Measurements of delayed neutrons yields and time spectra from 1 GeV protons interacting with thick natPb, 209Bi and natFe targets

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Abstract. This paper presents the experimental results on the DN yields and time spectra from 1 GeV protons interacting with the natPb, 209Bi and natFe targets of variable thicknesses from 5 cm to 55 cm. Both absolute yields and time constants were obtained. 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 modelling are of great importance for the new generation spallation neutron sources based on liquid metal technologies. Finally, the above experiment allowed the determination of the production cross sections of a number of DN precursors as, ^{87}Br, ^{88}Br and ^{17}N. These results permit the examination of two different reaction mechanisms, namely fission and fragmentation, implemented in high energy transport codes.

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 requires the use of liquid metal targets such as Hg, Pb, or Pb-Bi. Radioactive nuclides produced in such targets are transported into hot cells, into pumps with radiation sensitive components and electronics, etc. Due to a short transit time a significant amount of the delayed neutron (DN) precursor activity can be accumulated in the target fluid contributing significantly to the activation and dose rate. This phenomenon was discussed in detail in our earlier work [1] including the estimation of the DN flux and corresponding time spectra in the case of the MegaPie spallation target at PSI (Switzerland) [2]. In ref. [1] we also demonstrated that the final estimates of DNs were very much model-dependent within the MCNPX code [3].

The goal of this work was to measure the DN yields from high energy fission-spallation reactions on the Pb, Bi and Fe targets of variable thicknesses. These measurements were realized in December 2005 and April 2006 at PNPI Gatchina (Russia). Here we present our results from the analysis of the DN yields and time spectra from 1 GeV protons interacting with thick natFe, natPb and 209Bi targets. Corresponding calculations with MCNPX [2] and PHITS [4] codes were also performed leading to some recommendations related to the use of these tools. We note that the data obtained with thick and thin natPb targets were already partially reported in [5].

2 Experimental

A schematic view of the experiment is shown in figure 1. 1 GeV protons from accelerator impinged on the natPb and Bi targets of different thicknesses (0.5, 5, 10, 20, 40 and 55 cm). The emitted DNs were detected with the optimized He-3 detector following specific irradiation periods after the beam is switched off. Long (300 s), intermediate (20 s) and short irradiation ($350 \mu s$ – a single pulse) times were used to optimize the extraction of different time parameters of DN groups. Each individual irradiation-decay cycle was recorded on line and summed off line afterwards to accumulate the statistics.

Fig. 1. A photo of the experimental setup with a cylindrical target (in the centre) and the He-3 counter (in the back).

The He-3 counter was surrounded by a 5 cm thick polyethylene (CH_2) and coated by 1 mm natCd foils in order to increase the neutron detection efficiency in terms of neutron moderation and to avoid the background due to the thermal neutrons correspondingly. The detector optimization was

performed using Monte Carlo simulations with MCNPX [2]. Finally, the detector was calibrated with a standard ^{252}Cf neutron source, and its efficiency was reproduced within 9% by MCNPX.

The proton beam intensity was monitored with relative errors around 10% by activation of ^{27}Al foils (see fig. 1) and the γspectroscopy off line from the ^{7}Be, ^{22}Na and ^{24}Na activity. The diameter of the beam spot and its alignment was obtained using the photo-films as presented in figure 1.

Using the same technique, some measurements without target and with iron or brick-concrete targets were also performed to characterize the active background contribution due to the environment of the experimental area. During these test measurements without any target and with non-fissile targets we observed only two dominating decay periods of $T_{1/2} \sim 4.2$ s and ~ 0.18 s, which we attributed to the reaction products ^{17}N and ^{9}Li with $T_{1/2} = 4.173$ s and $T_{1/2} = 0.178$ s, respectively (see fig. 2).

Fig. 2. The accumulated DN decay curve from p (1 GeV) + Fe (55 cm) (on the top) and from p (1 GeV) + "brick"(thick) (on the bottom). The individual contributions from different precursors are indicated separately.

Indeed, these two DN precursors can be produced in the reactions from 1 GeV protons on different targets as Al, Cu, Fe or any other materials present in the experimental hall or accelerator structures [6]. It is important to note that no longer half-lives were observed during these test irradiations as shown in figure 2. Once the irradiations with ^{nat}Pb or Bi targets started, we noticed that the DN decay curves also

extended to the longer half-lives than ~ 4 s [7], which are characteristic for neutron-rich fission products only.

3 Results

Analysis of the DN decay curves for all ^{nat}Pb and Bi target thicknesses and with identical duration for irradiation and decay (300 s) were performed by fitting data with exponential sums using the TMinuit class implemented in the ROOT toolkit [8]. The following expression was used to obtain the $\{a_i, \lambda_i\}$ values:

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

with $T_{1/2}^i = \ln 2/\lambda_i$, C a constant representing the background, and the irradiation time $T_{irr} = 300$ s. Systematic errors due to the estimation of the C values have been taken into account.

Contrary to the conventional 6-group approach to reproduce the DN decay curves from neutron induced fission on actinides [9], only 4 terms of the above expression were sufficient in our case. Half-lives for these 4 terms correspond to the previously identified spallation products, namely ^{9}Li and ^{17}N, in addition to two fission products ^{87}Br and ^{88}Br (with $T_{1/2} = 55.6$ s and $T_{1/2} = 16.29$ s, respectively).

