Current Status of Spallation Product Data: Nuclear Engineering View-Point

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Abstract

Proton induced isotopic transformation in spallation targets of accelerator-driven systems (ADS) is becoming a key factor in selecting the reference target material&design options. The present paper gives an outlook of the current status of data on spallation products from the view-point of nuclear engineering stressing the isotopes whose accumulation would significantly effect the target performance.

1. INTRODUCTION

The maturing of the accelerator technology toward practical implementation in the form of accelerator-driven systems gave rise to the specific concern about isotope transformation in neutron producing targets. Associated terminology sometimes looks confusing. The term "spallation" that in nuclear physics is generally reserved for identification of specific nuclear reaction with emission of secondary nucleons, by engineering community is taken in a complex like "spallation neutron source" or "spallation target" to identify the vital design element responsible for neutron generation induced by accelerated ions. Strictly speaking, in this element pure spallation reaction might be accompanied by reactions like, for example, high energy fission and multi-fragmentation having a different mechanism from classical spallation. In addition to that, if the target is large enough, the secondary nucleons emitted through spallation will be moderated and trigger the nuclear reactions through mechanism of compound nucleus, like capture reaction.

Leaving the terminological issue to the field of nuclear physics, by "spallation products" (SP) in this paper we mean those nuclides accumulated in neutron producing target which are different from initial target material. Nowadays, at the edge of ADS implementation, it seems to be instructive to compare the current status of SP-data with that of fission products (FP) at the edge of implementation of critical nuclear reactors. In his classical monograph on physics of nuclear kinetics, G. Keepin pointed out that measurement of about 120 FP resulted in a reasonably accurate picture of mass and charge distribution for major fuels [1]. Available data on FP yields in neutron induced fission is usually tabulated for thermal, 1 MeV and 14 MeV of incident neutron energies and this covers all the spheres of fission technology application. The situation with SP is much more complicated. Though the list of materials for spallation target of ADS is limited to several heavy elements (Sn, Ta, W, Hg, Pb, Bi, Th, U), the energy of incident protons considered in ADS ranges from hundred MeV to several GeV. In this energy interval nuclides with mass number characteristic to FP form only the subset of long list of SP ranging from light fragments like tritons and helium to isotopes closed to initial target materials. Because of high price and complexity of the experiments with accelerator, experimental data base on SP yields is rather limited. Before the burst of interest to ADS from technological community (that happened about two decades ago) the high energy experiments were almost entirely focused on specific aspects of nuclear physics, like problem-oriented cross-section systematics. As for the engineering community, in early ADS projects, it was sufficient to know spatial distribution of neutron and heat generation to estimate target design characteristics and this was well supported

by simulation with computer codes based on existing models validated on some integral experiments. This stage of research was comprehensively reviewed in Ref.2.

Further maturing of ADS development put forward the necessity of detailed knowledge of nuclear processes to estimate the dose impact for personnel, material damage, target cooling time and decommissioning scenarios. These challenges were considered in Ref.**3** which made an attempt to prioritize the needed experiments. In brief, the high priority list of isotopes and reactions was elaborated with the upper energy assumed at 200 MeV. Above this energy the high-energy transport codes are recognized to reveal sufficient predictive power. In the list of isotopes four nuclides belonging to the class of target materials ^{184}W, ^{208}Pb, ^{232}Th, ^{238}U were nominated [3].

Currently, for technological reasons liquid metal spallation targets keep the leading positions among candidates for ADS, lead and lead-bismuth eutectic being of the highest priority. Recent progress and history of studies on residual nuclides in lead irradiated by protons are summarized and reviewed in Ref.4. It is worth to refer briefly the activity of three experimental groups which keep nowadays the leadership in experimenting on spallation products' yields and the number of reactions inherent to analysis of lead targets. Given in Ref.4 are the results of experiments on 127 residual nuclides from lead for proton energies between 64.8 and 2.6 GeV identified in ZSR (Hannover) by gamma spectrometry [4]. For fixed proton energy of 1 GeV, production cross-sections of more than 400 nuclides were measured with direct gamma spectrometry at ITEP (Moscow) [5]. By the inverse kinematic method realized in GSI (Darmstadt) with magnetic fragment separation about 900 nuclides from 1 GeV/A ^{208}Pb+p reaction were identified [6]. As one can see the list of SP depends on experimental technique and this list anyway exceeds the list of fission products recognized as essential for reactor kinetic. Obviously such a long list of nuclides illustrates the skills of experimental groups and definitely might help to improve the theoretical models responsible for simulation of high-energy nuclear reactions. However the question about significance of SP accumulation is still open and this paper attempts in identification of particular group of SP that might effect on ADS performance. This is done on the basis of lead target which is treated as the number one candidate for ADS designing.

