Estimation of Transmutation of Long-lived Fission Products by Proton Beam

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Abstract

A set of nuclear transformations during an interaction of 600-MeV protons with macroscopic targets containing radionuclides ^{90}Sr, ^{129}I, and ^{137}Cs is considered using the semiempirical formulae and the SHIELD code. This work seeks to obtain some data for quantitative estimation of an efficiency of long lived nuclear power waste transmutation in proton induced reactions. The calculated data on yield of radionuclides, some stable isotopes, and secondary particles formation in radioactive targets are listed. The target activity reducing during an after irradiation cooling process is estimated. An experimental justification of the obtained evaluations on the base of the proton accelerator in Moscow Meson Facility is discussed.

1. Introduction : Alternative approaches to the transmutation problem

The nuclear power development perspectives are defined largely by possibilities of a potential danger level decreasing from an accumulation and long-time keeping of high-active wastes. Besides a controlled burial in stable geological formations, other versions are also considered, such as an extraction of some fractions in the space, as well as a transmutation using fast and thermal neutron reactors, charged particle accelerators and electric-nuclear type hybrid systems. In turn, the transmutation problem is subdivided into comparatively independent transmutation tasks for actinides and long-lived fission products.

Considering of the transmutation induced directly by photons or charged particles with low energies (of about 100 MeV), shows their insufficient effectiveness in comparing with neutrons obtained by means of reactor or electric-nuclear technique [1].

A possibility of their posterior using as a nuclear fuel [2] is an advantage of actinides transmutation in fast neutron reactors. However, a large number of cycles needed for an initial actinides amount transmutation leads to a high radiation loading of an equipment and a staff. This fact serves as a base of some pessimistic views at such a technique realization expediency [3]. In this connection, the actinides transmutation in high-flux thermal reactors gets an additional attractiveness because a possibility of achievement higher depletion during an irradiation cycle decreases actinides losses and increases a security during the processing time [4]. As to long-lived fission products, the efficiency estimation of their transmutation has been carried out both for fast [5] and for thermal [6] neutrons.

In addition to nuclear reactors, some transmutation versions for radioactive wastes are considered up to now using the electric-nuclear plants based on high-current charged particle accelerators both low and intermediate energies, which have relevant advantages from the stand-point of the nuclear security as compared to reactors.

The most elaborated versions of electric-nuclear installations are based on the neutron generation by the proton beam from the linear accelerator with an energy of about 1 GeV, in a target for which two schema are on the whole considered. In accordance with the PHOENIX design developed in the BNL (USA) [7], a target is constructed as a subcritical fast neutron reactor with a metal or oxide fuel. A conception of the ATW (Accelerator Transmutation of Waste) LANL (USA) [8] supposes a scheme with a liquid lead-bismuth target, surrounded by a heavy water, serving as a neutron reflector and moderator. In a heavy water reflector there are some loops in which the molted actinide fluorides circulate. A liquid-salted system is also considered, containing the actinides and strontium- and caesium-fluorides [9]. Cyclic systems with the light nuclei multiple passing at energies of about 100 MeV through a target-convertor [10,11] maintain approximately the same relative efficiency of a neutron generation as is supposed for high-energy $(1.0 \div 1.5)$ GeV beams.

We must note, without considering the problems dealing with the actinides transmutations, that among radionuclides, as the fission products, the most radiative danger is due to ^{90}Sr and ^{137}Cs, whose initial activity is $(1.8 \div 1.9) \times 10^5$ Ci/t and $(2.4 \div 2.5) \times 10^5$ Ci/t, respectively, that equals about 15 % of total fission products activity, for a cooling during 1 year, and about 90 % of a total activity, for a cooling during 10 years. As we can find in [5] some effective transmutation of ^{90}Sr and ^{137}Cs by neutrons may be carried out only in reactors with neutron fluxes exceeding $10^{16}\ n/cm^2 \times s$. Under this condition, an amount of ^{90}Sr and ^{137}Cs nuclei in their limits may be decreased by a factor of 10 (^{90}Sr) and 4 (^{137}Cs), and taking into account the real technical opportunities, we can get 5 and 2.5 times, respectively. On the other hand, according with estimations obtained in [12], expenditures intended to the proton transmutation

of ^{90}Sr and ^{137}Cs could be $12 \div 15\%$ of electric power, obtained during their generation process in the nuclear power plant. However, the real expenditures are probably more, which makes the transmutation ineffective and returns to a conception of long-time controlled keeping of these radionuclides during about 500–600 years.

