Subgroup 13
IEC-150

STATUS REPORT
On Intermediate Energy Nuclear Data Activity
IPPE, Obninsk, May 1996

1. The calculations according to Specifications for an International Codes and Model Intercomparison for Intermediate Energy Activation Data were performed. Three codes were used for different energy regions: ALICE/IPPE, DISCA, CASCADE.
2. Data library MENDL-2 was completed for proton induced reactions up to 200 MeV. At the moment some testing and analysis is carrying out. After final choice of FORMAT representation the library will be delivered to NDS IAEA and NEA Data Bank ~1 October 1996.
3. The calculations and analysis of isotope concentrations and accumulation of long-lived activity in heavy liquid metal targets (Pb, Pb-Bi, Hg) have been performed for various energies (400, 800, 1000 and 1600 MeV). The dominating components to the total radioactivity resulting from spallation reactions, fission and radiative capture for various irradiation and cooling times were determined. The estimations of spectral component contributions of neutron and proton fluxes to the accumulated activity were carried out. Most important energy regions for the problem were determined. The contributions of fission products to the target activity and partial activities of main long-lived fission products were estimated. The production of tritium in the targets was estimated and energy dependence of tritium production cross sections was analyzed.
4. Calculations and analysis of the excitation functions for the more than 50 reactions, induced by protons deuterons and alpha-particles, that are important for medical radioisotope production in the mass region Z=51-55 and Z=80-83 have been performed in a wide energy region up to 100 MeV.
5. The sensitivity to optical potential parameters and nuclear structure in reaction cross section calculations is investigated. The effect of imaginary part of optical potential on the intranuclear transition rates in hybrid model, on the calculated nonequilibrium emission spectra was considered. The comparison of calculated and experimental spectra indicate the anomalous energy dependence of imaginary part of optical potential. The probable reasons for such dependence are analyzed.

The effect of nuclear structure on the level density and on calculated reaction cross sections at intermediate energies was considered. The geometry dependent hybrid model has been used to obtain preequilibrium particle spectra. The calculated cross sections were compared with experimental data in the energy region up to 200 MeV. It is shown that generalized superfluid model is preferable for the better description of experimental data.
6. The description of the ALICE/IPPE and DISCA codes used for calculations are in preparation and can be presented ~1 October 1996.

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NUCLEAR DATA IN ACCELERATOR DRIVEN TRANSMUTATION PROBLEM.

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The nuclear data needed for the calculations and analysis for accelerator driven transmutation problem are discussed on the example of investigation of the nuclei concentrations and long-lived residual radioactivity accumulated in Pb,Pb-Bi and Hg targets irradiated by high energy proton beam. The dominating component to the total radioactivity of radionuclides resulting from fission, spallation reactions and radiative capture by target nuclei for various irradiation and cooling times were analyzed. The estimations of spectral component contributions of neutron and proton fluxes to the accumulated activity were carried out. The contributions of fission products to the target activity and partial activities of main long-lived fission products were evaluated. The accumulation of Po isotopes due to the reactions induced by secondary alpha-particles were found to be important for the Pb target as compared with two step radiative capture. The production of tritium in the targets and its contribution to the total target activity was considered and the status of data needed for this task are discussed. The nuclear data connected with beam attenuation, gas production and radiation damage in the accelerator-target window was considered also. The necessity of the evaluated cross section data base development at intermediate energies is stressed.

