PRINCIPLE OF NATURAL AND ARTIFICIAL RADIOACTIVE SERIES EQUIVALENCY

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Abstract

In present paper one approach used under development of radioactive waste management conception is under consideration. This approach is based on the principle of natural and artificial radioactive series radiotoxic equivalency. The radioactivity of natural and artificial radioactive series has been calculated for 10^9- years period. The toxicity evaluation for natural and artificial series has also been made. The correlation between natural radioactive series and their predecessors - actinides produced in thermal and fast reactors has been considered. It has been shown that systematized reactor series data had the great scientific significance and the principle of differential calculation of radiotoxicity was necessary to realize long-lived radioactive waste and uranium and thorium ore radiotoxicity equivalency conception. The calculations show the execution of equivalency principle is possible for uranium series (4n + 2, 4n + 1). It is problem for thorium series. This principle is impracticable for neptunium series.

1. INTRODUCTION

Great quantities of radioactive waste connected with nuclear power engineering are accumulated in the world. They require utilization to provide the radiate safe for population. There are different ways to solve the problem connected with the decrease of nuclear power engineering waste radiotoxicity. The waste management conceptions are developed.

In present paper one approach used under development of radioactive waste management conception is under consideration. This approach is based on the principle of natural and artificial radioactive series radiotoxic equivalency. The authors try to estimate the possibility of this principle realization for the different radioactive series.

2. RADIOTOXICITY EQUIVALENCY PRINCIPLE

2.1. Natural radioactive series

The basic natural radioactive elements are included into four radioactive series as shown in Table I. These are: thorium series, neptunium series, uranium series and uranium-actinium series.

All of radioactive series articles are bond by irreversible reciprocal transformations. Therefore closed systems reach the equilibrium during the definite period.

Natural series contains the radioactive elements accumulated during millions of years. Great quantities of the radioactive daughter isotopes are in equilibrium with uranium and thorium. The series elements leave the system thanks to geochemical processes, difference of physical and chemical properties and miming [1]. Thus the system may be opened. It causes the breach of radioactive equilibrium. Therefore it is necessary to know the degree and time of system open state to estimate the series radioactivity.

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TABLE I. NATURAL RADIOACTIVE SERIES

Series	Mass changing	Radioactive parent isotope	Stable daughter isotope
Thorium	4n	^{232}Th ($T_{1/2}$ =1,405 × 10^{10} y)	^{208}Pb
Neptunium	4n + 1	^{237}Np ($T_{1/2}$ =2,14 × 10^{6} y)	^{209}Bi
Uranium	4n + 2	^{238}U ($T_{1/2}$ =4,47 × 10^{9} y)	^{206}Pb
Uranium-actinium	4n + 3	^{235}U ($T_{1/2}$ =7,04 × 10^{8} y)	^{207}Pb

2.2. Reactor radioactive series

Last 50 years the radioactive series are filled up by new transuranium elements.

Quantities of produced actinides increase. The safe management of long-lived radionuclides is one of the factors defining the rates of nuclear power engineering development. The reactor actinides are dangerous as radioisotopes of long standing. The activity dynamics of some isotopes is represented in Fig. 1-8. Discovered artificial actinides are predecessors for defined radioactive series [1,2]. Thus ^{248}Cm ($T_{1/2}$ = 3,39 • 10^{5} y) relates to the predecessors for thorium series; ^{245}Cm ($T_{1/2}$ = 8,5 • 10^{3} y) is predecessor for neptunium series; ^{246}Cm ($T_{1/2}$ = 4,73 • 10^{3} y) and ^{250}Cm ($T_{1/2}$ = 6,9 • 10^{3} y) are predecessors for uranium series; ^{247}Cm ($T_{1/2}$ = 1,56 • 10^{7} y) is predecessor for uranium-actinium series.

The actinide production depends on correlation between fission and capture cross-section and neutron spectrum. Consequently it depends on fuel kind and reactor type [3]. Thus the thermal reactor