As an alternative approach to this problem, we suppose, that it's expedient to consider a possibility of direct transmutation of fission products directly in the interaction of a proton beam with a conversion target, containing ^{90}Sr and ^{137}Cs isotopes that doesn't contradict the ATW conception as a whole. Despite that using of such a target leads to the neutron yield decreasing for the actinides transmutation by few times as compared with a target of heavy (lead, bismuth) materials, a possible reducing of the controlled keeping time for the high-active wastes is, in our opinion, a serious argument for the good of proposed opportunity investigation.

2 An interaction of proton beam with a conversion target.

2.1 Proton-induced reaction cross sections.

To estimate the direct transmutation correctly, a detailed analysis is needed of all of the possible channels of nuclear reactions, initiated by protons and leading to radioactive or stable isotopes production, which requires some information on the proton-nuclei interaction cross sections. However, the experimental data on nuclear reaction cross sections in radioactive targets are absent. This leads to a necessity of using of computational methods with an obligatory testing of the computed results by the experimental data, measured, to our regret, for stable isotopes of a considered element.

However, in the energy range near 1 GeV the experimental data for Sr and Cs are very scant [13]. That's why we carried out some cross section measurements for radionuclides formation at the energy of 1 GeV in targets containing Sr and Cs. These measurements were performed at the booster of the Institute of High Energy Physics IHEP (Russia) [14]. As a result, we have obtained some information on formation cross sections of more than 80 radionuclides, listed in tables 1 and 2. The table 3 shows that our measurement results agree well with available not numerous results of [15,16] carried out on the same target nuclei. In the tables 1 and 2, all of our obtained experimental cross section values are compared to the calculation results. We used the version of the cascade-evaporation model of nuclear reactions, realized in the hadron-nuclear generator of the SHIELD code [17], and an original SILBER code, based on the semiempirical formulae of Silberberg and Tsao [18]. The agreement degree of the calculated and experimental cross section values shows that both the cascade-evaporation model and the semiempirical formulae, on the whole, render correctly the quantitative characteristics of the proton-nuclear interactions for given target nuclei and initiating particle energies. The obtained results allow to turn to the next research step such as computational-experimental studying of the proton-nuclear interactions in macroscopic targets.

2.2 The transmutation rate estimation procedure.

To obtain the initial data in substantiation of the quantitative estimation of the transmutation effectivity for some long-lived fission products in the present work an amount of product nuclei formed by a proton beam was calculated.

For the targets in question, the excitation functions were calculated for the nuclear reactions with the stable and radioactive product nuclei formation in the atomic mass range of 7–136.

150 excitation functions were calculated for the ^{90}Sr-target, 259–for the ^{129}I-target, and 264–for the ^{137}Cs one. These excitation functions were calculated using the SILBER code. The subsequent numeric integration of these excitation functions by the proton ionization range in the target material allowed to obtain the values of stable and radioactive product nuclei yields, and for the last ones, the values of activities at the time of the irradiation finishing.

To obtain the quantitative estimation of the transmutation effectivity for the fission products in question, we used the following hypothetical parameters: the proton energy – 600 MeV; the average beam current – 50 mA; the target irradiation duration – 1000 hours; the target area – 1 cm^2; the target thickness equals to the proton ionization range; the target material – ^{90}Sr, ^{129}I and ^{137}Cs.

Under these conditions, almost 100 % of all target nuclei transmutate, which is in a good accordance with the previous transmutation rate estimations [15], obtained by using of similar initial parameters. For the test-control purpose an independent calculation was performed of the interactions amount by means of the SHIELD code. Table 4 lists the obtained data on the formation of the secondary neutrons and the product nuclei. All of the results are normalized by one initiating proton. A good agreement of the results obtained by means of SHIELD and SILBER increases a reliability of the resulting estimations.

2.3 Discussion of the results.

An analysis of a decay curve with a component extraction taking the main contribution to the total activity in any step of the radioactive decay is of practical interest.

Fig. 1 shows a dependence of relative (comparing to the initial one) change of ^{90}Sr and ^{137}Cs target activities with the time passing. These data confirm of that in 30 years an initial target activity decreases by a factor of about 10^2. If after a three-year cooling one could additionally realize a radiochemical separation procedure of ^{85}Kr from Sr and ^{137}Ba from Cs, in 30 years a waited activity decreasing would be about five exponents.

Fig. 2 shows the same dependencies for the ^{129}I-target. This proton-fission product transmutation is obviously ineffective, because the rest activity is greater than the initial even after 30-year-cooling. The radiochemical separation of ^{14}C, ^{39}Ar, ^{91}Nb, and ^{93}Mo doesn't change the situation radically, in spite of a greater difficulty comparing to Sr and Cs.