Introduction

It is clear that to study a feasibility of principal technologies in accelerator based transmutation problem and to work out the nuclear waste management concept it is necessary to know the nuclear data on spectra and reaction cross sections both for structural materials, long-lived fission product nuclei and minor actinides in a very broad energy range. Practically it covers the energy interval from thermal energies to thousands MeV. The status of nuclear data differs for different energy regions. It is well known how much efforts have been spent to get the libraries of evaluated neutron data ENDF/B, JENDL, BROND etc. So the situation is the best for energy region below 20 MeV. For the energy higher than 20 MeV neutron data are absent practically, and proton data are not systematized. The lack of the experimental data has to be compensated by the development of reliable calculation methods for the spectra and cross sections. The energy interval where data are needed is rather large, so various approaches have been developed that are based on different physical assumptions. Their predictive abilities are dependent on the applicability limits, details of elaboration, the experimental test possibility and the formulation of problem for which the method was worked out. To get the data needed various methods are used: both systematics with parameters selected from comparison with experimental data and models based on various assumptions. For the high energy region the calculation codes have been developed based on the intranuclear cascade model [1-4] (INC), combined with the evaporation model and radiation transport. The experience showed that these methods give the satisfactory results only for higher energies more than 100 MeV. On the other hand the evaluated data libraries (ENDF/B, JENDL, BROND etc.) are limited by energy 20 MeV. So the energy region 20-100 MeV requires special care. The high energy protons incident on the thick target of heavy elements lead to the spallation reactions with the emission both light particles (neutron, proton, alpha) and more massive nuclei. As a result one has rather hard neutron spectrum that extends from thermal energies to hundreds MeV. It is this neutron flux that give rise to various nuclear reactions leading to the transmutation both transuranium nuclei and fission products. Therefore to estimate the isotope composition and the activation of the target one needs the data in whole energy region and for all mass numbers. At the present for the whole energy interval from 1 to 1000 MeV the data on (n,xn) and (p,xn) reactions cross sections have been estimated by S.Pearlstein[5,6] and T.Fukahory and S.Pearlstein[7] for Fe, Pb and Bi nuclei only. It is impossible to have all needed data in a short time. Thus it is reasonable to evaluate data for concrete problems and to distinguish most important reactions, energy regions and nuclei for a given problem.

Energy Release, Beam Attenuation and Gas Production in the Window

Main part of the energy released by the beam in a thin window is connected with ionization losses. Nuclear interactions increase ionization losses by some 10-15%. Attenuation of the intensity of proton beam per cm of window thickness is approximately equal for both materials: 6.5 % for steel and 3.9 % for titanium. Estimates of energy release done by various groups agree within the accuracy of calculations which is 15-20 %.

Estimates of gas production in the target depend in a large degree on the reaction cross section reliability in a wide energy region. Figs. 1,2 show proton and alpha-particle production cross section on ^{56}Fe.

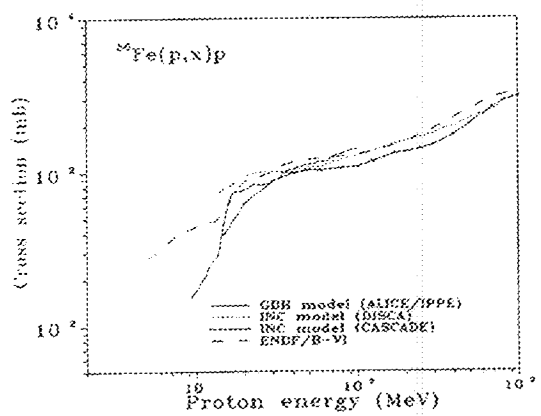

Fig. 1 Proton production cross section on ^{56}Fe.

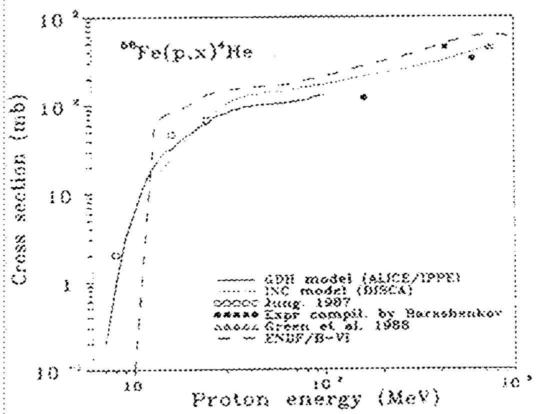

Fig. 2 Alpha-particle production cross section for $^{56}Fe(p,x\alpha)$ reactions.

The difference of evaluated cross sections for alpha-particle production taken from ENDF/B-IV and MENDL-2 [8] is about 20 % or more. According to proton yield evaluation the accumulation of hydrogen is approximately 6 times more than alpha-particle accumulation. For thin window approximately one half of alpha-particles and 2/3 of all protons will escape the window. On the other hand other particles such as deuterons, tritons and helium-3 will be accumulated also. The cross sections for these are evaluated with considerably larger errors than alpha-particle emission cross section. The uncertainties of such evaluations are probably not less than 50 %.