2. GLANCE AT MASS DISTRIBUTION

Figure 1 gives a typical illustration of SP yields obtained with computer code CASCADE (original Russian code developed especially for the high-energy simulation in Joint Institute of Nuclear Research (Dubna) and modified in Obninsk Institute of Nuclear Power Engineering (Obninsk) [7]. The reference energy is 1.6 GeV which is the highest one among ADS projects. Figure 2 incorporates the experimental data obtained by leading experimental groups with reference energy of 1 GeV [5]. As one can see from Fig.1 typical SP distribution curve reveals three classes of spallation products. One is represented by sharp peak in the region of light nuclides (isotopes of hydrogen and helium). The second one comprises intermediate nuclides with mass numbers characteristic to fission products of nuclear reactors. The third is formed near the mass number of initial target nuclei. The gap between the last two peaks for heavy nuclides (W, Pb, U) is filled by either products of high-energy fissions (descending heavy fragment tail) or residuals from evaporation stage following spallation reaction (sometimes they refer to this as "deep spallation"). Non-uniqueness of nuclear mechanism responsible for this region leads to significant uncertainties as it is shown in Fig.2 that gives an order-of-magnitude difference in experimental data. Simulation with help of theoretical models gives even more pronounced difference. Figure 3 reflects this fact by showing production cross-sections for ytterbium, neodymium and lead isotopes (incorporated data was taken partly from Ref.10 and added with results obtained with CASCADE [7] and MCNPX [11] codes. As it is seen for the case of lead isotopes, the closer residual nuclide to initial target material, the better agreement among the theoretical results and experimental data is observed. Ytterbium and neodymium

isotopes are representative of the residuals close to the valley between two peaks and theoretical modeling in this region definitely needs to be improved.

The detailed comparison of the models is out of the scope of the present report, though significance of the nuclides of this region is stressed in the next section.

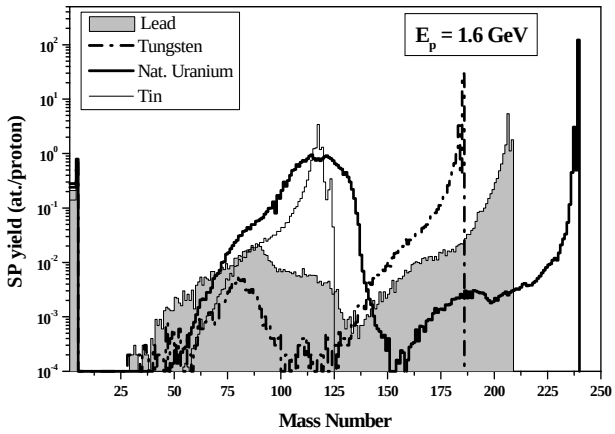


Fig.1 Mass distribution of spallation products in various targets irradiated by 1.6 GeV protons (calculated by CASCADE [7]) [8]

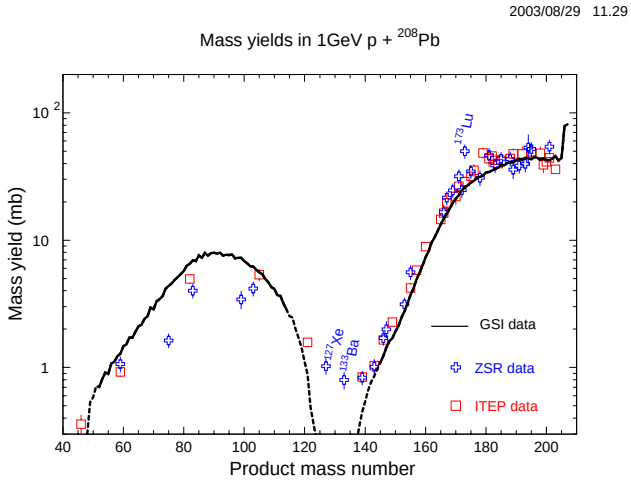


Fig.2 Mass distribution of spallation products from ^{208}Pb irradiated by 1 GeV protons (comparison of various experimental data [9])