Fig. 3 shows a time-changing of an activity part due to ^{90}Sr and ^{137}Cs in the activity of a mixture of nonseparated fission products. The obtained data show that the proton transmutation of ^{90}Sr and ^{137}Cs accompanied with the subsequent radiochemical separation, allows to reduce effectively the defining contribution of these radionuclides to the total fission product activity.

Table 5 lists the effective half-life period values for the fission products in question. These values are calculated in accordance with an expression given in [19] for different flux densities of 600-MeV-protons initiating the transmutation. The calculation results [19] made for relevant thermal neutron flux densities are listed in brackets. It follows from the table 5 that in the case of ^{90}Sr proton and neutron using is practically similar. For ^{137}Cs, proton using is more preferable. As it was mentioned above, the ^{129}I transmutation with proton using is ineffectible.

Note, that in 500 years, in the burial condition, the ^{90}Sr– and ^{137}Cs–activities decrease by a factor of 5.5×10^6 and 9.6×10^6, respectively. At the same time, our calculated estimations show that the proton transmutation accompanied with the radiochemical separation gives a relevant reducing of these fission products controlled keeping time (more than an order). A transition to higher energies of initiating protons and taking into account the transmutation processes, due to the secondary neutrons, may lead to much more favorable estimations.

3 Conclusion.

The performance of more detailed investigations must include, in our opinion, a complex of computational-experimental tasks, in particular:

- space-energetic distribution calculations for the primary beam particles and the secondary nucleons in macroscopic targets of a real composition
- calculations of yields of stable and radioactive product nuclei, time dependencies for target activities and energy deposition parameters
- some experiments intended to measurement of depth and radial radionuclide yields from macroscopic stable targets and dynamics of an activity time changing
- an experiment intended to a measurement of proton-induced nuclear reaction cross sections in radioactive targets
- calculation methods testing in accordance with the experimental results

Putting into operation in the Moscow Meson Facility of the high-current accelerator with the proton energy of 600 MeV allows to realize a set of model and test experiments with macroscopic targets, including some experiments with radioactive targets not performed up to now. It's essential that a technical equipment of such experiments doesn't contain any unique elements and instruments, while means of detection and information processing are multipurpose enough. The experimental results obtained for macroscopic targets allow to realize a special-purpose verification of the calculation techniques, that is, to orient them to a comparatively narrow range of the target nuclei which results in an increasing of the calculation accuracy up to a level sufficient for the project estimations requirements.

The authors wish to acknowledge their gratitude to Yu.Ya. Stavissky for the helpful discussions during this work fulfillment.

References

[1] Tolstov K.D. On the transmutation problem for nuclear fission products. Preprint JINR, 3-90-583, Dubna, 1990.

[2] Doblin K. Transmutation of LWR high-level waste in LMR'S. Trans. Amer. Nucl. Soc. 64 (1991) 122.

[3] Sivintsev Yu.V. The radioactive waste transmutations in nuclear reactors. Atomn. technika za rubezhom, 11 (1992) 3.

[4] Kozar A.A. Waste ecological safety increasing after the actinide irradiation by thermal neutrons. Atomn. Energ. 75 (1993) 188.

[5] Bergelson B.R., Murisova L.T. The expedience and possibility substantiation of long-lived fission products transmutation in thermal neutron reactors. Preprint ITEP, 27-92, Moscow, 1992.

[6] Artisyuk V.V., Konobeev A.Yu., Korovin Yu.A., Sosnin V.N. Burn up of the long-lived radioactive products of ^{90}Sr and ^{137}Cs fission in a fast neutron flux. Atomn. Energ. 71 (1991) 184.

[7] Van Tuyle G., Takahashi H., Todosow M. e.a. The PHOENIX concept. Proposed incineration of long-lived radioactive wastes to produce electric power. BNL, Upton, N.Y., April 1990, Revised August 1990.

[8] Schriber S. Transmutation of waste using particle accelerators. Tenth Session of the Intern. Sem. of Nuclear War. Erice, Italy, August 19-24, 1990.

[9] Kazaritsky V.D., Blagovolin P.P. Radioactive wastes transmutation in a liquid-salted target from a high-power proton accelerator. Preprint ITEP, 56-91, Moscow, 1991.

[10] Kazaritsky V.D., Stepanov N.V. The transmutation yields in light nuclear beam irradiation. Preprint ITEP, 32-91, Moscow. 1991.

