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Nuclear Data and Activation of Heavy Metal Targets in Accelerator Driven Technology

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ABSTRACT

The nuclear data needed for the calculations and analysis for accelerator driven technology are discussed using the example of investigation of the nuclei concentrations and long-lived residual radioactivity accumulated in heavy liquid metal targets irradiated by high energy proton beam. The status of data used for spallation reaction cross sections and fission product contributions is analyzed. The necessity of the evaluated cross section data base development at intermediate energies is stressed.

1. Introduction

Requirements for nuclear data for Accelerator-Based Transmutation were discussed repeatedly [1] Practically they cover the energy interval from thermal energies to thousands MeV. Thus we should determine most important energy regions and reactions for a given problem. As an example we consider the problem of the accumulation of long-lived activity and isotope composition of heavy metal liquid targets, irradiated with a high energy proton beam.

2. The Analysis of Long-Lived Radioactivity

The analysis of radioactive nuclei accumulation was performed in a several papers [2-7]. The results of the calculations reduced to the same irradiation conditions differ sometimes by a few orders of magnitude (in particular, for isotope ^{210}Po ($T_{1/2}$=138 d) in lead-bismuth target). We have carried out the calculations of the activity accumulation and isotope concentrations in lead, lead-bismuth and mercury targets for various energies (400, 800, 1000 and 1600 MeV) and various geometry [8]. Main components were determined of the long-lived residual radioactivity and dominating channels were investigated. Special consideration of tritium accumulation was made and the comparison between different targets was performed. Analysis was made of the contribution of fission products to the target's activity and partial activities of main long-lived fission products were evaluated. Common features are found for the all target materials investigated.

3. Results and discussion

The results of calculations of the total activity and activities of some most important nuclides for the lead-bismuth target (60 cm length, 40 cm diameter) irradiated with 1 GeV protons as a function of cooling time are shown in Fig. 1.

One can see that various isotopes of Pt, Au, Hg, Tl, Pb and Bi make more significant contribution to the long-lived ($T_{1/2}$>100 d) activity than ^{210}Po isotope. The nuclides making main contributions to the total activity of the target after different cooling times were identified. The isotope ^{195}Au (half-life $T_{1/2}$=186 d) provides main contribution in the cooling time range 10 days 1 year together with the isotope ^{204}Tl ($T_{1/2}$=3.78 years, begins to

dominate after 3 years). The ^{195}Pt contribution becomes significant later. The ^{210}Po activity is 3-4 times lower than the activity of ^{195}Au. After three years the total activity is determined by ^{207}Bi nuclide ($T_{1/2}$=32.2 years).

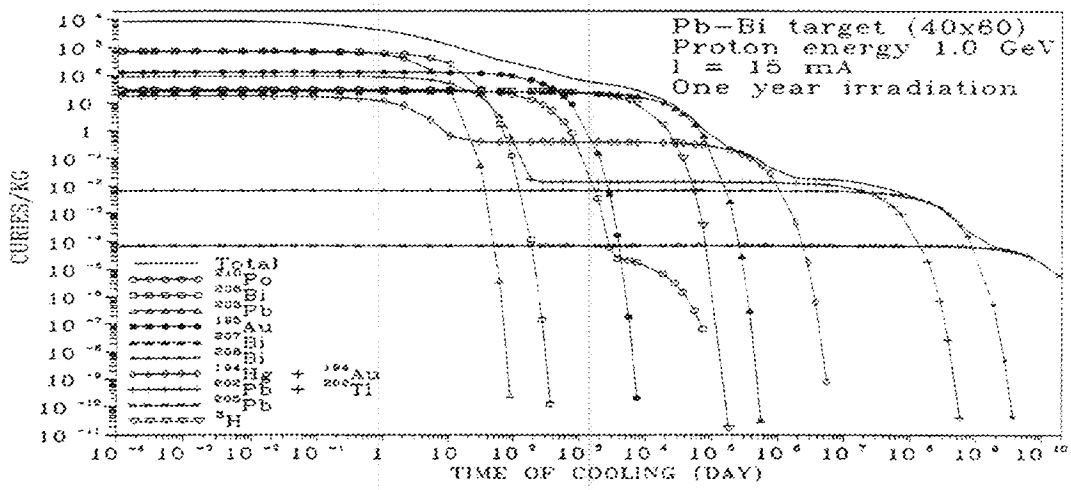

Fig. 1. Total and partial activities of lead-bismuth target irradiated with 1 GeV protons.