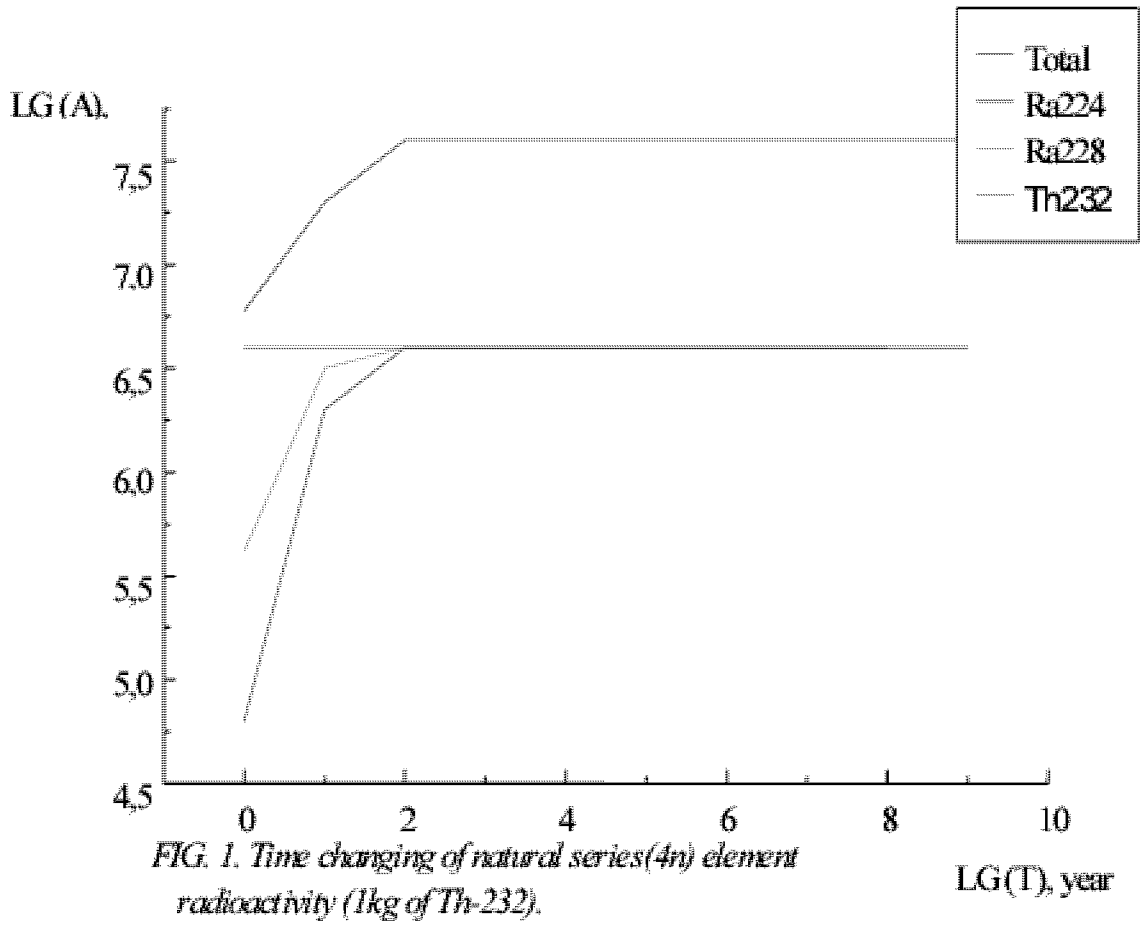

FIG. 1. Time changing of natural series(4n) element radioactivity (1kg of Th-232).

