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Transmutation of radioactive nuclear waste – present status and requirement for the problem-oriented nuclear data base

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Abstract. Transmutation of long-lived actinides and fission products becomes an important issue of the overall nuclear fuel cycle assessment, both for existing and future reactor systems. Reliable nuclear data are required for analysis of associated neutronics. The present paper gives a review of the status of nuclear data analysis focusing on the waste transmutation problem.

Keywords. Long-lived wastes; burner; transmutation efficiency; transmutation period.

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1. Introduction

A widely discussed issue is to resolve the problem of long-lived radiactive waste. There have been a number of attempts to estimate nuclear data needs related to the problem of radio-waste transmutation and to highlight priorities for future experimental and theoretical efforts [1–5]. The most important results are quantitatively reviewed here. To evaluate uncertainties in both the available and the required data on minor actinides, their effect on neutronics characteristics were investigated for the BN-800 reactor core design adjusted to transmutation of minor actinides (MA) [3,4]. The results of the analysis are presented in table 1.

Table 1. Existing and required (in brackets) uncertainties of actinide cross–sections [3,4].

Nuclide	Capture cross-section %	Fission cross-section %	Inelastic scattering cross-section %
Np-237	15 (5)	7 (3)	30 (10)
Pu-238	25 (10)	10 (5)	40 (30)
Pu-239	6 (4)	3 (5)	20 (15)
Pu-240	10 (5)	5 (5)	20 (15)
Pu-241	15 (5)	5 (3)	20 (20)
Am-241	10 (5)	10 (5)	30 (10)
Am-242m	30 (10)	15 (5)	40 (30)
Am-243	30 (10)	10 (5)	30 (30)
Cm-242	50 (10)	15 (5)	30 (30)
Cm-243	50 (10)	15 (5)	30 (30)
Cm-244	30 (20)	10 (5)	30 (30)

Similar approach was performed for sub critical fast reactor core with Pu/MA nitride fuel (MA/Pu ratio equal to 2) cooled by lead–bismuth [1]. This burner is a representative case reflecting most of the currently proposed accelerator-driven systems (ADS). The results are given in table 2.

It should be stressed that the main uncertainty in reactivity behavior (and associated decay heat) is revealed by MA in the energy range below 20 MeV, i.e. in the main part of the neutron spectrum. High-energy (above 20 MeV) data is essential only for radiation damage analysis.

Table 3 clearly indicates that the required uncertainty for MA data are comparable to that of major actinides and Pu isotopes currently used for reactivity analysis of conventional reactors.

An important point is that there are no reliable data concerning the effect of $(n, 2n)$ reaction in lead and bismuth on reactor criticality. Thus, to carry out evaluation of ^{206,207}Pb, inelastic scattering is considered to be the best for experimental analysis.

To sum up, it should be mentioned that the following areas cause much concern in terms of uncertainties:

- Data on the following microscopic MA cross-sections: $\sigma_{\mathrm{f}}, \sigma_{\mathrm{c}}, \sigma_{\mathrm{in}}$. It is imperative to carry out experiments and to obtain evaluated data in the energy range of 10 MeV to 0.1–1 keV.
- Data in the energy range above 20 MeV which have a significant influence on certain reactor parameters, in particular, on the radiation damage evaluation.
- Data on decay heat which basically depend on the data of minor actinides (Cm, in particular).

Table 2. K_{eff} uncertainties (%) for isotopes (no energy correlations) [1].