[11] Ado Yu.M., Ufimtsev F.G., Artisyuk V.V., et al. High-effective neutron generation in the deuteron accumulator for the radionuclide transmutation. Atomn. Energ. 73 (1992) 289.

[12] Blagovolin P.P., Kazaritsky V.D., Kiselev G.B. e.a. Transmutation of long-lived radioactive nuclear power wastes. Atomn. Energ. 70 (1991) 380.

[13] Iljinov, A.S., Semenov, V.G., Semenova, M.P., Sobolevsky, N.M., Udovenko, L.V.: Production of Radionuclides at Intermediate Energies. Springer-Verlag, Landolt Bernstein/New Series, subvolumes I/13a (1991), I/13b (1992), I/13c (1993), I/13d (1994), and I/13e (1994).

[14] Aleksandrov V.N., Semenov V.G., Sobolevsky N.M. Measurements and calculations of cross sections for spallation of strontium and cesium by 1 GeV protons. Workshop on nuclear transmutation of long-lived nuclear power radiowastes. 223, Obninsk, 1991.

[15] Felice P., Ocone R., Rindi A.: High energy proton interactions with Sr and Cs. Nucl. Instr. and Meth. 212 (1983) 359.

[16] Alekseev E.G., Guselnikov V.S., Zaitsev V.M. e.a. Production of bromine-76 and bromine-77 isotopes using the spallation reactions at proton energy of 1 GeV. Radiochimya 5 (1984) 667.

[17] Dementyev A.V., Sobolevsky N.M. SHIELD - a Monte Carlo Hadron Transport Code. See this Proceed.

[18] Silberberg R., Tsao S.H.: Partial Cross-Sections in High-Energy Nuclear Reactions and Astrophysical Applications. 1. Targets with Z<=28. 2. Targets Heavier then Nickel. Astrophys. J., Suppl. Ser. 25 (1973) 315.

[19] Bowman C.D., Arthur E.D., Lisowski P.W. et. al. Nuclear energy generation and waste transmutation using an accelerator-driven intense thermal neutron source. Nucl. Instr. Meth. A320 (1992) 336.

Table 1: **Experimental and calculated cross section values for radionuclides formation in strontium.**

	Radio-nuclide		Experiment			Calculation		
						SHIELD		SILBER
	Y-88		2.2	±	0.4	0.5		
*	Y-87		5.1	±	0.8	2.2	(m+g)	
*	Y-87	m	24.0	±	4.3			
	Sr-87	m	30.0	±	6.1	60.0	(m+g)	50.8
	Sr-85	m	7.7	±	1.2	17.6	(m+g)	15.8
	Sr-81		11.9	±	2.7	4.5		1.9
	Rb-86	(m+g)	25.0	±	4.2	16.4	(m+g)	21.4
	Rb-84		15.0	±	2.3	13.4	(m+g)	22.1
	Rb-84	m	13.7	±	2.4			
	Rb-83		25.0	±	4.5	14.3		21.9
	Rb-82	m	23.0	±	3.8	8.7	(m+g)	21.8
*	Rb-81		24.0	±	3.8	9.4		19.2
*	Kr-85	m	1.95	±	0.32	3.7	(m+g)	1.9
*	Kr-79	(m+g)	17.0	±	3.1	18.2	(m+g)	19.3
*	Kr-77		5.7	±	1.1	9.3		3.6
	Br-82	(m+g)	2.7	±	0.45	1.7	(m+g)	0.17
*	Br-77	(m+g)	17.0	±	2.7	9.6	(m+g)	22.1
	Br-75		12.0	±	2.1	6.8		5.2
	Se-75		18.0	±	2.8	15.1		17.1
	Se-73	(m+g)	13.0	±	2.3	12.0	(m+g)	9.0
*	Se-72		7.0	±	2.3	12.9		2.7
	As-74		7.6	±	1.2	4.1		6.2
*	As-71		9.7	±	1.8	8.2		8.2
*	Ge-69		12.0	±	1.9	11.5		8.7
*	Ge-68		9.0	±	2.0	13.9		4.4
*	Ga-67		6.1	±	1.1	8.5		10.1
	Zn-71	(m+g)	0.3	±	0.05	< 0.2	(m+g)	0.02
*	Zn-65		13.8	±	2.4	12.3		8.9
	Co-60	(m+g)	3.0	±	0.5	1.5	(m+g)	2.2
	Co-58	(m+g)	6.4	±	1.1	3.6	(m+g)	3.9
	Co-57		2.4	±	0.4	4.3		1.8
	Co-56		1.1	±	0.18	0.5		0.40
	Fe-59		0.85	±	0.14	1.2		0.46
	Mn-54		3.9	±	0.65	1.7		3.2
	Mn-52	(m+g)	1.1	±	0.18	< 0.3	(m+g)	0.51
*	Cr-51		3.6	±	0.57	3.6		1.8
	V-48		1.1	±	0.17	< 0.2		0.59