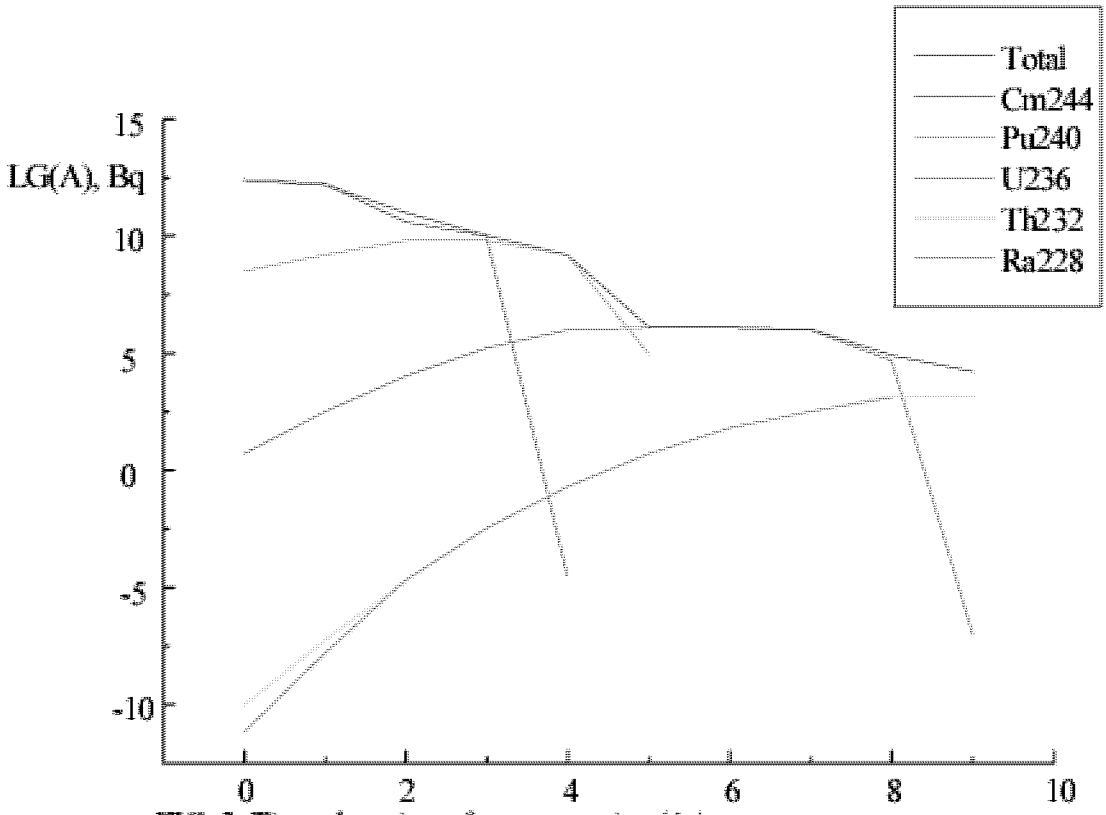

FIG. 2. Time changing of reactor series (4n) element radioactivity (1g of Cm-244).

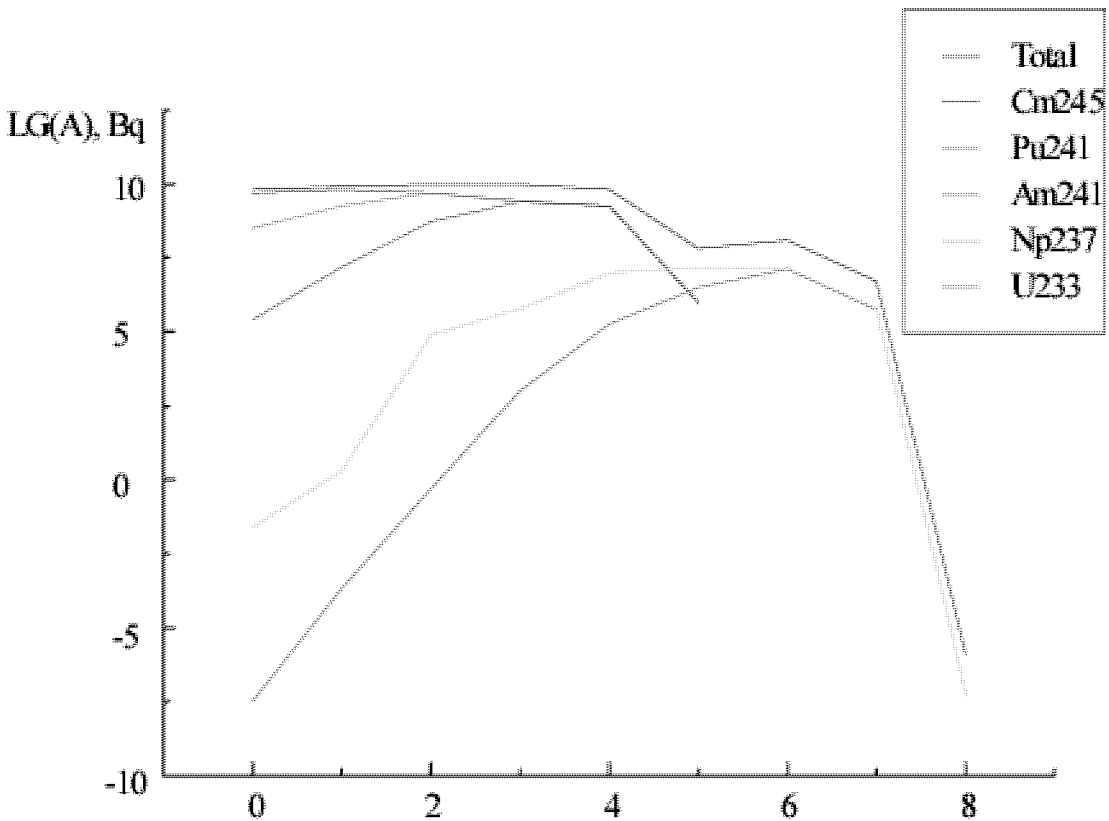

FIG. 3. Time changing of reactor series (4n + 1) element radioactivity (1g of Cm-245).