Isotope	σ_{cap}	σ_{fiss}	ν	σ_{el}	σ_{inel}	$\sigma_{(n,2n)}$	Total*
Pu-238	0.01	0.11	0.02	–	–	–	0.11
Pu-239	0.04	0.51	0.11	–	0.04	–	0.53
Pu-240	0.05	0.18	0.05	–	0.02	–	0.19
Pu-241	0.04	0.30	0.03	–	0.01	–	0.31
Pu-242	0.01	0.05	0.02	–	0.01	–	0.06
Np-237	0.24	0.70	0.21	–	0.14	–	0.78
Am-241	1.32	1.12	0.38	–	0.22	–	1.79
Am-242m	0.01	0.09	0.03	–	0.01	–	0.10
Am-243	0.74	0.59	0.21	–	0.60	–	1.14
Cm-242	–	–	–	–	–	–	–
Cm-243	–	0.05	0.01	–	–	–	0.05
Cm-244	0.13	1.09	0.18	–	0.07	–	1.11
Cm-245	0.01	0.41	0.08	–	0.01	–	0.42
Cm-246	–	–	–	–	–	–	–
Fe-56	0.03	–	–	0.05	0.49	–	0.50
Fe-57	–	–	–	–	0.06	–	0.06
Cr-52	0.01	–	–	0.01	0.03	–	0.03
Ni-58	–	–	–	–	–	–	–
Zr	0.03	–	–	0.03	0.07	–	0.09
N-15	–	–	–	0.19	0.01	–	0.19
Pb	0.02	–	–	0.10	0.41	0.02	0.43
Bi	0.04	–	–	0.11	0.49	0.03	0.50
Total*	1.54	1.97	0.54	0.25	1.05	0.04	2.77

2. Current status of nuclear model development

This section particularly deals with the nuclear reactor models engaged in theoretical evaluation. It is common to consider three energy intervals: <20 MeV, 20–200 MeV and >200 MeV. For the low energy region (below 20 MeV) the statistical models work well and major problems are related to the correct choice of model parameters (fission barriers and level density parameters). For energies higher than 20 MeV, the contribution of pre-equilibrium processes is essential. At the same time, it is widely accepted to use intra-nuclear cascade models only above 200 MeV, while at energies below 200 MeV various versions of statistical models with pre-equilibrium emission can be applied. As for quantum-statistical approaches, significant efforts are required to improve nuclear models.

The inconsistency of models at 200 MeV is illustrated in figure 1, where calculations of neptunium fission cross-sections by neutrons are done using known codes CEM [6] and STAPRE [7]. It is important to note that TALYS [8] code below 200 MeV and INCL [9] intra-nuclear cascade models have the same problem.

Cascade models were primarily developed to describe high-energy processes paying less attention to the statistical part. This simplification was to save computer

Table 3. Cross-section uncertainties for selected cross-sections: original uncertainty and required uncertainty to meet integral parameter target accuracy [1].

Isotope	Cross-section	Bound$^{(a)}$	Accuracy achieved (%)	Accuracy required (%)
Pu-239	$\sigma_{\rm fiss}$	4	6.5	3.4
		5	4	3.1
Pu-241	$\sigma_{\rm fiss}$	6	10	5.6
Np-237	$\sigma_{\rm fiss}$	3	25	8.0
		4	25	5.1
	ν	4	5	4.1
Am-241	$\sigma_{\rm cap}$	4	40	7.5
		5	40	5.5
		6	40	5.1
		7	20	5.9
		8	20	6.3
		9	20	6.9
	$\sigma_{\rm fiss}$	2	20	5.6
		3	20	4.6
		4	20	3.9
		3	5	3.8
	ν	4	5	3.3
Am-243	$\sigma_{\rm cap}$	4	40	10.4
		5	40	5.5
		6	40	5.1
		7	20	5.9
		8	20	6.3
		3	20	6.2
		4	20	5.4
		3	50	12.6
	$\sigma_{\rm inel}$	4	50	7.6
		5	50	12.0
		6	50	12.2
Cm-241	$\sigma_{\rm fiss}$	2	40	10.0
		3	40	8.5
		4	40	5.0
Cm-245	$\sigma_{\rm fiss}$	5	30	9.7
		6	30	9.6
Fe-56	$\sigma_{\rm inel}$	4	20	4.9
N-15	$\sigma_{\rm el}$	4	5	3.9
Pb	$\sigma_{\rm inel}$	1	40	20.4
		2	40	9.8
		3	40	10.6
		4	40	10.1
	$\sigma_{(n,2n)}$	1	100	21.5

Table 3. *Contd...*

Isotope	Cross-section	Bound[(a)]	Accuracy achieved (%)	Accuracy required (%)
Bi	σ_{inel}	1	40	18.8
		2	40	8.1
		3	40	9.3
		4	40	14.0
	$\sigma_{(n,2n)}$	1	100	17.5
	σ_{dpa}	1	20	20.0
		2	20	12.0
		3	20	12.1
		4	20	8.8
		5	20	20.0
		6	20	20.0
		7	20	10.9
	$\sigma_{(n,\alpha)}$	1	20	10.8
		2	20	20.0
	$\sigma_{(n,p)}$	1	20	15.1
		2	20	12.4
		3	20	20.0

[a]The group boundaries are given in table 4.