Radiation Damage of Target Window

Basic aspects of spallation radiation damage in accelerator-driven neutron sources were discussed by Wechsler M.S. et al. on example of tungsten [9] at Las Vegas Conference. Authors note that major concerns are associated with displacement, helium, and transmutation production The results of calculations of displacement cross section for tungsten using different approaches are shown in Fig. 3 taken from [9]. Our results are given with open cycles. One can see that the difference between calculation results is not so small and is probably due to inconsistency of data used in LAHET calculations.

The experimental data on target window radiation resistance for high energy protons are reviewed and analyzed in the paper of Ireland J.R. [10]. On the base of these data conclusion was made that the replacement of the window in the project of transmutation installation will be necessary only in a half of a year (if it will be necessary at all).

The results of the radiation damage calculations, that are defined by the values of the displacement per atom and hydrogen and helium accumulation, are shown in Table 1, where the results of Ref. [11] (OMEGA project), Refs. [12,13] (BNL project) and also the estimates on the base of Ref. [14] are given. The thickness of all windows was taken 0.5 cm, structural material was stainless steel. In the last column of Table 1 the values of displacements per atom in a year are given that were calculated using relation: dpa = $\sigma_d \cdot \Phi_p \cdot T$, where σ_d, the displacement cross section, was taken equal 3200 barn [14], T is the number of seconds in a year.

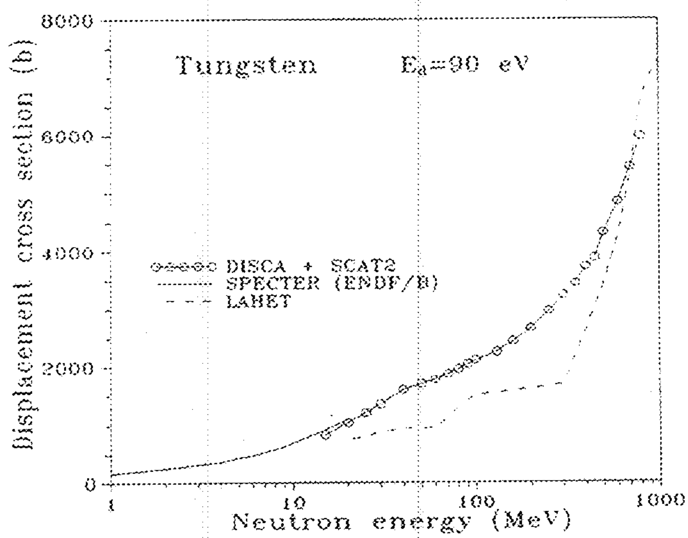

Fig. 3. Displacement cross section for tungsten.

The authors of paper [11] used the NMTC/JAERI codes, in Refs. [12,13] the LAHET and HTAPE codes were used. We see that the results of estimations not only are different, but contradict each other: to twice as larger fluence corresponds smaller (by order of magnitude) value of the displacement per atom. Our results are given in the last two lines. One can see that the discrepancies between different gas production estimates are not so large as compared with the values of dpa but are not negligible. Thus, the calculations of the radiation damage of the target window, that is important for technology problems, are rather sensitive to be a touchstone for the intercomparison of various approaches and codes, for the test of validity of the assumptions used in calculation and analysis.

Table 1. Radiation damage evaluations for target window.

Refs.	R,cm	E, GeV	I, mA	$\Phi_p, 10^{15}$	dpa/year	dpa'/year
[3]	23	1,5	39	0,146	60	15
[4-5]	8	1	10	0,312	6,07-8.8	32
	10	1	25	0,5		51
	10	1	60	1,2		123

The Analysis of Long-Lived Radioactivity

The analysis of radioactive nuclei accumulation was performed in a several papers [10,15,16,17]. 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). Such significant discrepancies can be due to cross sections used for some channels of isotope accumulation. That is why it is important to have reliable tested libraries of evaluated data.

Dominating Components of The Long-Lived Activity.

The results of calculations of the total activity and activities of some most important nuclides for the lead and lead-bismuth targets are shown in Figs. 4,5.

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 for both targets. The nuclides making main contributions to the total activity of the targets after different cooling times were identified. The isotope ^{195}Au (half-life $T_{1/2}$=186 days) provides main contribution in the cooling time range 10 days 1 year together with the isotope ^{204}Tl (half-life $T_{1/2}$=3.78 years, begins to dominate after 3 years). The ^{193}Pt contribution becomes significant later. For lead target the ^{210}Po contribution is by a few orders of magnitude lower than the contribution of those isotopes. Alternative isotopes dominate also in lead-bismuth target activity for the same range of cooling times. The ^{210}Po activity is only 2.5 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).

