Measurements of Delayed Neutrons Yields and Time Spectra from 1 GeV protons interacting with thick ^{nat}Pb targets

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Abstract

This paper presents the preliminary results on measured delayed neutron (DN) yields and time spectra from 1 GeV protons interacting with thick ^{nat}Pb targets. In parallel, the MCNPX and PHITS codes were used to predict the DN precursors and construct the theoretical DN tables. Different model parameters are examined and show significant dependence on the choice of the intra-nuclear cascade and fission-evaporation models used. These data and modeling are of great importance for the new generation spallation neutron sources based on liquid metal technologies where a significant amount of the DN precursor activity can be accumulated in the target fluid.

KEYWORDS: delayed neutrons, high energy fission-spallation reactions, intensive neutron sources, liquid metal targets

1. Introduction

The next generation spallation neutron sources, neutrino factories or RIB production facilities currently being designed and constructed around the world will increase the average proton beam power on target by a few orders of magnitude. Increased proton beam power results in target thermal hydraulic issues leading to new target designs, very often based on flowing liquid metal targets such as Hg, Pb, Pb-Bi. Radioactive nuclides produced in liquid metal targets are transported into hot cells, into pumps with radiation sensitive components and electronics, etc. Besides the considerable amount of photon activity in the irradiated liquid metal, a significant amount of the DN precursor activity can be accumulated in the target fluid. The transit time from the front of a liquid metal target into areas, where DNs may be important, can be as short as a few seconds, well within one half-life of many DN precursors. Therefore, it is necessary to evaluate the total neutron flux (including DNs) as a function of time and determine if DNs may contribute significantly to the activation and dose rate.

In the earlier work [1] the DN flux and corresponding time spectra were estimated in the case of the MegaPie spallation target at PSI (Switzerland). The DN tables within 6-group model were constructed for the first time for high energy fission-spallation reactions. In the same Ref. [1] it was demonstrated that the final estimates of DNs were very much

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model-dependent the physics models used within the MCNPX code [2]. To our knowledge, no experimental data were available for DN yields from high energy fission-spallation reactions on Pb targets. Therefore, the experiment to perform these measurements was realized in December 2005.

In this work we present some preliminary analysis on measured DN yields and time spectra from 1 GeV protons interacting with thick ^{nat}Pb targets. In addition, a number of model calculations with MCNPX [2] and PHITS [3] codes were performed as long as predictions of DNs from high energy fission-spallation reactions are concerned. Comparison between the experimental data and code results are discussed.

2. Experimental setup

A schematic view of the experiment carried out at PNPI Gatchina (Russia) is shown in Fig. 1. A 1 GeV proton beam impinged on the natural Pb target of different thicknesses producing a huge number of spallation-fission products (including DN emitters). After the beam was switched off, DNs were detected with optimized He-3 detector following specific irradiation periods. Long, intermediate and short irradiation cycles were used to optimize the extraction of different time parameters of DN groups.

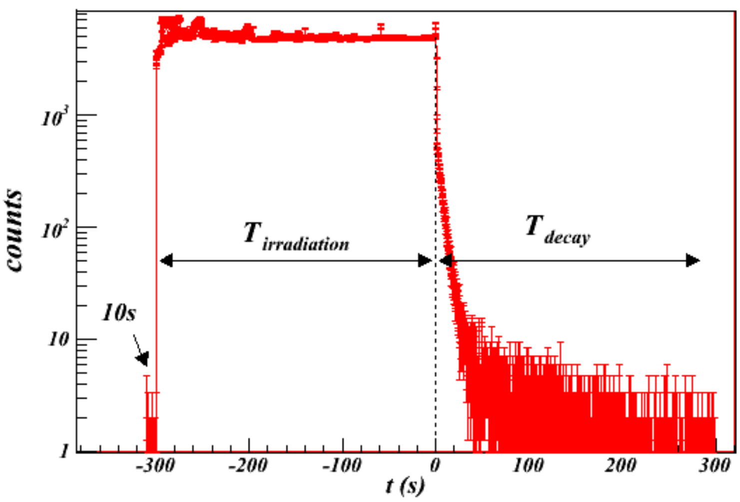
Figure 1: A photo of the experimental setup to measure DNs from p(1GeV)+Pb with the proton beam line (on the right), the Pb target (in the center) and He-3 counter (on the left).