Table 4. Group boundaries.

Group	Upper boundary (MeV)	Group	Upper boundary (MeV)
1	19.6	9	$9.12{\cdot}10^{-3}$
2	6.07	10	$2.04{\cdot}10^{-3}$
3	2.23	11	$4.54{\cdot}10^{-4}$
4	1.35	12	$2.26{\cdot}10^{-5}$
5	0.498	13	$4.00{\cdot}10^{-6}$
6	0.183	14	$5.40{\cdot}10^{-7}$
7	$6.74{\cdot}10^{-2}$	15	$1.00{\cdot}10^{-7}$
8	$2.48{\cdot}10^{-2}$		

time when describing the branching reactions. As of now, this problem is not a crucial one and incorporation of pre-equlibrium model with multiple particle emission from excited nuclei is desirable. Certainly, there still exist many problems in determining basic physical values influencing the reaction mechanism. It is necessary to improve calculation of fission barriers with accuracy better than 1 MeV. The same might be addressed to masses of nuclei (the use of experimental values along with calculations of Mueller–Nix type inevitably leads to jump in values at the boundaries of experimentally known masses). Finally, it is necessary to develop models and perform calculations of fragment yields from excited nuclei both for evaluation of ADS target activation and for estimation of their influence on the ADS performance.

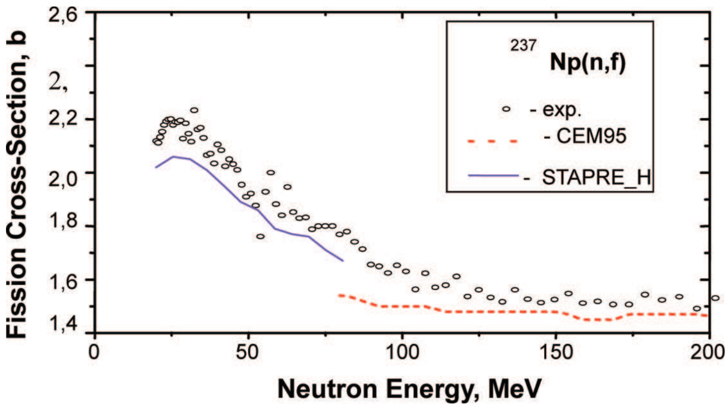

Figure 1. Neutron-induced fission cross-section of ^{237}Np at energies up to 200 MeV [10].

3. Effect of nuclear data uncertainties on radiation damage of structural materials

As it was stressed above, radiation damage is essential for high energy region. This problem includes gas production rate as well since the high level of gas production rate affects mechanical properties of materials. To obtain realistic estimation of lifetimes of the materials in such conditions, experimental data are necessary, whose amount at present is rather scarce [11]. In ref. [1] the following data on dpa and gas production rate uncertainties are shown (see table 5).

Partial correlation in energy

Currently, the code system NJOY is used for calculation of associated cross-sections. The NJOY code uses the NRT formulas [11,12] for the calculation of the damage energy. Such calculations are not correct for:

- the light secondary ions at the energies, where electronic loss is not proportional to $E^{1/2}$,
- the case of noticeable difference between Z and A for the secondary ion and material,
- mixtures of elements.

The situation may be improved by the use of the IOTA code [13]. In that case the number of defects calculated by the IOTA code should be introduced directly in the NJOY code substituting the defect number calculation by the DF function in the HEATR module.

The code IOTA was developed to obtain the total number of primary defects created in materials, the displacement cross-section and the space defect distribution. The calculations are performed with the help of different approaches described below. The simulation of the ion movement in the media is based on the binary

Table 5. Uncertainties (%) in radiation damage and gas accumulation rates in reference ADS [1].

