CREATION OF TRANSURANIUMS UNDER NEUTRON FLUXES OF NUCLEAR EXPLOSIONS

Yu. S. Lutostansky[1] and V. I. Lyashuk [2, 1, §]

[1]*National Research Center "Kurchatov Institute", Moscow, Russia*
[2]*Institute for Nuclear Researches of the Russian Academy of Sciences, Moscow, Russia*

Abstract: The artificial r(rapid)-process of nucleosynthesis goes under high neutron flux densities: the obtained neutron fluences in the irradiated volume of thermonuclear devices reach $\sim 10^{25}$ neutrons/cm^2 during the time interval $\sim 10^{-6}$ s. The extreme condition (in neutron flux and temperature ($\sim 10^8$ °K) can be reached under conditions of (thermo)nuclear explosions. The creation of transuranium nuclides under pulsed neutron fluxes of thermonuclear explosions is investigated by means of dynamical model (as in the kinetic model of the astrophysical *r*-process) taking into account the time dependence of the external parameters and the processes accompanying the beta decays of neutron-rich nuclei. Time-dependent neutron fluxes in the interval $\sim 10^{-6}$ s were simulated within the framework of the developed adiabatic binary model (ABM). The beta-delayed processes are taken into account for isotope yields correction after the pulse neutron wave. Calculations of transuranium nuclides yields $Y(A)$ are made for five large scale explosion USA experiments ("Mike", "Anacostia", "Par", "Barbel" and "Vulcan" and it were obtained good or satisfied agreement. The corresponding root-mean-square deviations of the model yields compare to the experimental data (r.m.s.) are: 91% (for "Mike"); 70% ("Anacostia"); 33% ("Par"); 29% ("Barbel"); and 45% ("Vulcan"). An even-odd anomaly in the observed yields of transuraniums is explained by the predominant processes of decays after creation of neutron rich isotopes.

1. INTRODUCTION. MODEL OF ARTIFICIAL RAPID NYCLEOSYNTHESIS

In nature the creation of nuclides with mass A≈ 65–70 occurs in *r* (rapid) and *s* (slow) processes of (n,γ)-captures. The cosmological *r*-process taking place in supernova explosions [1] is really fast one compare to *s*-process (as at the helium and carbon burning in red giants): so, the corresponding *r*-duration is up to $\sim 10^2$ s when the slow process – up to thousands of years. Duration of the artificial *r*-process considered in this article is determined by the very small mass of the explosives (compare to stars) and as a result is strongly shorter (than cosmological one) with the time length $\sim 10^{-6}$s. An artificial *r*-process under the explosions of (thermo)nuclear devices goes intensively in the specially constructed target installed within the volume of the exploding experimental installation. For successful results (i.e., creation of the most heaviest elements by (n,γ)-captures) the target is manufactured from uranium or transuranium isotopes. The most full and significant results were obtained with ^{238}U-target.

Studies of the transuranium nuclei formation under thermonuclear tests were widely carried out in the USA (starting from 1952 year when it was realized the thermonuclear test "Mike") in (thermo)nuclear explosions. Namely in air debris of "Mike" it were first detected the transuranium isotopes (up to ^{255}Fm) [2, 3]. Later for peaceful use of nuclear explosions and scientific purposes it were proposed the USA program “Plowshare” [4] which were

§ lyashuk@itep.ru

continued from 1958 to 1975 years. In the frame of the Plowshare program it were conducted 27 (thermo)nuclear experiments. Specifications of some experiments (in which the transuranium isotopes were detected) are indicated in the table 1.

Table 1. Experiments on production of transuranium isotopes.