To evaluate correctly the possible uncertainties of the calculations and the influence of cross section data errors on the results it is necessary to analyze the spectral contributions of neutrons and protons to the accumulating activities. The components of the long-lived radioactivity in lead-bismuth target, accumulated due to the *(p,xn)* and *(n,xn)* reactions induced by various incident particles energies, are presented in Figure 2.

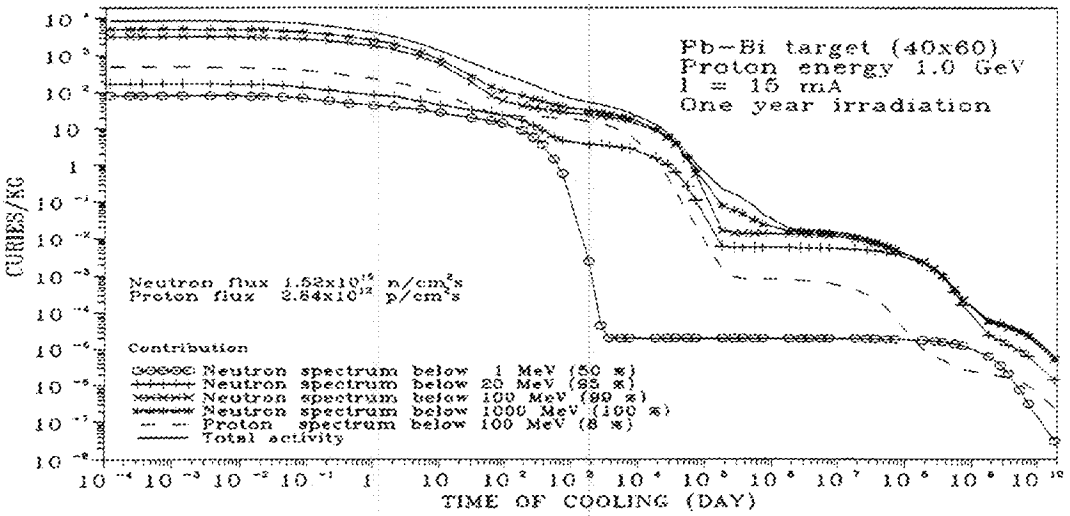

Fig. 2. Activity of lead-bismuth target due to various spectral components.

These results indicate that the dominating long-lived activities determined by platinum, gold, mercury and thallium isotopes are formed by hard components of proton and neutron spectra with the energies above 20 MeV. The soft component of neutron spectrum with the energies below 20 MeV corresponding to 95 % of the total neutron flux makes some 10-10^2 times lower contribution to the total long-lived activity ($T_{1/2}$>1000 d) than protons and neutrons from the hard part of the spectra comprising less than 5 % of the total flux of the particles. It must be pointed out however that the accumulation of long-lived isotopes ^{207}Bi ($T_{1/2}$= 1.39·10^4 d), ^{208}Bi ($T_{1/2}$=1.34·10^8 d) and ^{210}Po ($T_{1/2}$=138 d) is due to the *(n,γ)* reaction at low

energy. The total activity of those isotopes is several orders of magnitude lower than that of gold, mercury and thallium for cooling times about 1 year.
The status of the data for the production of spallation reaction products can be seen for the case of one of the main contributors - ^{193} Pt shown in Fig. 3.