	Max. dpa	Max. helium production rate	Max. hydrogen production rate	Max. (He/dpa)
$\Delta I_{\text{no correlation}}$	±29.9	±43.6	±28.5	±45.5
$\Delta I^{(a)}_{\text{PEC}}$	±48.9	±59.1	±53.1	±67.4

[a]The group boundaries are given in table 4.

collision approximation (BCA) and the Monte Carlo method. The experimental data for ion stopping power are used for the calculation.

Besides that, the special edition of the IOTA code allows to consider the inelastic nuclear interactions of the primary protons with materials. It uses the inelastic displacement cross-sections calculated with the help of the intranuclear cascade evaporation model for a wide number of materials [14,15].

4. Current status of activity in Russia for justification of nuclear data for transmutation

This section reviews the results obtained under 40 ISTC (International Science and Technology Center) projects dealing with measurements and evaluations of nuclear data for minor actinides, structural and heat carrying materials.

The main results of the ISTC projects are summarized as follows:

1. The spectra of prompt spontaneous fission neutrons from ^{240}Pu and ^{242}Pu as well as fission neutrons from thermal neutron fission of ^{243}Cm and ^{245}Cm were measured. Also the prompt neutron multiplicity distributions were measured for spontaneous fission of ^{244}Cm and ^{248}Cm and for fission of ^{235}U and ^{239}Pu by thermal neutrons.
2. Measurements of integral neutron inelastic scattering cross-sections, fission neutron spectra and mass-energy distributions of fission fragments for energies of emitted neutrons were done for ^{237}Np corresponding to the first plateau of the fission cross-section.
3. In the energy range from 1 to 7 MeV, detailed measurements of fission cross-sections for ^{238}Pu, $^{242\text{m}}$Am, ^{243,244,245,247}Cm were done.
4. The fission cross-sections for neutron-induced fission of ^{204,206,207,208}Pb, ^{209}Bi ^{232}Th, ^{233}U, ^{237}Np, ^{239}Pu and ^{243}Am at the energies from reaction threshold up to 200 MeV were measured.
5. The energy dependence change of delayed neutron yields for ^{237}Np in the energy range corresponding to the first plateau was measured. The analogous experiments are carried out at present for ^{233}U, ^{232}Th, ^{239}Pu and ^{241}Am, and for ^{239}Pu the incident neutron energy range will be expanded up to 18.0 MeV.
6. The systematic measurements and analysis for the basic structural materials of spectra and integral gamma-ray production yields from the interactions with 14-MeV neutrons were performed.