The He-3 tube was surrounded by cylindrical polyethylene (CH_2) moderator in order to increase the neutron detection efficiency. Our Monte Carlo simulations showed that an optimal CH_2 thickness is around 5 cm for neutrons in the energy range of 100 keV – 1 MeV, i.e. in the DN energy range. To avoid the background due to the thermal neutrons, the polyethylene moderator was coated by 1 mm ^{nat}Cd foils (see Fig 1.). In addition, the detector was placed in the shielding "house" with 8-16 cm thick walls made of borated (3%) polyethylene in order to protect the detection system from fast and low-energy room neutrons. The detector was calibrated with a standard ^{252}Cf neutron source, and its efficiency was confirmed within 15 % by Monte Carlo calculations performed with MCNPX [2]. The proton beam intensity was monitored by activation of ^{27}Al foils and the γ-spectroscopy off line. The gamma lines of ^{7}Be, ^{22}Na and ^{24}Na were identified and analyzed for this purpose.

3. Experimental procedure

Fig. 2 explains the experimental procedure used in this work, where the case of a long irradiation and long decay cycle (300 s – 300 s) is shown. It is clearly seen that the He-3 counter is saturated during the irradiation, and comes back to its normal operation right after (typically within 15-20 ms) the beam is switched off. Each individual irradiation-decay cycle was recorded on line and summed off line afterwards to accumulate the statistics.

Figure 2: The measurement procedure of DN time spectra (decay curves). T_{irrad} and T_{decay} regions are shown explicitly.



Before the final experiment some tests were performed in order to characterize the background in the experimental hall. These tests were done using 1 GeV incident proton beam in two configurations: A) without any target, representing active background, and B) with the Pb target of 20 cm thick, representing active background and physical signal.

After irradiation without any target the detector clearly measured some DNs (beam off) with the decay period of $T_{1/2}$ ~ 4 s (see Fig. 3 – on the left). After trying different collimation systems, different detector positions, etc., the corresponding decay time of the background neutrons remained unchanged. We concluded that this background was due to ^{17}N (β-n with $T_{1/2}$ ~ 4.173 s), created mainly from ^{18}O via different reactions (e.g. (γ,p), (n,d), (p,2p), etc. typically of the order of 10 mb). Note that spallation reactions from 1 GeV protons on Al, Cu, Fe and other materials present in the experimental hall or accelerator structures also lead to the creation of ^{17}N (e.g. σ(p(1GeV)+Cu ^{17}N) = 0.149 mb or σ(p(1GeV)+Al ^{17}N) = 0.66 mb [4]). By placing 20 cm thick Pb target in the beam, the $T_{1/2}$ ~ 4 s contribution remained, but DNs with higher half-lives became also visible (see Fig. 3 - on the right). This proved the feasibility of the final experimental campaign.

We also checked the above background hypothesis experimentally by irradiating thick iron cylinder and massive brick being representative materials for the accelerator structures and experimental hall. The results are presented in Fig. 4. Indeed, the DN curves can be reproduced with only two half-lives in both cases, namely $T_{1/2}$ ~ 4 s and $T_{1/2}$ ~ 0.2 s. We confirm that in high energy protons interacting with intermediate mass nuclei the reaction products like ^{17}N ($T_{1/2}$ = 4.173 s), ^{9}Li ($T_{1/2}$ = 0.178 s) and eventually ^{16}C ($T_{1/2}$ = 0.747 s) will be the major delayed neutron precursors [4]. It is important to note that no longer half lives were observed during these test irradiations.