The comparison of the activities of lead and lead-bismuth targets irradiated by 800 MeV protons for one year demonstrates that the total activities of the targets begin to differ significantly only after one year. This is due to the formation of long-lived bismuth isotopes in the (n,2n) and (n,3n) reactions in lead-bismuth target (^{207}Bi and, for longer times, ^{208}Bi).

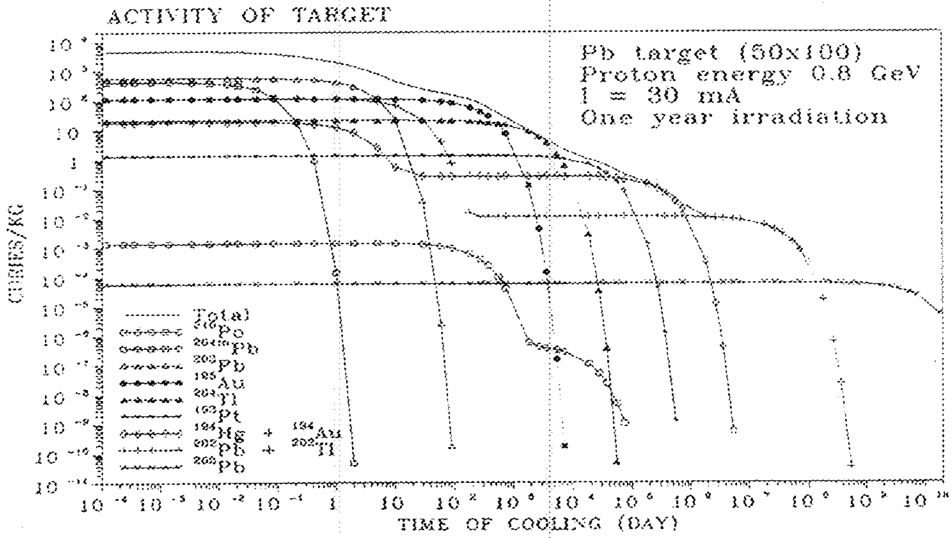

Fig. 4. Total and partial activities of lead target as a function of cooling time.

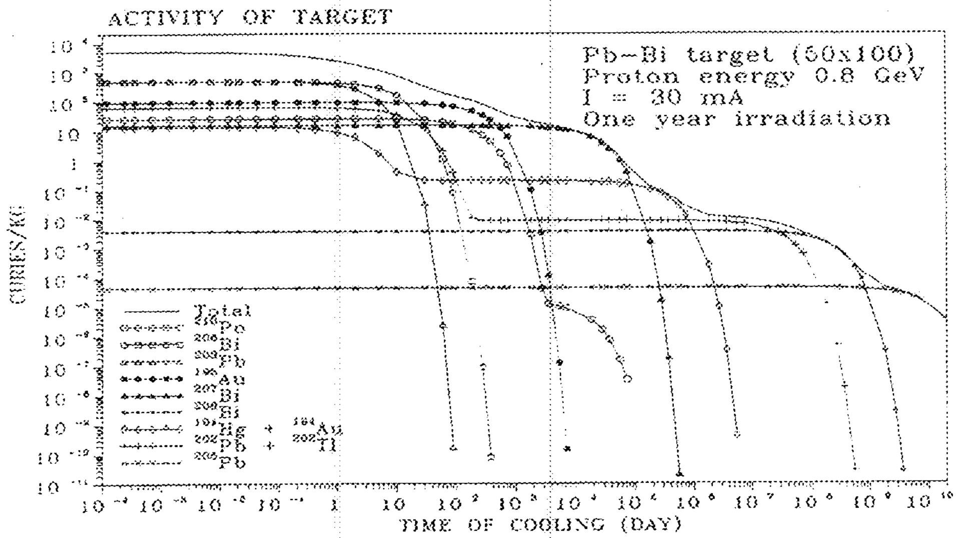

Fig. 5. The same as in Fig. 3 for lead-bismuth target.

Analysis of the Contributions of the Proton and Neutron Spectral Components to the Accumulating Activity

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 target, accumulated due to the (p,xn) and (n,xn) reactions, are presented in Figure 6.

