.

Another code system applied for simulation the experiment is the KASKAD-S code [4] which uses a discrete ordinate algorithm for coupled charges/neutral particle transport calculations in 2D pencil beam problems. The multigroup cross-section library SADKO-2 for nucleon-meson cascade calculations coupled with CONSYST/ABBN-93 neutron and gamma-ray cross-section libraries below 20MeV is used.

Reaction cross sections for neutrons with energies up to 100 MeV and protons with energies up to 200 MeV were taken from MENDL2 [5] and MENDL2P [6] libraries respectively. Reaction cross section are obtained via integral product of spectra and cross section ( $R = \int \sigma(E) \phi(E) dE$ ).

The discrepancy between calculated and experimental results was estimated with the use of  $\langle F \rangle$ , the root-mean-square discrepancy factor which is defined in [7].

## Results

The preliminary processing of gamma spectra from Al samples placed on the target outside surface and in the target center makes it possible to estimate generation rates of  $^{24}\text{Na}$  and  $^{27}\text{Mg}$ . Experimental and calculated values obtained using LAHET code are presented in Fig. 3. Measurements of gamma-spectra of all the samples irradiated are still being in progress and all the reaction rates listed in Table 2 are expected to be determined after complete gamma-spectra processing.

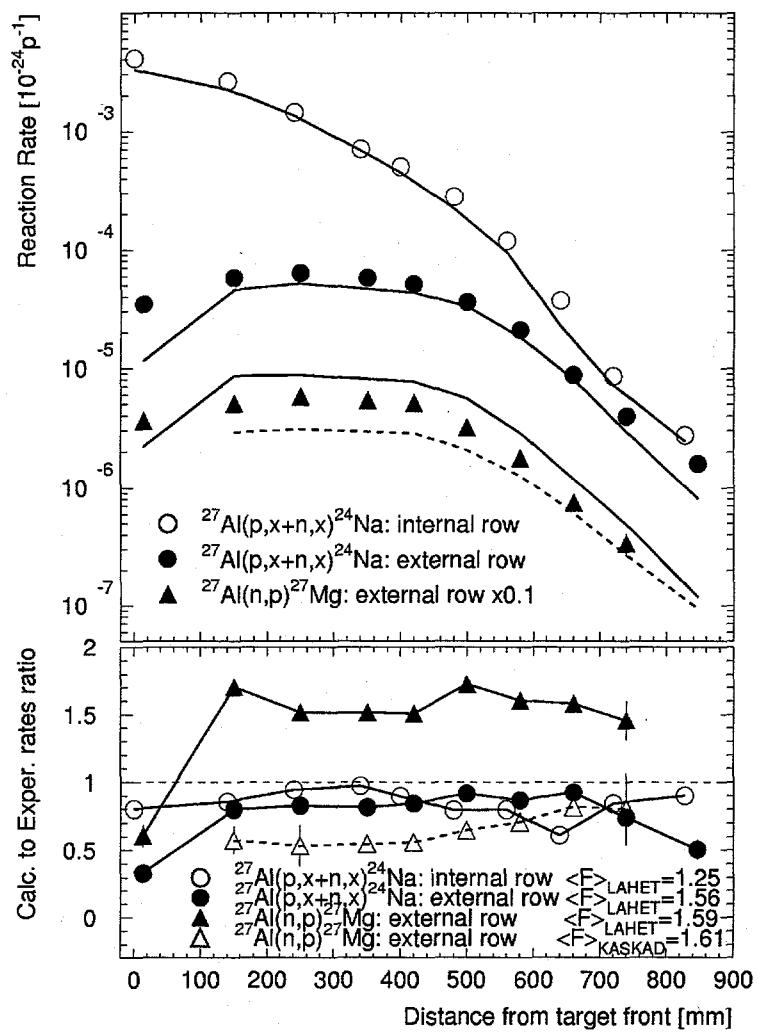


Fig. 3. Reaction rates of  $^{27}\text{Al(p,x+n,x)}^{24}\text{Na}$  and  $^{27}\text{Al(n,p)}^{27}\text{Mg}$  at target axis and on surface. Calculation using the LAHET and the KASKAD-S codes are identified by solid lines, and dashed lines, respectively. The ratio of reaction rate calculated values to experimental are shown in the bottom figure.

### Conclusion on convergence of calculated and experimental values.

The rate of  $^{24}\text{Na}$  in Al foils predicted using the LAHET code is in satisfactory agreement with experimental values (except the first and the last point on the surface) as is indicated in Fig. 3. Along with this, the predicted rates of  $^{27}\text{Mg}$  generation on the target surface are systematically (1.6 on the average) overestimated except the first point compared to reaction rates measured. At the same time,

$^{27}\text{Mg}$  production rates predicted using the KASKAD-S code are underestimated, on the average, by a factor of 1.6. The study of causes of detected discrepancies could be performed only after complete processing of gamma spectra of the rest of samples and determining both (n,p) – purely “neutron” - reactions and (p,n) – purely “proton” - reactions.

### Acknowledgement

The work was done with support of Japan Atomic Energy Research Institute within the framework of the ISTC project #1145 and was partially supported by the U. S. Department of Energy.

### References

- [1] T. Takizuka et al, Conceptual Design Study of Accelerator-Driven Systems for Nuclear Waste Transmutation, *Proc. of the Second International Conference on Accelerator Driven Transmutation Technologies and Applications*, June 3-7, 1996, Kalmar, Sweeden, pp. 179-185.
- [2] G.J. Van Tuyle, D.E. Beller, Accelerator Transmutation of waste technology and implementation? scenarios, *Proc. of the Third Int. Topical meeting on Nuclear Applications of Accelerator Technology*. Long Beach, CA, November 14-18, 1999, 337-346.
- [3] R.E. Prael, H.Lichtenstein, LANL Report LA-UR-89-3014 (1989).
- [4] M. Voloschenko, “An  $S_n$  Algorithm for Spallation Target Neutronics and Shielding Calculations,” *Proc. of International Conference on Mathematics and Computations, Reactor Physics, and Environmental Analyses in Nuclear Applications*, 27-30 September, 1999, Madrid, Spain, vol. 2, p. 975; A. M. Voloschenko, E. I. Yefimov, T. T. Ivanova, V. P. Kryuchkov and O. V. Sumaneev, “Using Discrete Ordinate Method in the SpallationTarget Neutronics and Shielding Calculations,” *Proc. 3rd International Conference on Accelerator Driven Transmutation Technologies and Applications*, June 7-11, 1999, Pruhonice, Chech Republic, Paper Code We-O-E12
- [5] Yu.N. Shubin et al., Cross Section Data Library MENDL-2 to Study Activation and Transmutation of Materials Irradiated by Nucleons of Intermediate Energies, IAEA, INDC(CCP)-385, Vienna, May 1995.
- [6] Yu.N. Shubin et al., Cross Section Data Library MENDL-2p to Study Activation and Transmutation of Materials Irradiated by Nucleons of Intermediate Energies, Nuclear Data for Science and Technology (Trietse 1997), IPS, Bologna, 1997, v.1., p.1054.
- [7] Yu.E. Titarenko et al. "Exerimental and Theoretical Study of the Yields of Radionuclides Produced in  $^{209}\text{Bi}$  thin target Irradiated by 1500 MeV and 130MeV Protons", Nucl.Instr. and Meth. A414 (1998) 73; Yu.E. Titarenko et al. "Yield study of residual nuclei of products generated in GeV proton interaction with thin  $^{208}\text{Pb}$  и  $^{nat}\text{W}$  targets". *Proceedings of SATIF-5 meeting*.