The final measurements were done with 1 GeV protons interacting with the natural Pb targets of 10 cm diameter and different thicknesses (5, 10, 20, 40 and 55 cm). Statistics was accumulated in periodic fashion for three different irradiation-decay cycles (300 s - 300 s; 20 s – 120 s; 1 pulse of ~700 µs – 2 s). For all different configurations, new Al foils were irradiated each time for proton beam monitoring. Background measurements (no target) were equally performed.

Figure 3: Measured DN time spectra (decay curves) without any target - on the left and with a thick Pb target - on the right. Also see the text for details.

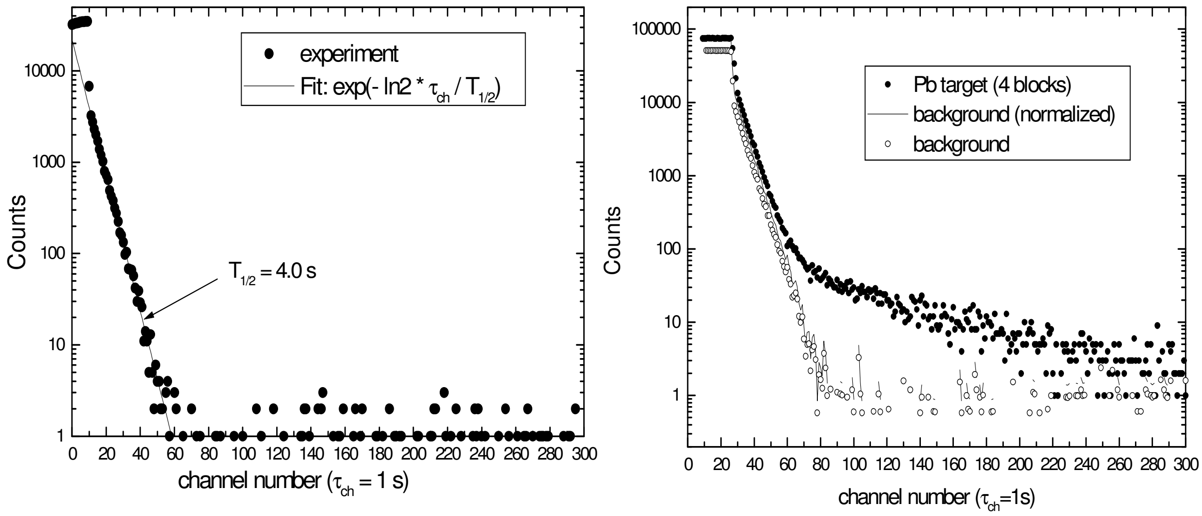
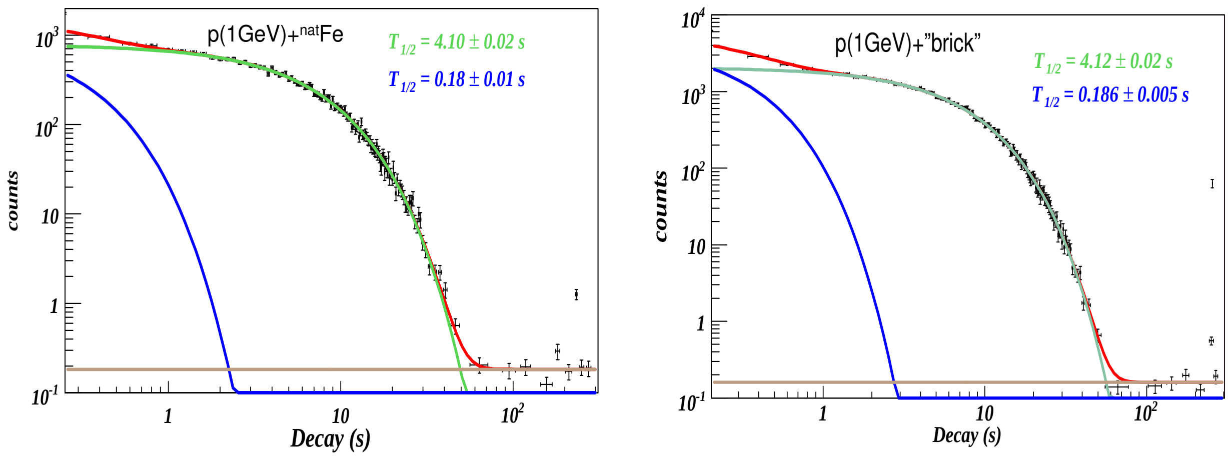