The accumulated data for the ^{209}Bi target (55 cm thick), are presented together with the fits of exponential sums in figure 3. Similar quality fits using the same 4 terms in the exponential sum, i.e., with four half-lives fixed to those of ^{9}Li, ^{17}N, ^{87}Br and ^{88}Br, were obtained for all target thicknesses and both ^{nat}Pb and ^{209}Bi targets [7] (not shown here).

Fig. 3. The accumulated DN decay curve from p(1 GeV)+^{209}Bi (55 cm); individual contributions from different precursors are indicated separately.

4 Discussion

Some conclusions can already be drawn just looking at the fitted DN decay curves for the ^{nat}Pb and ^{209}Bi targets (e.g., for ^{209}Bi in fig. 3). First, the DN contribution from light mass

products as ^{9}Li and ^{17}N is clearly more important than the contribution from fission products. In addition, this "unusual" DN emission dominates the DN decay curve for Pb and Bi targets up to the cooling time of 10–20 s (see figure 3). For longer decay times, 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]).

Thanks to the efficiency determination of the ^{3}He counter and to the proton beam intensity monitoring, it was possible to extract the DN yields in absolute values. In brief, once the DN(t) is parameterised during the fitting procedure (see figure 3 and equation 1) and when t approaches zero, one obtains

$$DN(t \to 0) = \sum_i a_i + C \qquad (2)$$

Here the following expression can be employed to calculate the individual DN yields:

$$P_n^i Y^i = a_i / (\varepsilon_{He-3} I_p \varDelta t_{ch} N_{cycles}),$$

with P_n^i being the DN emission probability, Y^i – the individual DN precursor yield (atoms per incident proton), ε_{He-3}– the total efficiency of the ^{3}He counter (events per emitted source neutron), I_p – proton beam intensity (protons per second), $\varDelta t_{ch}$ – the channel width (seconds), N_{cycles} – a number of accumulated irradiation-decay cycles.

Figure 4 presents the obtained DN precursor production yields Y^i for $i = {}^{17}$N, ^{87}Br and ^{88}Br as a function of target thickness. Note that in addition to the uncertainties in detector efficiency, proton beam intensity monitoring and statistical errors, we took into account systematic errors due to the fitting procedure and background contribution. We add that the extraction of ^{9}Li yields was impossible due to its short half life, which is comparable with the experimental channel width $\varDelta t_{ch} = 200$ ms.

Some comparisons of the data have also been made with PHITS code predictions (NMTC+JAM+GEM) [4] and prove that this code can reproduce rather accurately the formation of DN precursors in high energy spallation-fission reactions (see the same figure 4) - in most of the cases within a factor 2 or better.

Using the data taken with thin targets (5 mm and 5 cm), production cross section of ^{17}N has been extracted from 1 GeV protons interacting with ^{nat}Fe, ^{nat}Pb and ^{209}Bi. Our results are shown in figure 5 together with other data but for different targets. We note that our data is in good agreement with systematic established by Dostrovsky et al. [6].

Equally, the production cross sections of ^{87}Br and ^{88}Br have been obtained from 1 GeV protons interacting with ^{nat}Pb. Our results are shown in figure 6 including some older data from [10]. Again a good agreement with the earlier work is seen. As it was mentioned earlier in the case of thick targets, PHITS [4] gives reasonable results both for ^{17}N and ^{87,88}Br, while MCNPX can be trusted only for ^{87,88}Br (see figure 6). It is worth mentioning that ^{17}N and ^{88}Br cross sections reported here have been measured for the first time for 1 GeV protons both for the ^{nat}Pb and Bi targets.

Fig. 4. The DN precursor yields Y^i (atoms per proton) as a function of target thickness for Pb (top figure) and Bi (bottom figure). The experimental data (filled symbols) are compared with the PHITS predictions (open symbols). The lines are to guide the eye.

Fig. 5. Experimental production cross sections of the ^{17}N yields from 1 GeV protons interacting with different targets A. See the legend for details.

5 Conclusions

In this work we present the experimental data on measured DN yields and time spectra from 1 GeV protons interacting with thick ^{nat}Fe, ^{nat}Pb, and ^{209}Bi targets. For all these targets the emission of DNs is dominated by light reaction products as ^{9}Li and ^{17}N during the decay time from 0 to ~20-30 s. In the case of fissile targets after the longer decay time 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 and the majority of the DNs from 1 GeV

Fig. 6. Comparison of experimental and predicted production cross sections of the Br isotopes from 1 GeV protons interacting with ^{nat}Pb. See the legend for details.

protons are produced in the 1st half of the stopping target. In addition to the DN yields, the microscopic production cross sections of the DN precursors have been extracted for ^{17}N, ^{87}Br and ^{88}Br.

The above experimental observations are reproduced with the PHITS transport code, which is able to predict the production of the DN precursors within a factor of 2 or better. These new data are of great importance for the new generation high power spallation targets based on liquid metal technology as well as for further development of high energy spallation-fission-fragmentation models.

Finally, we add that in the framework of the Neutronic and Nuclear Assessment Task Group of the MegaPie experiment the DN flux at the top of the liquid PbBi target was recently measured. A preliminary comparison between the experimental DN decay curve at MegaPie and its interpretation using the DN parameters extracted from this work is rather good [11].

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