7. The evaluations were done and the general-purpose files were created for isotopes ^{232}Th, ^{231,233}Pa, ^{232,233,234,238}U, ^{239}Np, ^{238,242}Pu, $^{241,242g\&m,243}$Am and ^{243,245,246}Cm in the incident neutron energy range up to 20 MeV and for isotopes ^{237}Np, ^{240}Pu, ^{241}Am at neutron energies up to 150 MeV.
8. A comparison and verification of the methods for determining reaction product yields resulting from proton–nuclei interaction were made. An experiment was designed and performed whose aim was to irradiate targets made of ^{63}Cu and ^{65}Cu by 1.2 GeV protons with subsequent processing of results independently in ITEP and JAERI. To determine the yields of radioactive product nuclei formed in target and structural materials, various thin targets were irradiated 47 times (^{182,183,184,186}W at the proton energy of 0.2, 0.8 and 1.6 GeV; ^{nat}W, ^{56}Fe, ^{58}Ni and ^{93}Nb at 2.6 GeV; ^{232}Th, ^{nat}U, ^{99}Tc at 0.1, 0.2, 0.8, 1.2 and 1.6 GeV; ^{59}Co and ^{63,65}Cu at 0.2, 1.2, 1.6 and 2.6 GeV; ^{nat}Hg at 0.1, 0.2, 0.8 and 2.6 GeV and, in addition, ^{208}Pb at 1.0 GeV) and 4050 values of cumulative and independent yields of residual radioactive product nuclei with half-lives from 8 min to 32 yrs. The experimental values of nuclear reaction product yields were compared with the values calculated by LAHET, CEM95, CEM2k, CASCADE, CASCADE/INPE, YIELDX and INUCL which model hadron–nuclei interactions.
9. The independent and cumulative yields of radioactive residual product nuclei in thin targets made only of isotopes of Pb (^{206,207,208,nat}Pb) and bismuth (^{209}Bi) were determined experimentally and modeled theoretically. Thin targets made of ^{206,207,208,nat}Pb and ^{209}Bi were irradiated 55 times (at proton energy of 0.04, 0.07, 0.10, 0.15, 0.25, 0.4, 0.6, 0.8, 1.2, 1.4, 1.6 and 2.6 GeV) and more than 5900 cumulative and independent yields of residual radioactive product nuclei with half-lives from 8 min to 32 yrs were determined. Apart from the standard modeling of experimental results using various codes, some work was done on LAHET and CASCADE modification and new versions LAHETO and CASCADO were developed, respectively.
10. Measurements were made of secondary neutron spectra from ‘thin’ W – Na targets irradiated by 0.8 and 1.6 GeV protons. Prompt neutron spectra of the fission induced by bombarding isotopes of thorium, uranium and neptunium by 50 and 100 MeV protons were measured. The double differential cross-sections of neutron generation, with due account for errors, are given for 22 groups in the energy interval from 3.10 to 330 MeV. Fission cross-sections were measured for ^{204,206,207,208}Pb and for ^{209}Bi and ^{205}Tl. The fission cross-sections were determined for ^{nat}Pb and ^{209}Bi (mono-isotope). Absolute neutron- and proton-induced fission cross-sections for ^{nat}Pb and ^{209}Bi have uncertainties of about 10%. The data on neutron-induced fission cross-sections for the above nuclides are consistent with those found in literature, the maximum discrepancy (for ^{209}Bi at 97 MeV neutron energy) being about 20%. The data on proton-induced fission cross-sections are also consistent with the data available in literature.
11. The effective fission cross-sections and reactivity coefficients for Np-237 and Am-241 at the fast critical assemblies BFS-67, -69 (IPPE) spectra were measured.

12. The effective fission cross-sections for ^{235}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{244}Pu, ^{241}Am, ^{242m}Am, ^{243}Am, ^{243}Cm, ^{244}Cm, ^{245}Cm, ^{246}Cm, ^{247}Cm, ^{248}Cm were measured on the spectra of BIGR reactor, molten salt blankets MSB-1, -2 (VNIIEF) and thermal reactor MAKET (ITEP).
13. The isotopic composition change of U-235, Np-237, Pu-238, Pu-240, Am-241 and Cm-244 samples irradiated in the commercial fast reactor BN-350 with integral fluence of $\sim 2.2 \cdot 10^{23}$ n/cm^2 was obtained.
14. Interesting data were obtained while examining the power density distribution and threshold reaction rates in thick tungsten and NaCl+PbCl$_2$ targets irradiated by 0.8, 1.0 and 1.2 GeV protons. The investigated parameters are of great importance to the analysis and implementation of ADS projects. The measurement results of the threshold reaction rates for ^{209}Bi, ^{197}Au, ^{181}Ta, ^{169}Tm, ^{115}In, $^{\mathrm{nat}}$In, ^{93}Nb, ^{65}Cu, ^{63}Cu, ^{64}Zn, ^{59}Co, ^{27}Al, ^{19}F, ^{12}C and absorbed doses inside and on the external surface of 'thick' W–Na target irradiated by 0.8 GeV protons are presented. The calculation modeling of the measured reaction rates was made by means of the LAHET code used to calculate a stream of particles as well as excitation functions of the nuclides formed available in the MENDL, MENDL2p and IEAF libraries (in the range of $\sim$100 MeV) and in the LAHET calculations at energies above 100 MeV. The experimental and calculated results were compared and the most essential discrepancies were analyzed. A further study will be made on neutron field characteristics on the external surface of the 'thick' Pb target irradiated by 0.8 GeV protons.
15. The completed ISTC Project # 1486 devoted to investigation of feasibility to create MA burner on the basis of molten-salt cascade subcritical reactor revealed the necessity to improve the MA microscopic data, especially for ^{237}Np, ^{242m}Am.
16. The started ISTC Project # 2267 (SAD) might identify new requirements for future experiments and data evaluations in the course of Project implementation.