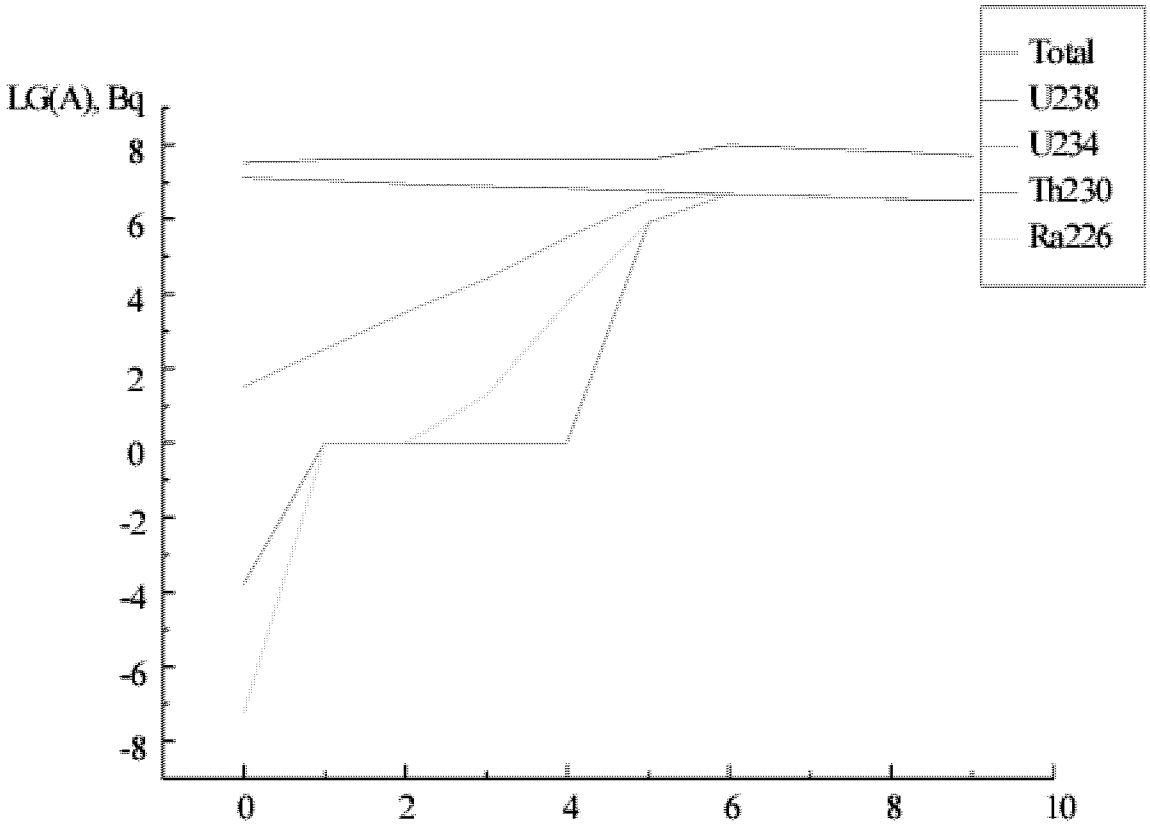

FIG. 4. Time changing of natural series (4n + 2) element radioactivity (1kg of U-238).