* – cumulative yield

Table 2: **Experimental and calculated cross section values for radionuclides formation in caesium.**

	Radio-nuclide		Experiment			Calculation SHIELD		SILBER
	Ba-131	(m+g)	3.2	±	0.5	2.1	(m+g)	
	Cs-132		53.0	±	10.5	64.7		49.3
	Xe-127	(m+g)	20.0	±	3.0	17.4	(m+g)	21.8
	Xe-125	(m+g)	28.0	±	6.3	18.0	(m+g)	21.5
	I-126		16.0	±	2.7	4.5		12.0
	I-124		11.0	±	1.8	8.3		6.0
	I-123		28.0	±	5.1	11.1		13.2
	I-119		13.0	±	2.1	5.7		4.5
	Te-123	m	3.4	±	0.6	5.3	(m+g)	
	Te-121		23.0	±	3.5	11.8	(m+g)	5.9
	Te-121	m	6.3	±	1.0			
	Te-119		18.0	±	3.1	15.1	(m+g)	16.7
	Te-119	m	7.8	±	1.4			
	Te-117		23.0	±	5.5	15.6		7.7
	Te-116		11.4	±	1.7	15.0		4.8
	Sb-124		0.34	±	0.06	< 0.3	(m+g)	
	Sb-120		2.2	±	0.35	2.8	(m+g)	1.4
	Sb-120	m	2.1	±	0.4			
	Sb-118	(m+g)	3.5	±	0.6	4.8	(m+g)	7.0
	Sb-116	m	14.8	±	2.5	5.8	(m+g)	14.8
*	Sb-115		23.0	±	4.1	8.4		11.1
	Sn-117	m	3.9	±	0.7	4.9	(m+g)	2.0
*	Sn-113	(m+g)	28.0	±	8.5	21.3	(m+g)	12.2
	In-114	(m+g)	1.5	±	0.25	2.5	(m+g)	2.6
*	In-111	(m+g)	20.0	±	3.6	7.5	(m+g)	15.1
	In-110	m	9.6	±	1.6	7.5	(m+g)	8.2
	In-109	(m+g)	6.8	±	1.3	8.6	(m+g)	4.7
	Cd-111	m	3.5	±	0.7	4.2	(m+g)	3.8
	Pd-100		5.1	±	0.9	11.2		0.57
	Rh-101	m	12.6	±	2.1	3.0	(m+g)	7.2
	Ag-110	(m+g)	0.82	±	0.15	< 0.5	(m+g)	0.83
	Ag-106		3.2	±	0.5	5.1	(m+g)	10.1
	Ag-106	m	5.5	±	1.0			
	Ag-105		19.0	±	4.8	6.6		7.0
*	Ru-103		0.24	±	0.42	< 0.3		0.29
	Ru-97		7.6	±	1.5	8.9		1.8
	Tc-96	(m+g)	4.0	±	0.7	1.7	(m+g)	3.6
	Tc-95	(m+g)	7.9	±	1.5	1.8	(m+g)	2.3
	Tc-94		1.6	±	0.35	2.4		0.79
	Mo-93	m	5.3	±	1.0	4.4	(m+g)	3.1
*	Zr-89	(m+g)	4.4	±	0.7	3.1	(m+g)	1.3
	Zr-88		4.8	±	1.3	4.0		0.70
	Y-88		1.3	±	0.4	1.0		0.90
*	Y-87		3.9	±	0.7	1.0		1.7
	Rb-84	(m+g)	1.1	±	0.17	< 0.07	(m+g)	0.3
*	Rb-83		0.68	±	0.12	< 0.2		0.75
*	Se-75		0.46	±	0.09	< 0.2		1.1

* – cumulative yield

Table 3: **Some experimental cross section values comparing for radionuclides formation in strontium and caesium.**