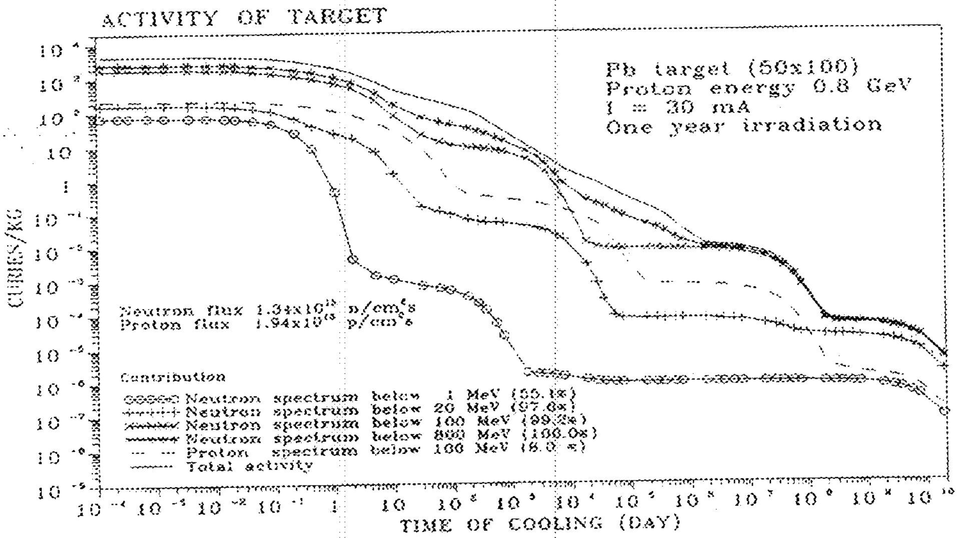

Fig. 6. Activity of lead target due to various spectral components.

The activities induced by the soft part of neutron spectra with En < 20 MeV are also presented. 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 96.6% of the total neutron flux makes some 10^3-10^4 times lower contribution to the total long-lived activity ($T_{1/2}$ > 1000 days) than protons and neutrons from the hard part of the spectra comprising less than 4% 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 days), ^{208}Bi ($T_{1/2}$=1.34·10^8 days) and ^{210}Po ($T_{1/2}$=138 days) is due to the (n,γ) reaction on soft neutrons. The total activity of those isotopes is more than 1000 times lower than that of gold, mercury and thallium for lead and lead-bismuth targets for cooling times longer than 1 year.

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 of protons and neutrons. The new version of MENDL library has been developed in IPPE [9] for neutrons up to 100 MeV and for protons up to 200 MeV. Working out the MENDL-2 library we took into account the preequlibrium emission of clusters, the pair correlations, shell and collective effects in nuclear level density on the base of the unified superfluid model. The library has been tested on experimental data for the threshold reactions.

Contribution of the Fission Products into Long-Lived Activity

To take into account the contribution of the fission products into targets activity it is necessary to include a lot of additional nuclides into kinetic equations. In our calculations we considered only the nuclides with half-lives longer than 5 hours. The empirical Silberberg-Tsao [18] formulae were used to determine fission product yields for fission induced by high energy protons and neutrons. The resulting total long-lived activities are given in Figure 7 as well as total activities of lead target irradiated for a year by the beam of 800 MeV protons.