Figure 4: Experimental DN decay curves from p(1GeV) + ^{nat}Fe (on the left) and p(1GeV) + “brick” (on the right) together with corresponding fits with the sum of two exponentials. Note the log-log scale.



4. Preliminary results

So far only very preliminary analysis of the DN decay curves was performed for all target thicknesses and with irradiation-decay cycles of 300 s – 300 s. The accumulated data are presented all together with corresponding fits of exponential sums in Fig. 5. The following expression was used to obtain the $\{a_i; \lambda_i\}$ values:

$$DN(t) = \sum_i a_i \exp(-\lambda_i t)(1-\exp(-\lambda_i T_{irrad})) + C,$$

with $\lambda_i = ln\ 2 / T^i_{1/2}$, T_{irrad} – irradiation time and C - the free constant background. Contrary to the conventional 6-group approach to reproduce the DN decay curves resulting from neutron induced fission on actinides, we found that from 4 to 5 terms of the above expression might be sufficient.

In addition, to make the fitting procedure more accurate, we analyzed in detail the earlier experimental work on isotopic yields of reaction products (including the DN precursors in particular) from 1 GeV proton induced reactions on Pb or neighboring targets [4, 5]. Indeed, this preparatory work permitted to restrain a number of half lives. For example, in the case of group 2 DN precursors as ^{136}Te ($T_{1/2}$ = 17.5 s), ^{137}I ($T_{1/2}$ = 24.5 s), and ^{141}Cs ($T_{1/2}$ = 24.9 s) with comparable half lives can be neglected due to their very small production cross sections (2 orders of magnitude lower compared to ^{88}Br). Table 1 summarizes the fixed time parameters in the above *DN(t)* dependence and identified corresponding DN precursors. Note that the constant background C and a_i values were kept free in all cases.

As a matter of fact, the chosen data analysis strategy seems to be justified at least for the half-lives higher than 1 s as seen from Fig. 5. Indeed, the experimental data are reproduced with good precision for all target thicknesses. On the other hand, much more careful analysis should be done in the decay region from 0 to 1-2 s, where the extracted a_4 (^{9}Li) and a_5 (not shown in these figures) values can vary considerably (up to 2 orders of magnitude) as a function of the target thickness.

Table 1: The half-life constraints during the fitting procedure of the DN decay curves. Also see Fig. 5 (at the very bottom) corresponding to the 55 cm thick Pb target for more details.

Group	Half-life, s	Identified precursor	P_n (β-n), %
1	55.60	^{87}Br	2.52
2	16.29	^{88}Br	6.58
3	4.173	^{17}N	95.10
4	0.178	^{9}Li	50.80
5	free, but < 1 s	^{16}C, ^{17}C, ^{14}Be, …	99, 32, 81, …

In our opinion, this observation could be explained either by the existence of the DN precursors with shorter half-lives or by the variable time needed for the He-3 counter to come back to its normal operation after the saturation. Hopefully, the future analysis of the DN curves with short irradiation periods, where much narrower time channels were used, will allow us to disentangle this problem.

5. Discussion

Although we still cannot provide the DN yields in absolute value (the data analysis of the irradiated Al foils for beam monitoring is still in progress) some interesting observations already can be outlined.