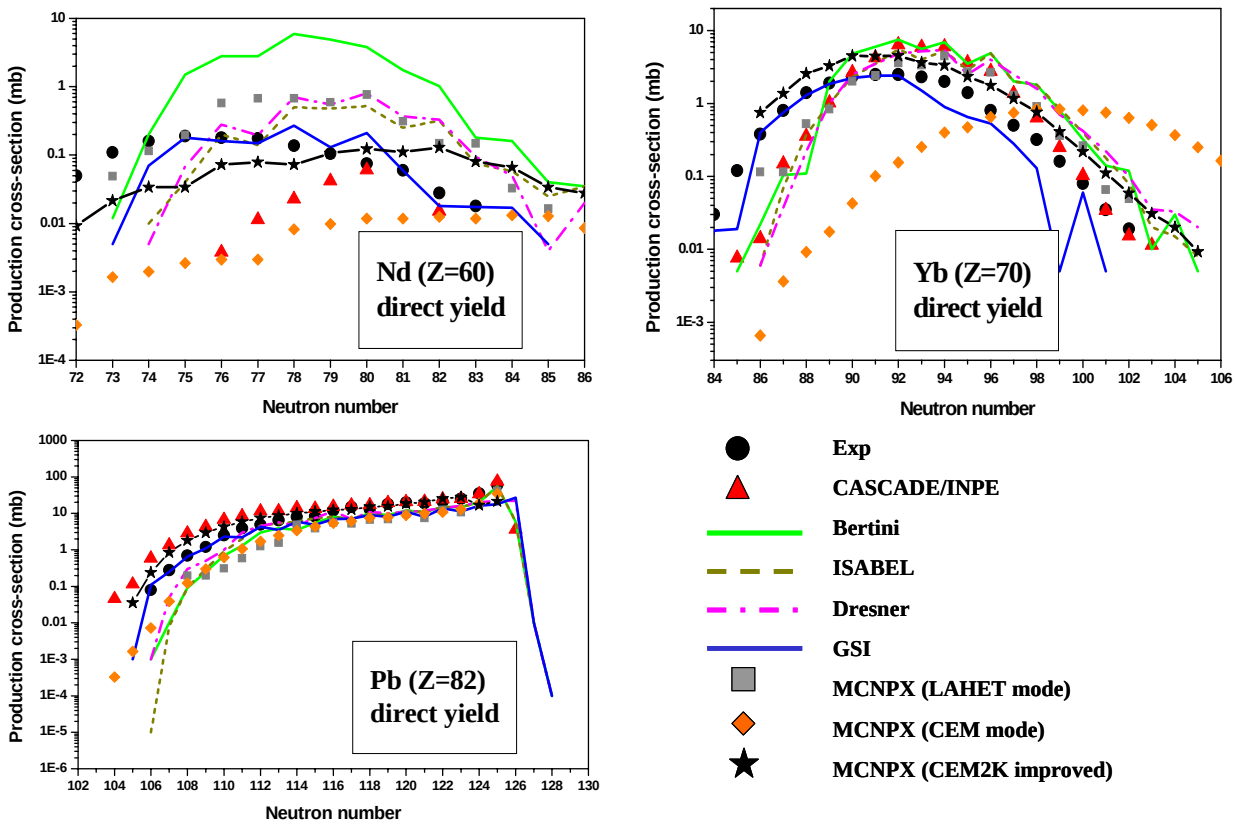


Fig.3 Independent yields of lead, ytterbium and neodymium isotopes in 1 GeV/A ^{208}Pb+p reaction obtained with various models and compared with experimental data reported in Ref.10

3. CHALLENGE OF SPALLATION PRODUCTS

So far the experimental studies on SP accumulation were oriented to validation of general predictive power of theoretical models and associated computer codes. In view of complexity of the processes involved it seems to be difficult to provide reasonable accuracy for all the range of mass numbers and, for this reason, it is instructive to select the nuclides of importance and concentrate the efforts on improving the accuracy in this particular area. Several criteria from technology side might be put forward. Among them radiation damage, corrosion and radiological issues are of the highest importance. Liquid metal targets give the possibility to reduce significantly the problem of radiation damage, (for example swelling due to accumulation of gaseous SP is avoided) and this fact stimulated interest to lead and lead-bismuth eutectic. Corrosion issues of SP accumulation have not been studied yet. The main focus was one the analysis of their radioactivity. Frequently targets are compared with respect to accumulation of polonium which is volatile and toxic (^{210}Po undergoes alpha decay with $T_{1/2}$ =138 days). Accumulation of ^{210}Po goes mainly by neutron capture in ^{209}Bi, thus giving the possibility of one-step production reaction in the Pb-Bi eutectic, and, at least two-step reaction in pure lead starting from neutron capture in double-magic nucleus of ^{208}Pb. Polonium was an important argument and continuous issue of contention in the debate of advantages and disadvantages of lead and lead-bismuth targets.