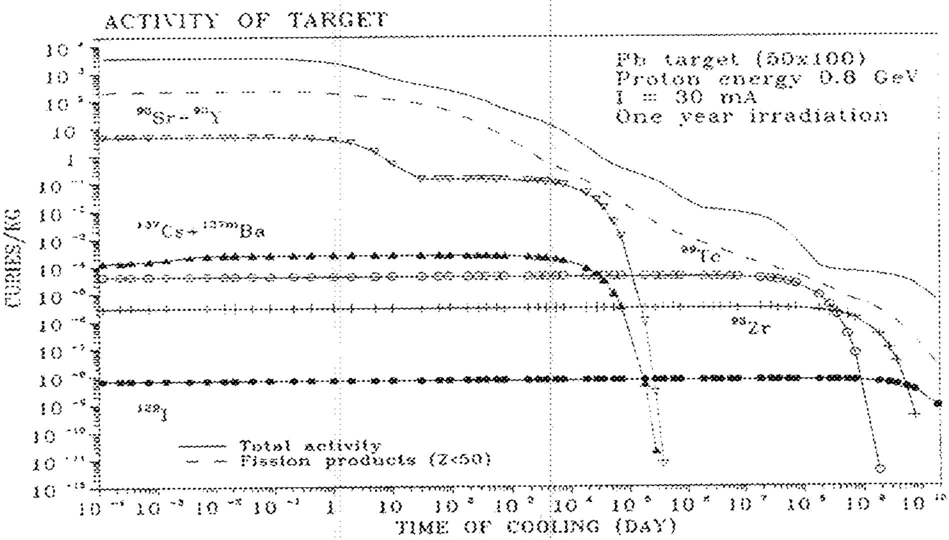

Fig. 7. Activity of lead target due to fission products.

The results demonstrate that fission products increase the activity of target by 10-15% for cooling times under 2 years and less than by 10% for longer cooling times. Well known long-lived isotopes ^{90}Sr, ^{137}Cs, $^{98,99}Tc$ and ^{129}I make largest contributions. The situation is different for Hg target. For large cooling times the activity of mercury target is determined by fission product activity (see Fig. 8).

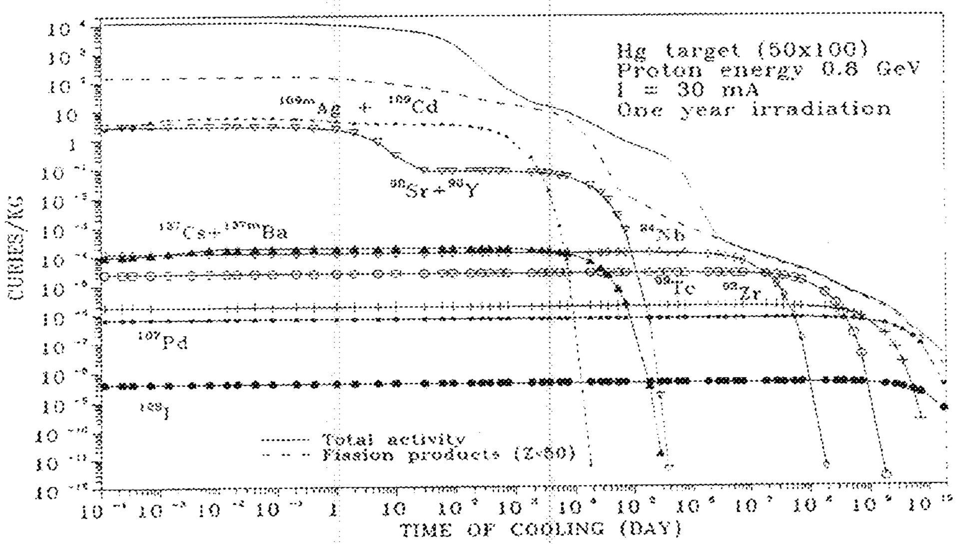

Fig. 8. Activity of mercury target due to fission products

Analysis of Tritium Accumulation in the Target

Tritium accumulation in the target requires special consideration due to special ecological danger associated with this isotope. To determine its accumulation in the reactions with high energy protons and neutrons it is necessary to have reliable enough evaluations of tritium production cross sections for nucleon energies above 100 MeV.

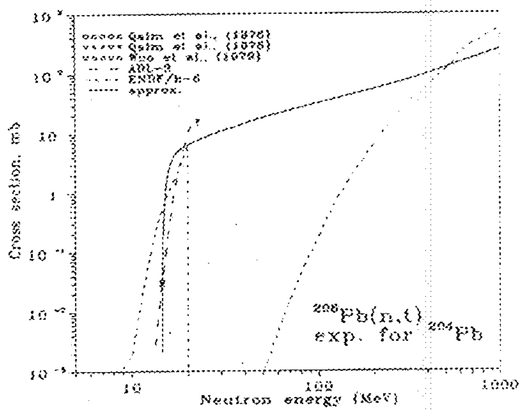

Fig. 9. Tritium production cross section for neutron induced reactions on lead.