First of all, the preliminary data analysis clearly shows that from 1 GeV protons interacting with thick Pb targets the major DN contribution is from light mass products as ^{9}Li and ^{17}N rather than from fission products as in the case of actinides. In addition, this “unusual” DN emission dominates the DN decay curve up to the period of 10-20 s (see Fig. 5). For longer decay time, say 50-100 s, the long lived DN precursors, being “usual” fission products as ^{88}Br and ^{87}Br, remain the only contributors to the DN activity. Note that this finding is very important in the case of high power liquid metal targets, where the liquid metal makes the "round trip" typically in the period of 10-30 s (e.g., the MegaPie loop makes a turn in ~20 s [1]).

Figure 5: The DN decay curves from p(1GeV)+Pb as a function of the target thickness.

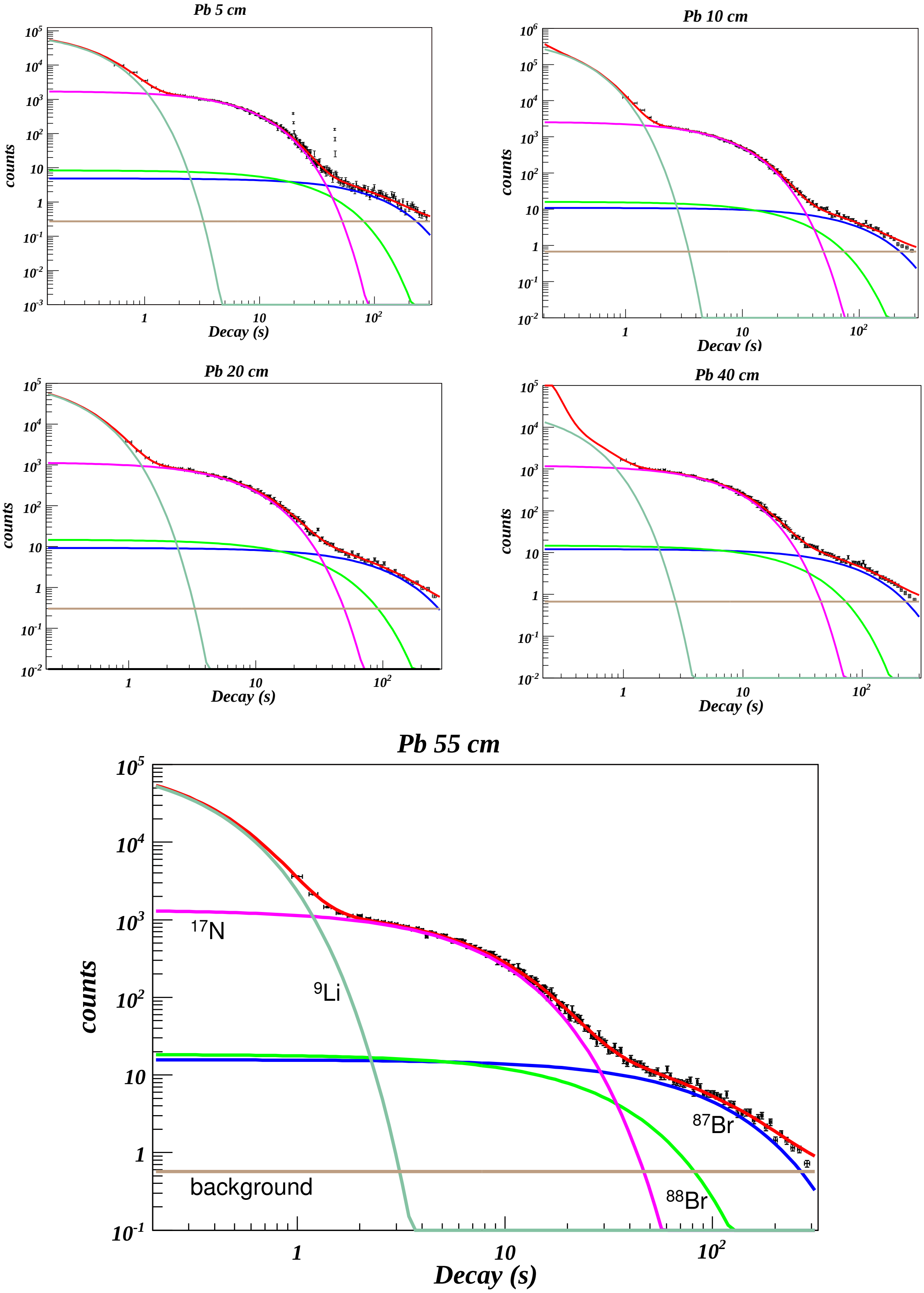


Table 2: Relative values of the experimental a_i ratios with respect to the $a_1(^{87}Br) = Y_{cum} \cdot P_n(^{87}Br)$ as a function the target thickness. Also see Fig. 5.

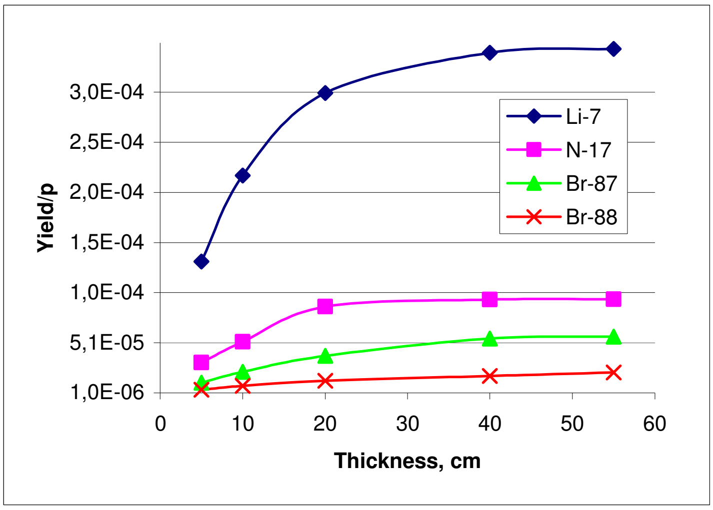
Target thickness, cm	$a_2(^{88}Br)/a_1(^{87}Br)$	$a_3(^{17}N)/a_1(^{87}Br)$
5	1.7	354
10	1.4	235
20	1.3	118
40	1.3	102
55	1.4	90