Several target design options were developed exclusively to response polonium challenge. Thus, polonium free operation was considered as the main point in favor of solid tungsten [12] and liquid tin [13] spallation targets. Lead-tin alloy (38.1Pb-61.9Sn) was proposed as a target material with no “serious polonium or toxic daughters’ accumulation” [14]. Opposite to aforementioned accentuation of polonium, in Russia there is an opinion that itsactivity is not a big problem and could not be a criterion in making the choice between lead and lead-bismuth, at least. The main reason, as they say, is that “the main contribution to long-lived activity ($T_{1/2}$ > 100 days) for both target materials is caused not by ^{210}Po, but by various isotopes of Bi, Te, Hg, and Au accumulated due to (p,xn) and (n,xn) reactions in high-energy part of proton and neutron spectra” [15]. For example, specific activity of mercury isotopes exceeds that of polonium by factor of 35, at proton energy and current of 800 MeV and 1–30 mA, correspondingly, and they single out the long-lived ^{194}Hg (520 yr) nuclide rather than polonium [16]. Whatever elaborated analysis of induced activity could be performed it would offer no more than pure nuclear physics and is insufficient to judge on how much adverse effect this brings to the human environment. Along with relatively high specific activity, implicitly included in the polonium concern is its alpha decay mode and volatile physical form whose combination brings polonium inhalation toxicity to an extreme. All this should emphasize the importance of toxicity units for characterization of induced activity. With respect to spallation products such a characterization was done by the authors of the present paper in Ref.17,18 that reveals the importance of rare earths. Among them there several alpha emitters (see Fig.4), four of them being with half-lives less than that of uranium $^{146}_{62}$***Sm*** ($T_{1/2}$=1×10^{8}yr), $^{148}_{64}$***Gd*** ($T_{1/2}$=74.6yr), $^{150}_{64}$***Gd*** ($T_{1/2}$=1.8×10^{6}yr) and $^{154}_{66}$***Dy*** ($T_{1/2}$=3×10^{6}yr). For incident proton energy of about 1 GeV, their location in mass distribution is in the problematic interval identified in the previous section. Figure 5 gives 90% fraction of the overall generated toxicity (which is just a translation of the cumulative yield (measured in atoms per proton) into toxicity units (ALI per proton)) for some representative reactions. For broad interval of proton energy these values are determined by only three nuclides: ^{113m}Cd ($T_{1/2}$=14.1 yr) for tin, ^{194}Hg($T_{1/2}$=520 yr) contributing generated toxicity of lead at low energies, and ^{148}Gd($T_{1/2}$=74.6 yr), which is significant for both tungsten and lead. Judging from Fig.5, overall generated toxicity (with exception of tin) is increased with energy increase and, what important, this growth is largely due to ^{148}Gd which is the RE isotope. Important to note, that tungsten advocated for his polonium-free advantage reveals the highest toxicity exclusively due to alpha activity of rare earths. Coming back to Fig.1 one can conclude that their

accumulation in W target is more pronounced than in Pb (or Pb-Bi) because mass number of initial nuclides is closer to this problematic region.