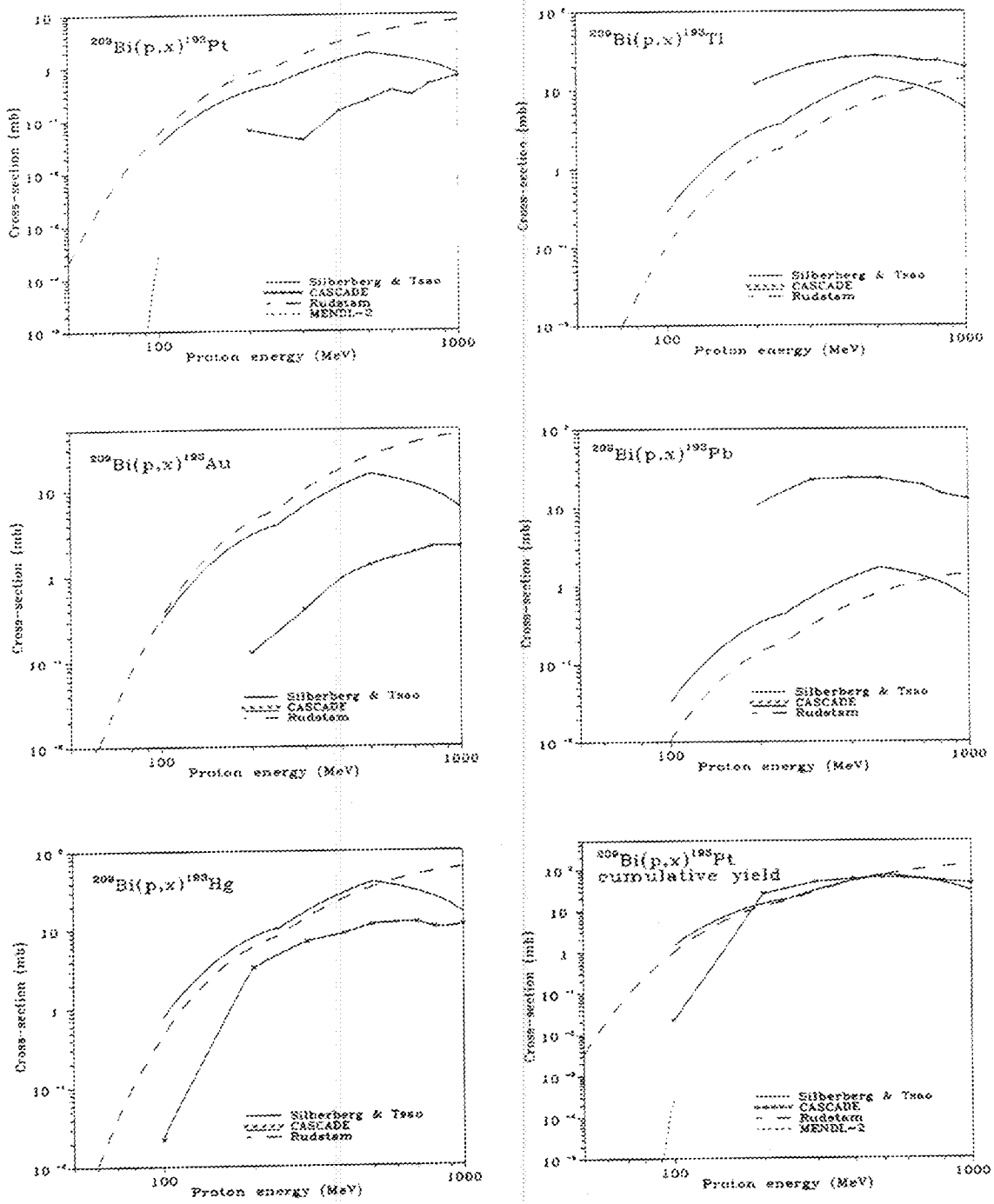

Fig. 3. The production cross sections for the nuclei with mass number 193 in ^{209}Bi(p,x) reactions calculated in various approaches.