Secondly, some interesting dependence of the relative a_i ratios, being the product of the cumulative yield Y_{cum} and DN emission probability P_n specific for each DN precursor, was noticed as a function of the target thickness. The experimental results are presented in Table 2, where it is clearly seen that one of these ratios, namely a_2/a_1 is nearly constant, while a_3/a_1 is constantly decreasing with increasing target thickness. This can be explained in terms of two different reaction mechanisms responsible for the production of these DN precursors, namely fission (Br) and fragmentation-evaporation (N and Li) respectively. In addition, the decrease of the ratio a_3/a_1 stabilizes with the target thickness of 40-55 cm. What means that the production cross sections of DN precursors by the above reaction mechanisms are much more important at higher energies, i.e. the most of the reactions take place in the 1st half of the stopping target. These findings are confirmed by the model predictions obtained using the PHITS code [3] with the results given in Table 3 and Fig. 6.

Table 3: Relative values of the PHITS predicted a_i ratios with respect to the $a_1(^{87}Br) = Y_{cum} \cdot P_n(^{87}Br)$ as a function the target thickness.

Target thickness, cm	$a_2(^{88}Br)/a_1(^{87}Br)$	$a_3(^{17}N)/a_1(^{87}Br)$	$a_4(^{9}Li)/a_1(^{87}Br)$
5	0.96	104	235
10	0.95	89	200
20	0.89	86	159
40	0.86	64	125
55	0.97	62	121

Figure 6: Production yields of Li, N and Br isotopes (predicted by PHITS) from the reaction p(1GeV)+Pb as a function of target thickness. The yields are normalized per incident proton.