Formation cross section, mb						
Target-nuclear	Radio-nuclide	Our data [14] 1.0 GeV	Data from [15] 0.6 GeV	Data from [16] 1.0 GeV
Sr	Y-88	2.2 ± 0.4	1.5 ± 0.3	
	* Y-87	5.1 ± 0.8	2.0 ± 0.4	
	Sr-87 m	30.0 ± 6.1	14.4 ± 4.6	
	Rb-84	15.0 ± 2.3	21.2 ± 3.3	20.5 ± 1.0
	Rb-82 m	23.0 ± 3.8	17.5 ± 1.9	
	Kr-79 m+g	17.0 ± 3.1		17.5 ± 1.9
	* Kr-77	5.7 ± 1.1		6.5 ± 1.2
	As-74	7.6 ± 1.2	10.6 ± 1.6	8.6 ± 0.9
Cs	Cs-132	53.0 ± 10.5	59.0 ± 6.2	
	I-124	11.0 ± 1.8	10.8 ± 1.5	
	Sb-118 m+g	3.5 ± 0.6	3.8 ± 0.7	

* – cumulative yield

Table 4: **Formation of the secondary neutrons and product nuclei in the interaction of the primary proton beam with the macroscopic targets.**

Target	Secondary neutrons SHIELD		Product-nuclei	
	E<10.5 MeV	E>10.5 MeV	SHIELD	SILBER
Sr-90	4.22	1.25	1.18	1.23
I-129	6.30	1.26	1.34	1.13
Cs-137	5.77	1.35	1.02	1.06

Table 5: **Effective half-life period decreasing for some fission products in dependence of the flux density of the particles initiating the transmutation.**

Particle flux density	Effective half-life period in years for some fission products					
$1/cm^2s$	^{90}Sr		^{129}I		^{137}Cs	
10^{14}	26.0	(28.6)	219.0	(11.0)	27.0	(30.0)
10^{15}	14.7	(15.0)	21.9	(0.9)	13.6	(25.0)
10^{16}	2.7	(2.0)	2.2	(0.08)	2.3	(7.0)
10^{17}	0.3	(0.25)	0.2	(0.009)	0.25	(0.9)