One can see that the cross sections for the production of nuclei having the same mass number A=193, calculated with the CASCADE code [9] and with semiempirical systematics

of Silberberg-Tsao [10] and Rudstam [11], differ very strongly up to two orders of magnitude. For the cumulative yield of ^{193}Pt, however, the differences are not so large.
Our analysis demonstrates that the possible uncertainties of the results of calculations of the long-lived activity of the targets are determined by the errors of the cross sections of the threshold reactions at intermediate energies. The new version of MENDL-2 library has been developed in IPPE for neutrons up to 100 MeV and for protons up to 200 MeV [12]. Working out the MENDL-2 library we took into account recent results of our investigations of the preequlibrium emission of clusters, the pair correlations, shell and collective effects in nuclear level density on the base of the unified superfluid model and the set of the other improvements. The library has been tested on experimental data for the threshold reactions.
The CASCADE code and the empirical Silberberg-Tsao formulae were used to determine fission product yields for fission induced by high energy protons and neutrons. The resulting total activity as well as fission product contributions for the same target calculated using reaction cross sections determined with the CASCADE code and the semiempirical Silberberg-Tsao formulae are given in Fig. 4. One can see that for the fission products the difference between these two results can make factor 2-3.

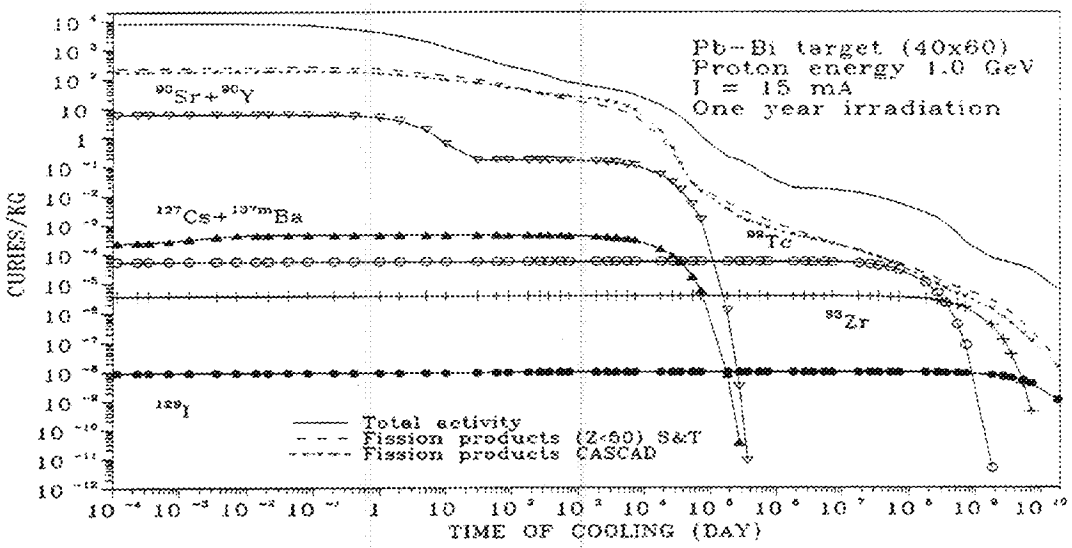

Fig. 4. Activity of lead-bismuth target due to fission products calculated using CASCADE code and the semiempirical Silberberg-Tsao formulae.

The results demonstrate that fission products increase the activity of target by 10-15 %. The situation is different for mercury target. For large cooling times the activity of mercury target is determined by fission product activity

Conclusions

The list of nuclei which make main contributions to the long-lived activity of Pb-Bi target was determined and analysis of the contributions of the proton and neutron spectral components to the accumulating activity was performed. The dominant contribution to the target activity is due to spallation reactions in a high energy (>20 MeV) parts of neutron and proton spectra. It is shown that in this energy region nuclear data calculated using different approaches differ considerably up to orders of magnitude and should be analyzed more carefully. Fission product yields calculated using various approaches can differ by factor 2-3.

Tritium accumulation is approximately equal in all targets and its contribution to the total activity is rather high. The library of evaluated data ENDF/B-VI does not describe tritium production cross section satisfactorily.

Acknowledgments

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