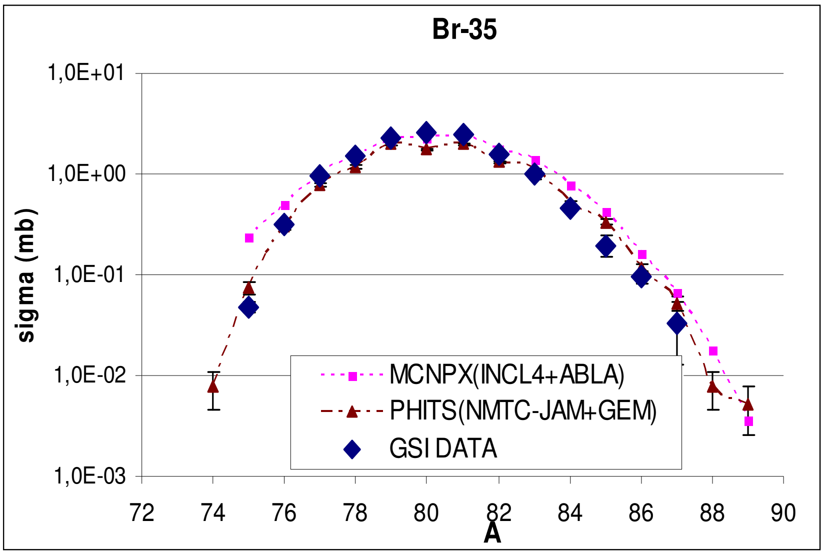


Finally, note that before performing the full scale transport calculations, we benchmarked the PHITS and MCNPX codes against the 1 GeV proton data from thin targets [4, 5]. The PHITS results (NMTC-JAM+GEM) [3] together with the best model combination within the MCNPX code (INCL4-ABLA) [2] are presented in Table 4 and Fig. 7. This brief comparison proves that PHITS can predict within a factor of 2 not only heavy but also light DN precursors from high-energy fission spallation reactions, while MCNPX gives zero values for the production of ^{9}Li and ^{17}N being of the major importance in our case. It will be very interesting to compare the PHITS code predictions with our experimental DN decay curves in absolute values when the data analysis is finalized.

Table 4: Production cross sections of some of the DN precursors from 1 GeV protons interacting with ^{208}Pb. Experimental data marked by "*" are extrapolated values.

σ (mb)	MCNPX	PHITS	Experiment
^{9}Li	0	0.981(39)	~1.0* [4]
^{17}N	0	0.249(17)	~0.6* [4]
^{87}Br	0.065	0.052(8)	0.033(20) [5]
^{88}Br	0.018	0.008 (3)	~0.01* [5]

Figure 7: Comparison of experimental [5] and predicted production cross sections of the Br isotopes from 1 GeV protons interacting with ^{208}Pb. Different model combinations are indicated in the legend.



6. Conclusions

In this work we present for the 1st time the experimental data on measured DN yields and time spectra from 1 GeV protons interacting with thick ^{nat}Pb targets. These results are of great importance for the new generation spallation neutron sources based on liquid metal technologies where a significant amount of the DN precursor activity can be accumulated in the target fluid.

Although the data analysis is still in progress, a number of important conclusions can be drawn already at this stage:

- The emission of DNs from p(1GeV)+^{nat}Pb is dominated by light reaction products as ^{9}Li and ^{17}N during the decay time from 0 to ~20-30 s, while after the decay time >30 s the fission fragments as ^{88}Br and ^{87}Br are the major contributors.

- The DN yield production per incident proton is increasing with the target thickness: the majority of the DNs from 1 GeV protons are produced in the 1st half of the stopping lead target.
- The above experimental observations are confirmed with the PHITS transport code, which is able to predict the production of the DN precursors within a factor of 2 or better.

We have also performed a similar experiment using the same 1 GeV proton beam and high purity ^{209}Bi targets of variable thicknesses at PNPI Gatchina (Russia). These new experimental data together with the final results of the present work will be reported elsewhere. Finally, we note that we plan a new experiment to measure the DN emission from liquid Hg target bombarded by ~1 GeV protons.

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