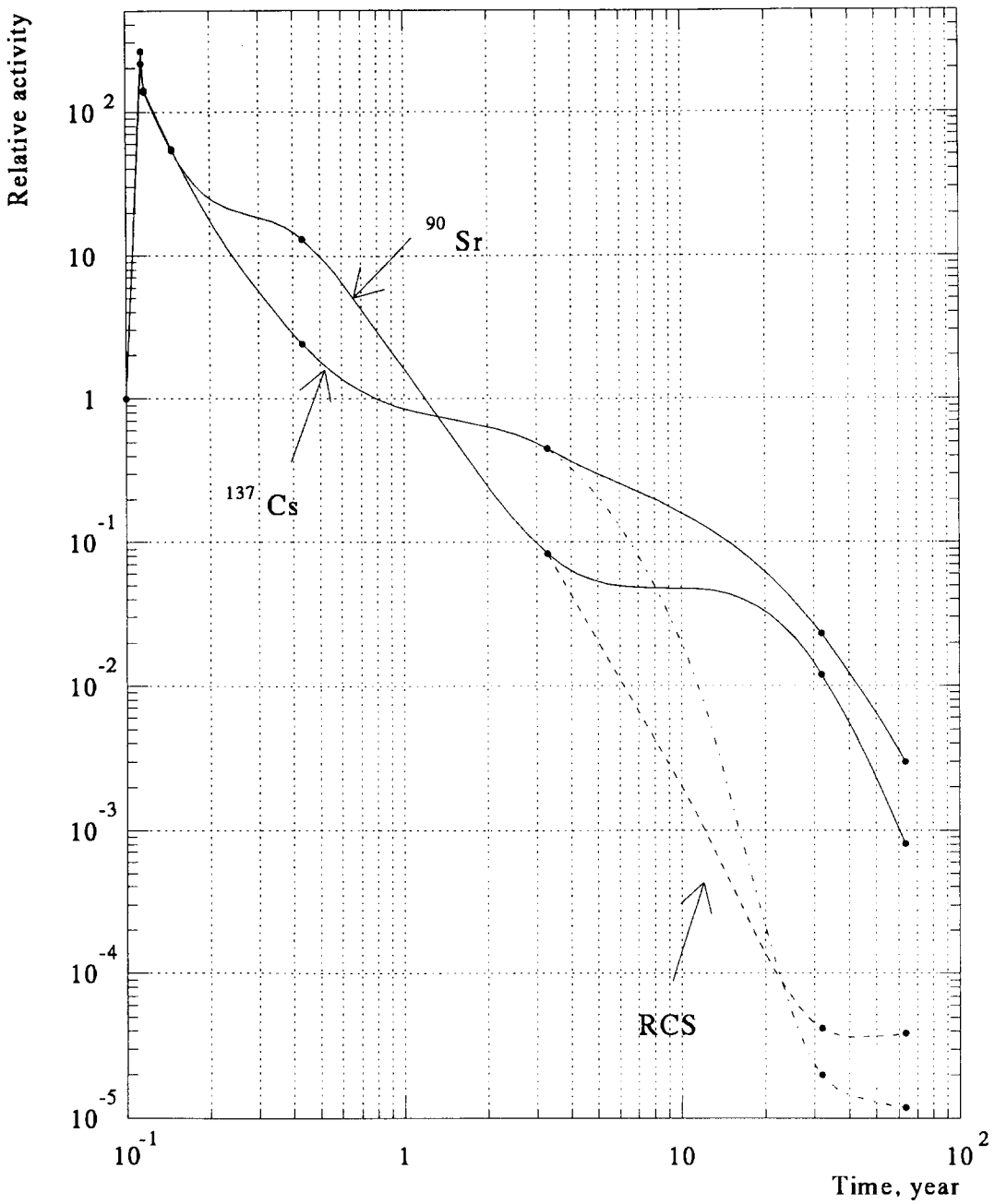

Figure 1: **Time dependence of the relative activities of ^{90}Sr and ^{137}Cs targets (RCS – radiochemical separation).**

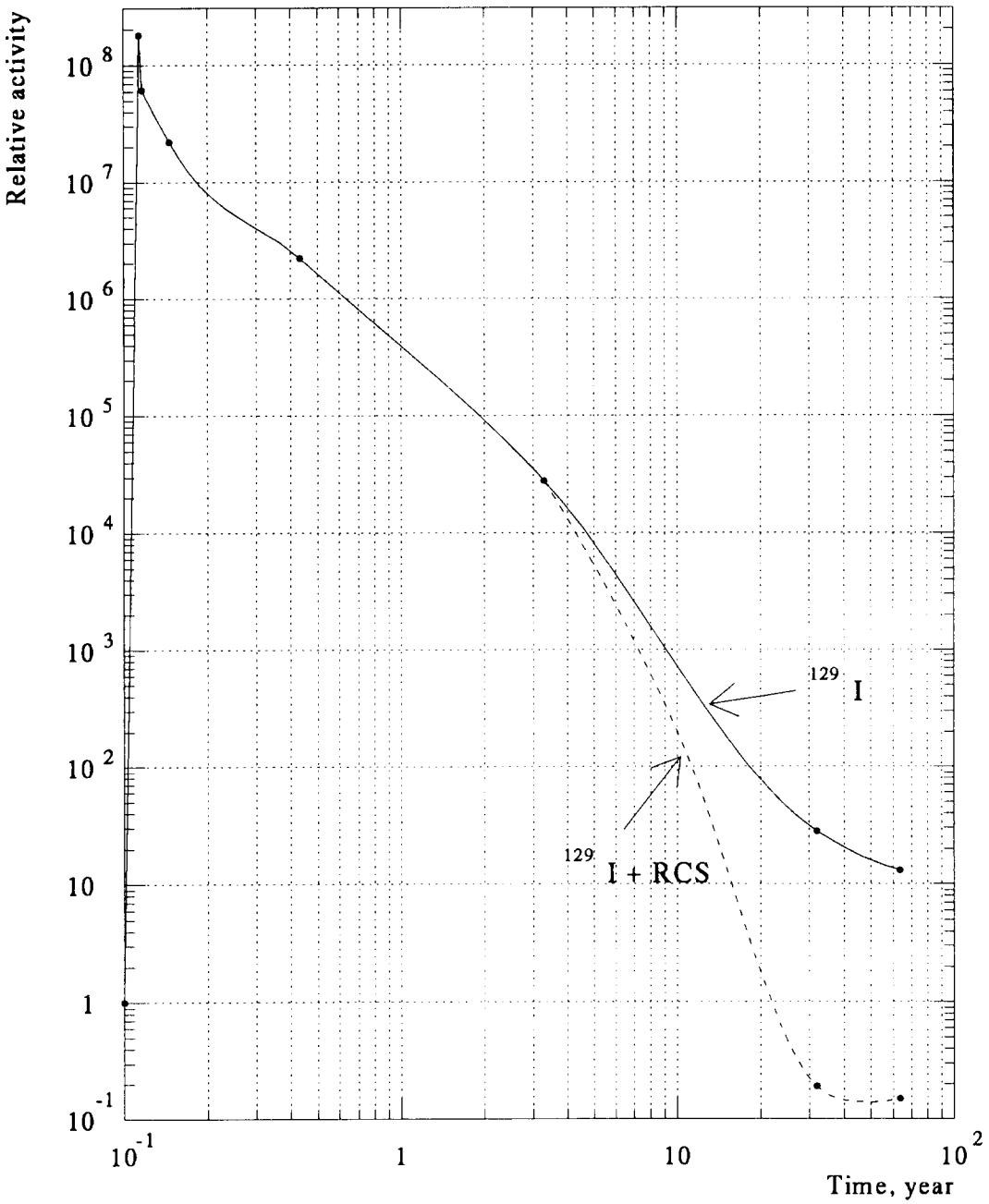

Figure 2: **Time dependence of the relative activities of ^{129}I target (RCS – radiochemical separation).**