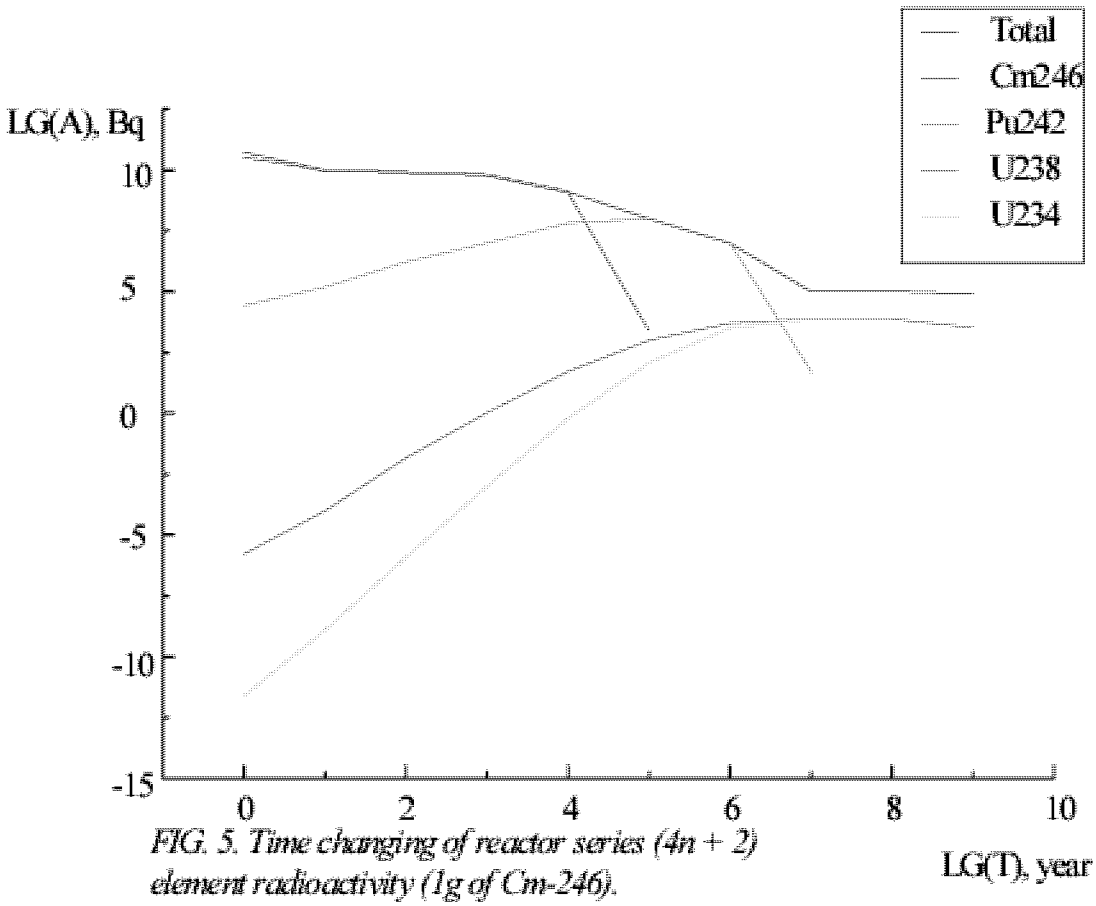

FIG. 5. Time changing of reactor series (4n + 2) element radioactivity (1g of Cm-246).

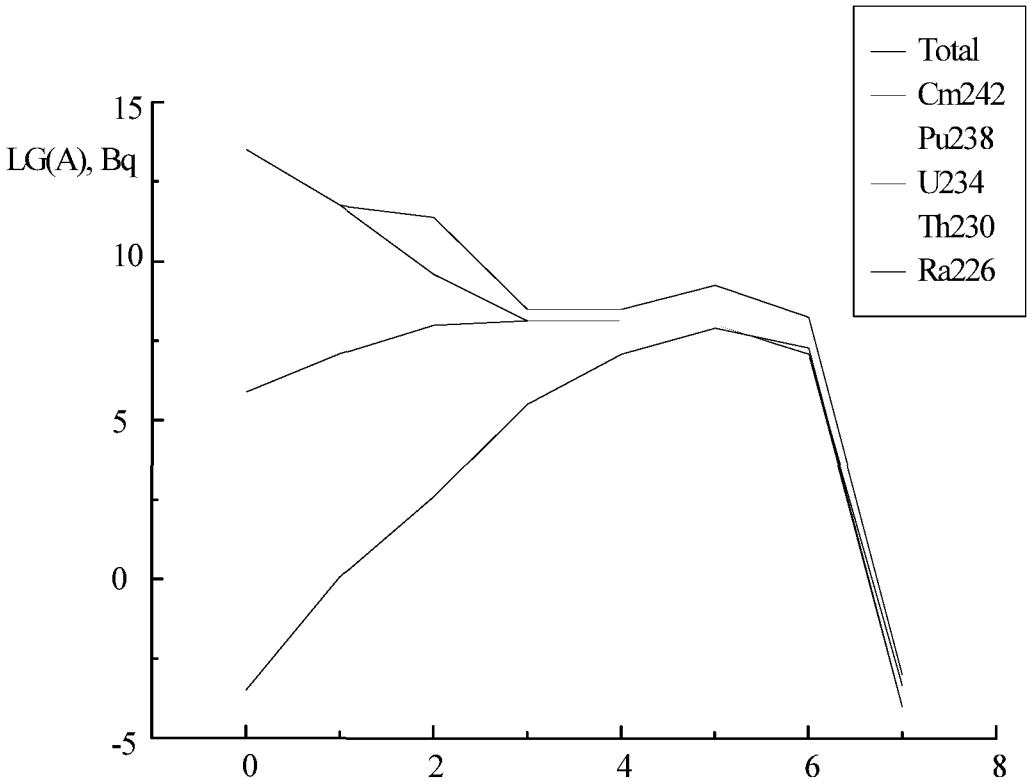

FIG. 6. Time changing of reactor series (4n + 2) element radioactivity (1g of Cm-242).