5. The nuclear data library TREF

As an attempt to fulfill nuclear data needs for transmutation problem, the neutron- and proton-induced nuclear data library for investigating particle transport in media has been prepared. The library entitled TREF (Transport Recommended Evaluated data Files) contains nuclear data for transport, heating and shielding applications in the energy region of primary particles from 10^{-5} eV up to 150 MeV [16]. Below 20 MeV neutron-induced data are taken mainly from ENDF/B-VI (Revision 8) [17]. For some isotopes the low energy data were adopted from JENDL-3.3 [18] and JEFF-3.1 [19] libraries. The TREF complements LA-150 library [20] with isotopes ranging from $Z = 8$ to $Z = 82$. The sub-library containing proton-induced data files is under development and should cover the same range of nuclides (from $Z =$ 8–82) as neutron sub-library does. Currently, proton-induced data files for 14 nuclides were completed. The evaluation of energy and angular distributions of secondary emitted particles above 20 MeV was performed with the help of the

ALICE/ASH code [21] and the analysis of the available experimental data. The total cross-sections, elastic cross-sections and elastic scattering angular distributions were calculated using the ECIS code realizing the coupled channel model [22].

6. Conclusions

Some of the essential conclusions are summarized below:

- The main bulk of the ISTC supported activity deals with improvement of nuclear data for transuranics isotopes (differential experiments: cross-sections in a broad energy interval, neutron yields, etc) and their effect on the integral characteristics of the facilities (integral experiments). Recognizing the importance of these studies, one should, however, stress that the nuclear data thus obtained are significant only at the early stages of transmutation i.e. for short irradiation time. In the foreseen accelerator-driven systems concepts, the equilibrium transmutation cycle is characterized by a dominant role of Pu-238 (about 25% in the isotopic mixture of all the heavy isotopes in the blanket). Uncertainty in its capture cross-section is 20% while the required accuracy is 10% (similar characteristics for Am-241 are 10 and 5%, correspondingly). It seems to be important to improve nuclear data particularly for Pu-238 and adjacent isotopes Np-237 and Np-238.
- Experimental data on the yield of residual nuclides accumulated in spallation target (the so-called spallation products) were obtained mainly for short-lived nuclides essential for the small proton fluences, while the accumulation of long-lived rare earth nuclides was not considered. Nevertheless some of these nuclides (like for example ^{146}Sm ($T_{1/2} = 1 \times 10^8$ yrs), ^{148}Gd ($T_{1/2} = 74.6$ yrs), ^{150}Gd ($T_{1/2} = 1.8 \times 10^6$ yrs), ^{154}Dy ($T_{1/2} = 3 \times 10^6$ yrs)) appear to be of high toxicity due to their alpha decay mode. In addition, some of them are known to be neutron poisons with anomalously large neutron capture cross-sections. It seems to be of importance to identify the domains of proton energies and neutron spectra in which their accumulation could significantly affect the safety of accelerator-driven transmuters.
- A large discrepancy in cross-sections has been observed between neutron-induced ($(n, xn), (n, pxn)$, $(n, 2pxn)$, etc.) and similar proton-induced reactions for transuranics, estimated by various models in the energy interval 20–150 MeV. It seems to be instructive to conduct the relevant experimental studies for isotopes of Th, U, Np, Pu and Am to validate theoretical models and computer codes. The first priority would be the reactions producing long-lived toxic nuclides like $^{238}\mathrm{U}(n, 7n)^{232}\mathrm{U}$, for example.
- The endurance of structural materials under irradiation might happen to be of crucial importance in justification of the transmutation strategy. Currently, the accumulated damage dose (in terms of displacements per atom), characteristic of gas accumulation is predicted with an accuracy of 15%. This accuracy was well assumed for the conventional reactors at the early stages of their development. At energies of about 1 GeV, uncertainties in these characteristics approach 25–50%. Needless to say, this gives a rather shaky estimation of the lifetime of structural components in the spallation target

region. In this connection, it seems to be instructive to improve the database for prediction of damage dose and gas accumulation.

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