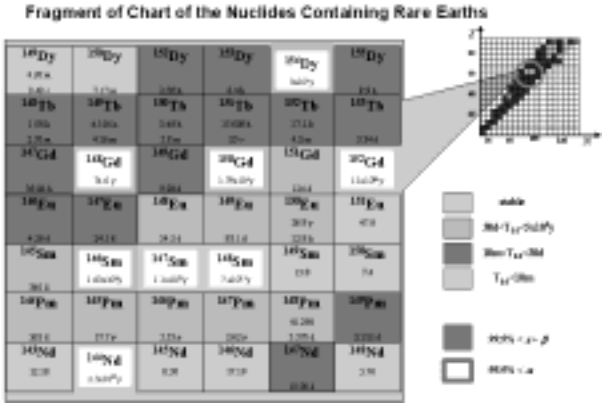


Fig.4 Fragment of Chart of Nuclides with alpha-emitting Rare Earths

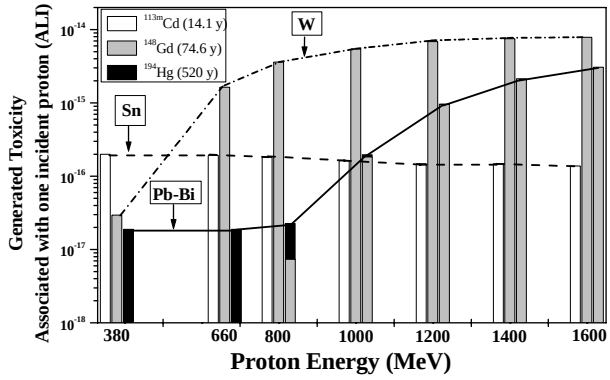
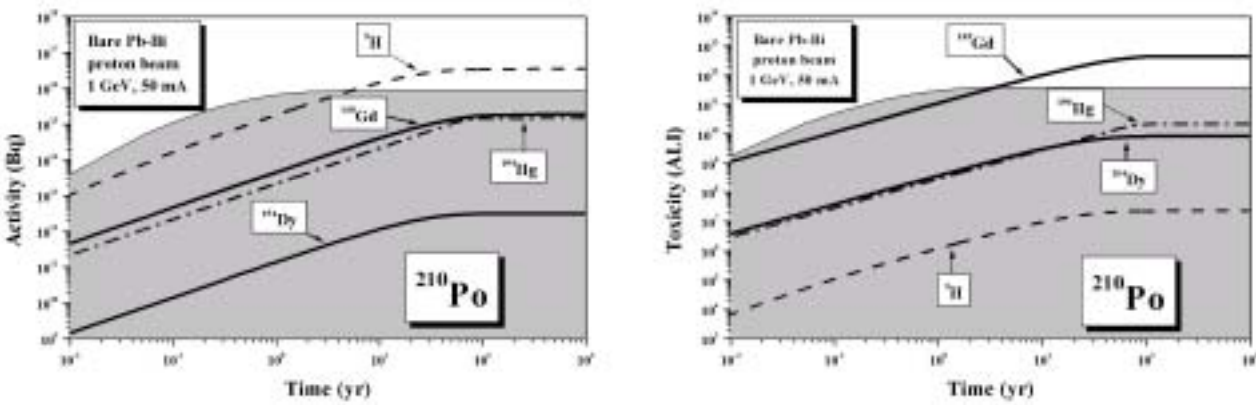


Fig.5 Toxicity from singular interaction of proton with one nucleus in Pb-Bi, W, and Sn targets as a function of proton energy [17]

Figure 6, a,b comprise the dynamics of ^{210}Po and important SP accumulation in large bare Pb-Bi target expressed in units of activity (Bq) and toxicity (ALI). Polonium reaches its equilibrium state within 1 year – operation time common for nuclear facilities between two successive shutdowns for maintenance. Other nuclides presented by Fig.6 require several decades. Since their half-lives longer than for ^{210}Po, their decay within maintenance periods is not essential, so, one can reasonably expect that all the selected nuclides reach equilibrium within the ADS lifetime. It is seen that for the reference beam parameters ^{210}Po equilibrium activity is inferior only to tritium (Fig. 6,a). Nevertheless, tritium hazard generally is not stressed in SP analysis. Implicitly, it warns that activity is a crude measure in hazard characterization. Figure 6,b underlines the significance of alpha emitting rare earths exemplified by ^{148}Gd ($T_{1/2}$=74.6 yr) whose equilibrium toxicity is almost by one order of magnitude higher than that of ^{210}Po. What important is that their toxicities become equal rather soon just after 3.5 years of operation. The essential result is that neither activity nor ^{210}Po is a criterion to select material for target of ADS. Rather than these, the toxicity of alpha emitting rare earths must be carefully looked at. Recently, similar conclusion was reported by Los Alamos group . They found that despite of low activity level of ^{148}Gd it encompasses almost two thirds of the total dose burden in the LANSCE facilities based on present yield estimates [19].



a) b)

Fig.6 Time dependent activity (Bq) and toxicity (ALI) of selected isotopes in Pb-Bi target irradiated with 1 GeV protons (beam current 50 mA)

Rare earths are known also for their large capture cross-sections and they are broadly used in reactors as burnable poisons. The effect of their accumulation on neutronics and reactivity coefficients in ADS is knot touched yet. That is subject of our future research.

3. CONCLUSIONS

The focus of this paper is on accumulation of spallation products from the view point of their effect on target performance. Rare earths spallation products were identified as the most problematic in terms of toxicity. Review of experiments on their accumulation showed the big discrepancy in available data. Theoretical evaluations do not agree well either. Thus improvement of data on rare earths production in spallation target is important for further successful ADS development.

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