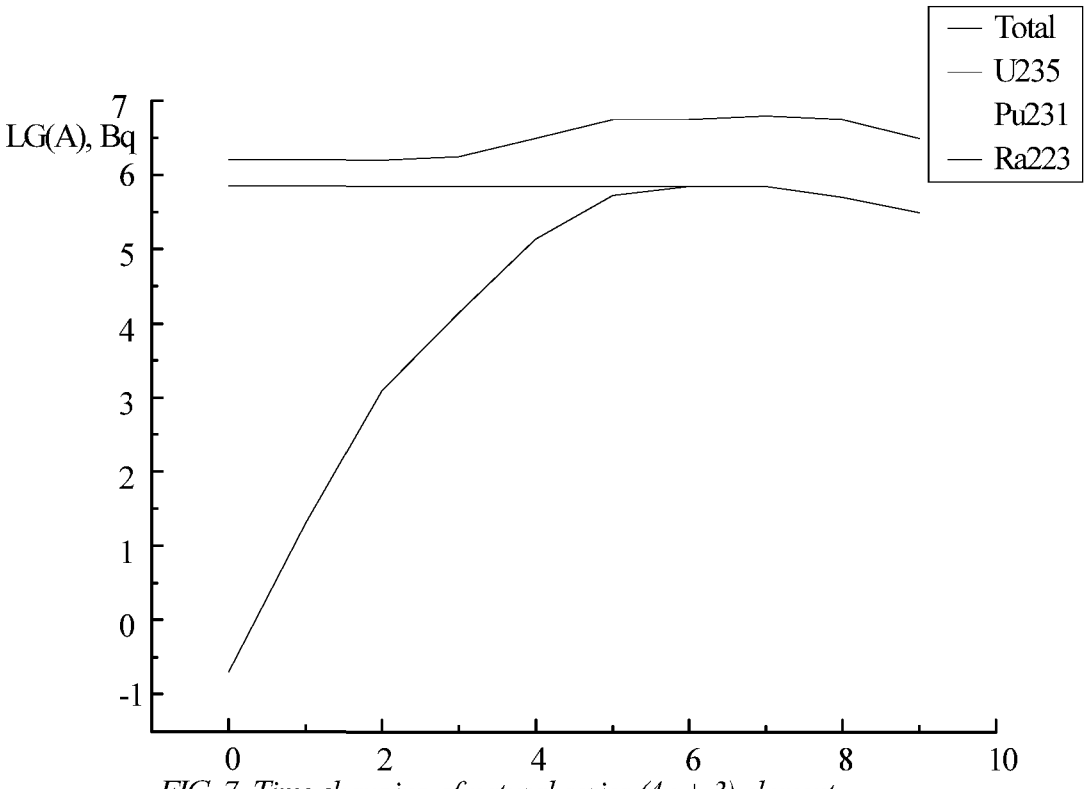

FIG. 7. Time changing of natural series (4n + 3) element radioactivity (1 kg of U-235).

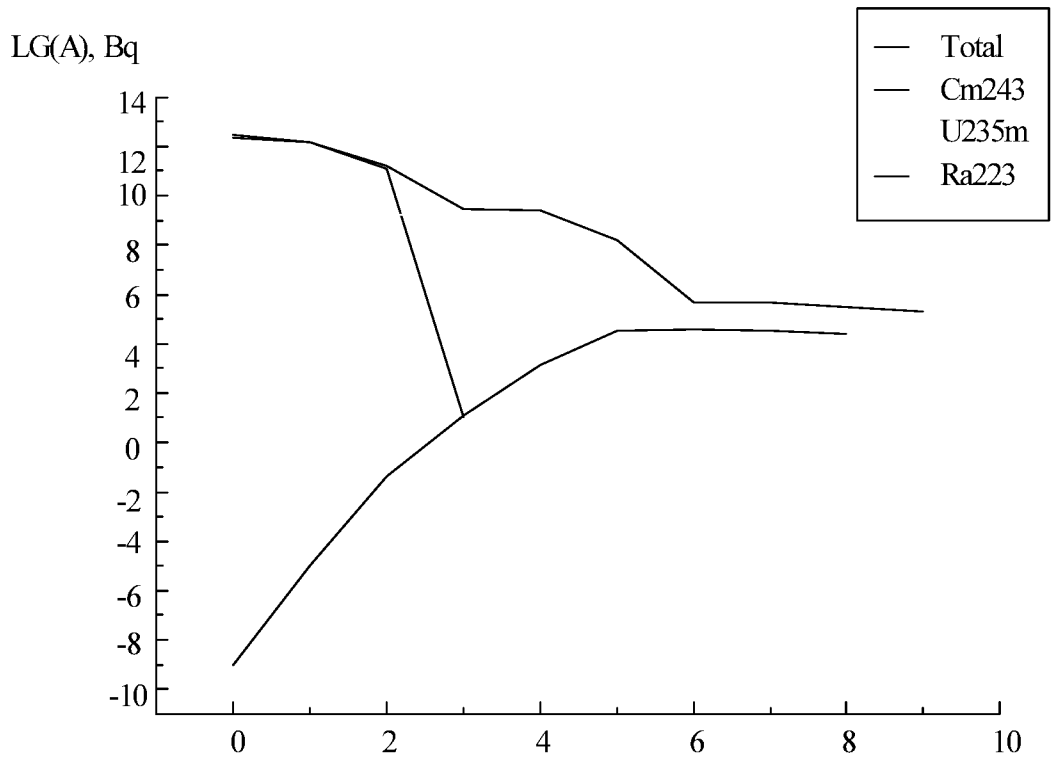

FIG. 8. Time changing of reactor series (4n + 3) element radioactivity (1g of Cm 243).

produces more neptunium series isotopes than the fast reactor (Table II). The quantity and correlation between actinides produced in reactor are defined by physical parameters of reactor, chemical and isotope composition of nuclear fuel, power and time regimes of exploitation and duration of fuel storage. Therefore the actinide spectra and their absolute value are different for the different fuel elements and reactor types. But there are some general regularities: the most quantity of produced isotopes is plutonium; neptunium and americium are produced less by one order than plutonium; curium isotopes are approximately produced less by two orders than plutonium [3]. Approximate actinide quantity submits to following correlation: Pu: (Np, Am): Cm ≈ 1: (0,1): 0,01 (Table III).