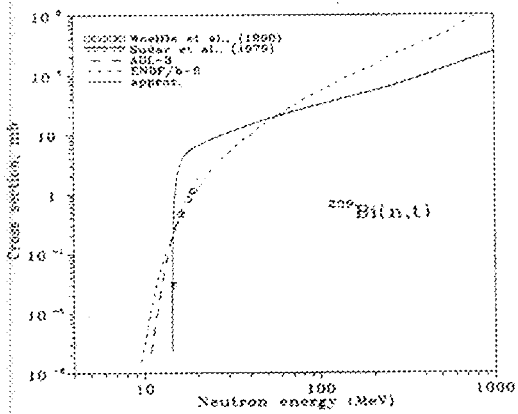

Fig. 11. Tritium production cross section for neutron induced reactions on bismuth.

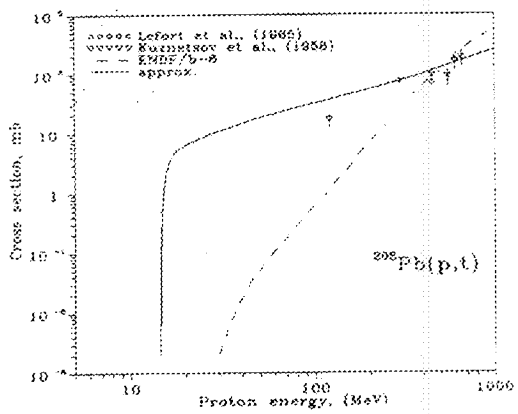

Fig. 10. Tritium production cross section for proton induced reactions on lead.

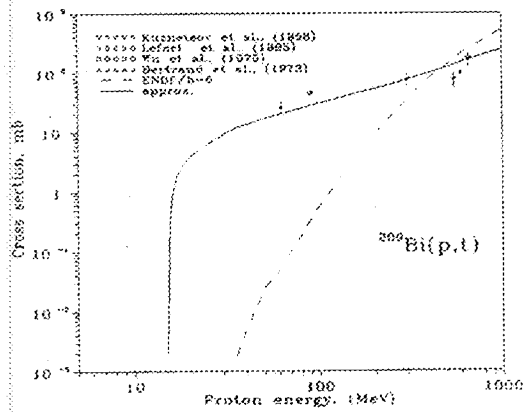

Fig. 12. Tritium production cross section for proton induced reactions on bismuth.

Experimental data available for lead and bismuth isotopes were analyzed together with different evaluations of tritium yields. ENDF/B-VI evaluation describes observable tritium production cross sections about 800 MeV satisfactorily but is in sharp disagreement with the experimental data available below 100 MeV. Semiempiric approximation of tritium production cross sections for high energy neutron and proton reactions [19] on lead and bismuth isotopes was developed on the base of experimental data and ADL-3 recommended values for the neutron energies below 20 MeV. Comparison of various evaluations with experimental data for neutron and proton induced reactions are shown in

Fig. 9-12 for the reactions $^{208}Pb(n,t)$, $^{208}Pb(p,t)$, $^{209}Bi(n,t)$ and $^{209}Bi(n,t)$. One can see that the evaluations of the ENDF/B-6 library are in a rather poor agreement with experimental data for the $^{208}Pb(n,t)$, $^{208}Pb(p,t)$ and $^{209}Bi(n,t)$ reaction. Tritium accumulation is approximately equal in both targets and its contribution to total activity is as high as 50% for lead target at cooling times from 3 to 30 years and is 2 times lower than the same figure for lead-bismuth target for the same time interval

Conclusions

The nuclei that make main contributions to the long-lived activity to Pb, Pb-Bi and Hg targets were 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 in this energy region where nuclear data should be analyzed to get reliable estimations for the task.

The contributions of fission products to the target activity comprise 10-15% of total activity for Pb and Pb-Bi target, well known long-lived isotopes make the largest contributions. For mercury target the activity of fission product determines total activity for large cooling times.

Tritium accumulation is approximately equal in all targets and its contribution to the total activity is as high as 50% for lead target at cooling times from 3 till 30 years and 2 times lower than the same figure for lead-bismuth target for the same time interval. The library of evaluated data ENDF/B-6 does not describe tritium production cross section satisfactorily.

Significant discrepancies exist in published evaluations of radiation damage of target window. The main reason of differences in this evaluations is the discrepancy in calculated spectra of secondary particles produced in nuclear collisions.

It is necessary to develop unified evaluated data base that is tested on most important element through intercomparison of various approaches.

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