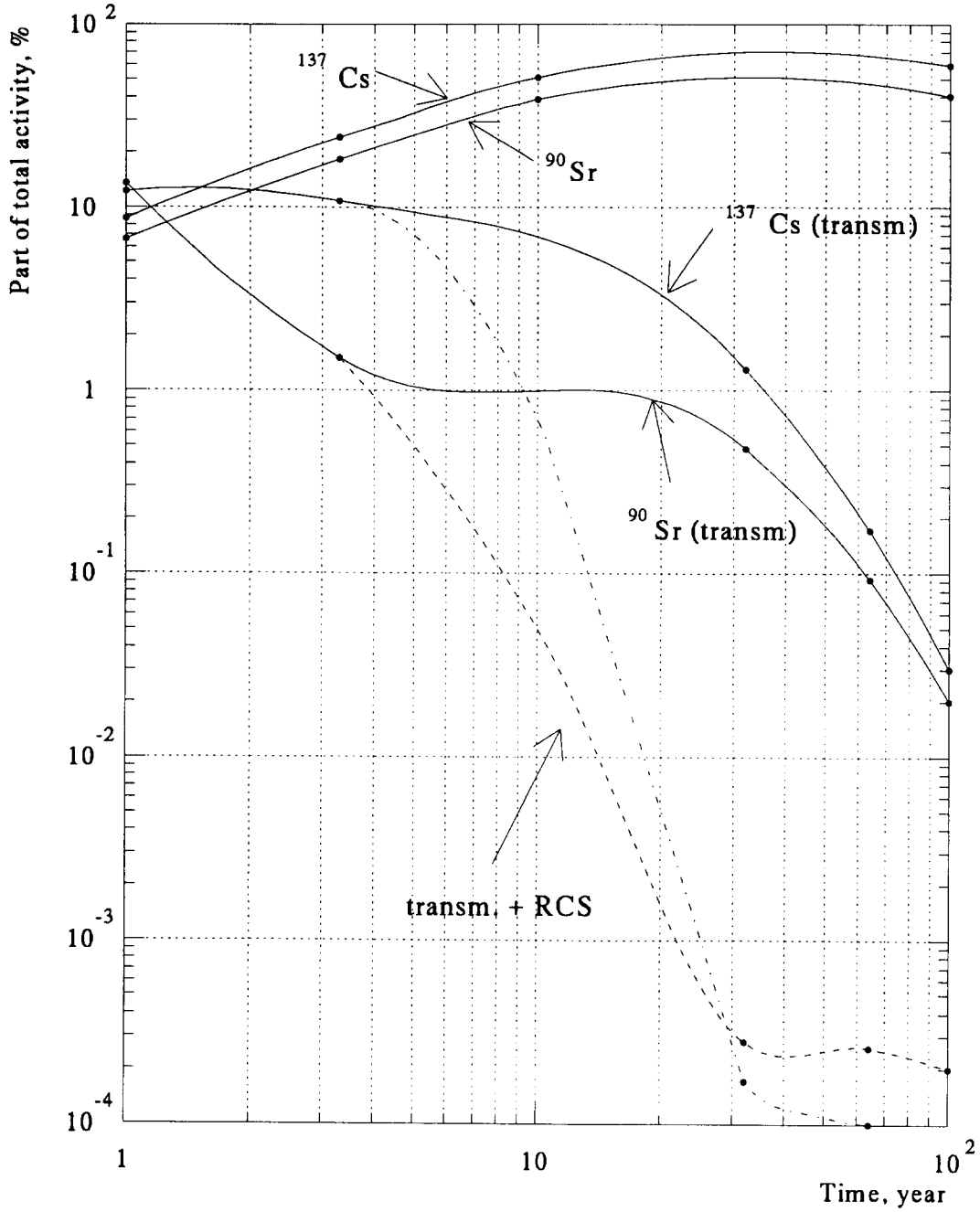

Figure 3: **Time dependence of activity contributed with ^{90}Sr and ^{137}Cs in the total activity of fission products (RCS – radiochemical separation).**