2.3. Radioactivity and radiotoxicity

The equivalency principle is one of the approaches under development of radioactive waste management conception.

The radioactive series elements emit the different articles (α,β) [4]. Part of decays accompanied by γ-quantum emission. Radioisotopes present the defined danger for health of person exposed to radioisotope radiation. This danger depends on the character of the isotope radiation.

The radiotoxicities of equal activity isotopes are different since the decay kind and radioactive decay energy is different (Table IV). Thus natural uranium-actinium series, taking the first radioactivity place, takes only third radiotoxicity place (Table IV). Series radiotoxicity is changing in the course of time because of the radionuclide decay.

The conception of differential radiotoxicity estimation is the base for realization above-mentioned principle.

To carry out this estimation it is necessary to know:

- Natural radioactive series radiotoxicity of closed balanced system;
- Natural radioactive series radiotoxicity of opened systems with the different opening times
  and degrees of it differentiation for separate or group of elements;
- Artificial radioactive series radiotoxicity of closed system, depending on the spectrum of irradiation neutrons;
- Artificial radioactive series radiotoxicity of open system, depending on time, opening regime and degree of fractionation.

The radioactivity of uranium ores related to 1 kg of natural uranium is four times as large than the radioactivity of thorium ores. The uranium ore radioactivities are changing little in the course of time as compared with maximum equilibrium quantity. Neptunium series is exception since it has zero radiotoxicity by present (Table IV). Therefore any measured quantity of neptunium series predecessors produced by reactor exceeds the natural series radiotoxicity (Tables IV, V) [3]. Reactor that does not produce neptunium series actinides is the ideal reactor to realize the equivalency principle.

TABLE II. THE CORRELATION BETWEEN RADIOACTIVE SERIES REACTOR ISOTOPES OF URANIUM FUEL (ATOMIC %) [3]

Series	Parent isotope	Reactors					Uranium daughter isotopes, ($T_{1/2}$)
		Thermal			Fast		
		PWR-800[a]	WWER-440[b]	WWER*[c]	BN-800[d]	BN-600[e]	
4n	^{240}Pu	20,3	20,65	22,67	27,31	3,5	^{236}U
	^{244}Cm	0,3	0,20	0,43	0,04	$3,6 \times 10^{-7}$	$(2,3 \times$
	Total	20,6	20,85	23,10	27,35	3,5	$\times 10^{7}$ y)
4n + 1	^{237}Np	3,8	2,75	2,99	0,14	1,32	^{233}U
	^{241}Pu	12,79	11,67	11,91	6,75	7×10^{-2}	$(1,6 \times$
	^{241}Am	2,0	2,07	2,82	1,46	1×10^{-2}	$\times 10^{6}$ y)
	^{245}Cm	$1,46 \times 10^{-2}$	$9,8 \bullet 10^{-3}$	$3,0 \times \times 10^{-3}$	$1,8 \times 10^{-3}$	$4,58 \times 10^{-9}$	
	Total	18,6	16,49	17,72	8,35	1,40	
4n + 2	^{238}Pu	1,69	0,7	1,18	0,42	0,1	^{238}U
	^{242}Pu	5,00	3,8	5,42	4,52	$1,3 \times 10^{-3}$	$(4,5 \times$
	^{242m}Am	$5,39 \times 10^{-3}$	-	$2,5 \times \times 10^{-3}$	$1,08 \times 10^{-2}$	$7,24 \times 10^{-6}$	$\times 10^{9}$ y)
	Total	6,69	4,5	6,6	4,95	0,1	
4n + 3	^{239}Pu	53,07	57,71	51,43	58,95	95,0	^{235}U
	^{239}Am	1,02	0,79	1,15	0,40	$1,63 \times 10^{-5}$	$(7,1 \times$
	^{243}Cm	$3,85 \times 10^{-3}$	-	$2,3 \times \times 10^{-3}$	$1,4 \times 10^{-3}$	$2,93 \times 10^{-7}$	$\times 10^{8}$ y)
	Total	54,09	58,50	52,58	59,35	95,0	

[a] 1 year after reactor operation.
[b] To 2000.
[c] After standing of 10 years.
[d] After standing of 3 years (3MO, U + Pu).
[e] After standing of 3 years (fuel element, U).

TABLE III. EXHAUSTED NUCLEAR FUEL TRANSURANIUM ISOTOPE COMPOSITION FOR THERMAL REACTOR

Series	Fraction, Atomic %	Isotopes	Fraction, atomic %
4n		^{240}Pu	10,49
(thorium)	31,19	^{244}Cm	17,85
		^{248}Cm	2,77
		^{252}Cf	0,08
4n + 1		^{237}Np	5,51
(neptunium)		^{241}Pu	9,48
	16,18	^{241}Am	0,54
		^{245}Cm	1,27
		^{249}Bk	0,05
		^{249}Cf	0,03
4n + 2		^{238}Pu	4,17
(uranium)		^{242}Pu	3,89
	20,00	^{242m}Am	0,02
		^{242}Cm	0,18
		^{246}Cm	11,71
		^{250}Cf	0,03
4n + 3		^{239}Pu	23,03
(uranium-actinium)		^{243}Am	8,11
	31,93	^{243}Cm	0,02
		^{247}Cm	0,75
		^{251}Cf	0,02
Total	100		100

TABLE IV. RADIOACTIVITY (A) AND RADIOTOXICITY (F [a]) OF NATURAL SERIES

Series	Parent isotope	Equilibrium establishment period, y	A, Bq/kg		F, L (H_2O)	
4n	^{232}Th	5×10^3	$4,1 \times 10^7$	(3)[b]	$7,37 \times 10^6$	(2)
4n + 1	^{237}Np	1×10^6	0	(4)	0	(4)
4n + 2	^{238}U	1×10^7	$1,6 \times 10^8$	(2)	$2,5 \times 10^7$	(1)
4n + 3	^{235}U	5×10^6	$3,7 \times 10^8$	(1)	$6,3 \times 10^6$	(3)

[a] Radiotoxicity is radioactivity divided by limited permissible concentration.
[b] Radiotoxicity place of series.

TABLE V. RADIOACTIVITY (A) AND RADIOTOXICITY (F [a]) OF REACTOR SERIES

Series	Parent isotope	1 year of standing		10^3 years of standing	
		A, Bq/g	F, L (H_2O)	A, Bq/g	F, L (H_2O)
4n	^{244}Cm	$2,8 \times 10^{12}$	$2,16 \times 10^{10}$	$7,58 \times 10^{10}$	$9,32 \times 10^7$
4n + 1	^{245}Cm	$6,6 \times 10^9$	$1,01 \times 10^8$	$1,66 \times 10^{10}$	$1,64 \times 10^8$
4n + 2	^{242},	$2,6 \times 10^{13}$	$6,4 \times 10^{10}$	$4,73 \times 10^8$	$8,03 \times 10^6$
	^{246}Cm	$1,14 \times 10^{10}$	$1,7 \times 10^8$	$9,85 \times 10^9$	$1,48 \times 10^8$
4n + 3	^{243}Cm	$1,86 \times 10^{12}$	$2,02 \times 10^{10}$	$4,5 \times 10^9$	$2,75 \times 10^7$

[a] Radiotoxicity is radioactivity divided by limited permissible concentration.

TABLE VI. RADIOACTIVITY (A) OF REACTOR SERIES

Series	Parent isotope	Time of standing	
		1 year	10^3 years
		A, Bq/g	A, Bq/g
4n	^{244}Cm	$2{,}8 \times 10^{12}$	$7{,}58 \times 10^{10}$
4n + 1	^{245}Cm	$6{,}6 \times 10^{9}$	$1{,}66 \times 10^{10}$
4n + 2	^{242,}	$2{,}6 \times 10^{13}$	$4{,}73 \times 10^{8}$
	^{246}Cm	$1{,}14 \times 10^{10}$	$9{,}85 \times 10^{9}$
4n + 3	^{243}Cm	$1{,}86 \times 10^{12}$	$4{,}5 \times 10^{9}$

3. ESTIMATION OF POSSIBILITY OF EQUIVALENCY PRINCIPLE REALIZATION

Basing on the closed nuclear fuel cycle (it foresees uranium, plutonium and neptunium inclusion), it is possible to estimate the radioactivity of radioactive waste reactor series, taking into consideration their basic radioactive elements. For example, 1 g-samples of corresponding curium isotope of 1 and 10^3 years standing each were taken as the parent isotope base (Table VI). The calculations show (taking into consideration the relation between uranium and curium (1: 10^{-5})) natural and reactor uranium series (4n + 2, 4n + 1) radioactivities are commensurable and the execution of equivalency principle is possible for these series (compare Tables IV and VI). It is possible to realize this principle for thorium series in tens of thousands of storage years.

The equivalency principle is impracticable for neptunium series since natural series has practically the background radioactivity and reactor neptunium series is increasing its radioactivity during hundreds of thousands of years (Fig. 3).

4. CONCLUSION

Systematized reactor series data have a great scientific significance. It is necessary to take into consideration the generality and correlation between series radioactive elements.

Proposed conception of differential estimation of series radioactivity for initial nuclear fuel and radioactive waste long-lived party (actinides) is useful for equivalency principle realization. As time is the technological parameter, it is necessary to take into consideration the opening degree of systems. It permits to calculate the exhausted nuclear fuel radioactivity; radioactivity of technological processes of exhausted nuclear fuel radiochemical treatment, and the radioactivity of radioactive waste immobilization products containing actinides.

The calculations show the execution of radioactivity equivalency principle is possible for uranium series (4n + 2, 4n + 1). It is a problem for thorium series. This principle is impracticable for neptunium series.

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