Test (experiment)	**Date**	**released energy**, kt	**Obtained neutron fluence**, neutron/cm^2	**irradiated target**	**detected isotopes up to *A*-mass (and not detected *A*-mass)**
"Mike" [2,3]	October 31, 1952	10400	$(1.2–1.8)\cdot10^{24}$	^{238}U	255 (250, 251)
"Anacostia" [5]	November 27, 1962	5.2	$1.8\cdot10^{24}$	^{238}U	254 (241, 247–251, 253)
"Kennebec" [6]	June 25, 1963	< 20	$(2.7–3.6)\cdot10^{24}$	^{238}U	–
"Par" [7]	October 9, 1964	30 [7], 38 [6]	$(4.2–4.8)\cdot10^{24}$ [7] $6.6\cdot10^{24}$ [6,8]	^{238}U	257
"Barbel" [9]	October 16, 1964	20 [9], <20 [6]	$6.6\cdot10^{24}$ [6,8]	^{238}U	257 (not measured: 251 and 256) [9]
"Tweed" [6,8]	May 21, 1965	< 20 [6]	$7.2\cdot10^{24}$ [6,8]	^{242}Pu and small part of ^{237}Np	255
"Cyclamen" [6,8]	May 5, 1966	12	$1.1\cdot10^{25}$	^{238}U and small part of ^{243}Am	257 (249, 256)
"Kankakee" [6,8,10]	May 15, 1966	(20-200)	$7.2\cdot10^{24}$	^{238}U	255 (249, 251)
"Vulcan" [6,8,10]	June 25, 1966	25	$7.2\cdot10^{24}$	^{238}U	257 (256)
"Hutch" [6,8,10,11]	July 16, 1969	(20-200)	$(2.1–2.4)\cdot10^{25}$	^{238}U and ^{232}Th	257 (249, 256)

The short duration of explosive nuclear process ($t < 10^{-6}$ s) allows to split it into two phases: neutron capturing process and the following decays of neutron-rich nuclei [12, 13]. Such a process can be called as "prompt rapid" or *pr*-process and solution of equations for the concentration $N_{A,Z}(t)$ of formed nuclei can be greatly simplified.

The significant part of Plowshare program was devoted to nucleosynthesis of thransuranium isotopes under neutron pulse of exploding devices. The complete analysis on identification of isotopes produced in underground tests is possible only after drilling from the surface to the zone of explosion to the produced cavity volume. In order to imagine the scale of drilling it is need to note that depth of drilling can be several hundred meters (up to about kilometer). So the "Hutch" device was exploded at the depth 600 m [8]. The process of debris recovery by drilling can take days: the first debris from "Hutch" was delivered to the laboratory after 7 days since the explosion. But an exceptions are possible as in case of "Cyclamen" test (when the debris were obtained within 24 hours) [8]. As a result the short lived isotopes decay before isotope analysis in the laboratory.

In Plowshare program the first experiment for transuraniums production was "Anacostia" [5] with strongly lower released energy yield (compare to "Mike") and significantly more poor list of created isotopes. The most complete data of transuraniums yields up to A = 257 were obtained in the "Par" experiment [7]. In the similar test "Barbel" test [9] a close fluence was achieved as in "Par", but yield of ^{257}Fm was lower. In “Cyclamen” test it was obtained more high fluence (~$1.1{\cdot}10^{24}$ neutron/cm^2) but without new isotopes creation. The more heavy shot “Vulcan” (compare to “Cyclamen”) was constructed as device-development experiment. But production of isotopes with A>257 were not detected too (see table 1). In the test “Hutch” it was ensured the record fluence (2.1–2.4)$\cdot10^{25}$ neutron/cm^2 but also without creation of isotopes with A>257.

The figure 1 shows the experimental data normalized on $Y(A_i)$ yields for five explosions “Mike" [2, 3], "Par" [7], Barbel" [9], “Anacostia” [5] and “Vulcan” [10]. The decreasing dependence of $Y(A)$ is fitted as follows:

$$Y(A)/\,Y(A_i) = \exp\{-\,b_i A + c_i\} \quad (1)$$

$$i = 1 \text{ (“Mike”) } A_1 = 239,\ b_1 = 1.570,\ c_1 = 375.491 \quad (1a)$$

$$i = 2 \text{ (“Barbel”) } A_2 = 244,\ b_2 = 1.395,\ c_2 = 340.584 \quad (1b)$$

$$i = 3 \text{ (“Par”) } A_3 = 245,\ b_3 = 1.388,\ c_3 = 341.015 \quad (1c)$$

$$i = 4 \text{ (“Anacostia”) } A_4 = 239,\ b_4 = 1.739,\ c_4 = 417.941 \quad (1d)$$

$$i = 5 \text{ (“Vulcan”) } A_5 = 244,\ b_5 = 1.325,\ c_5 = 370.441 \quad (1e)$$

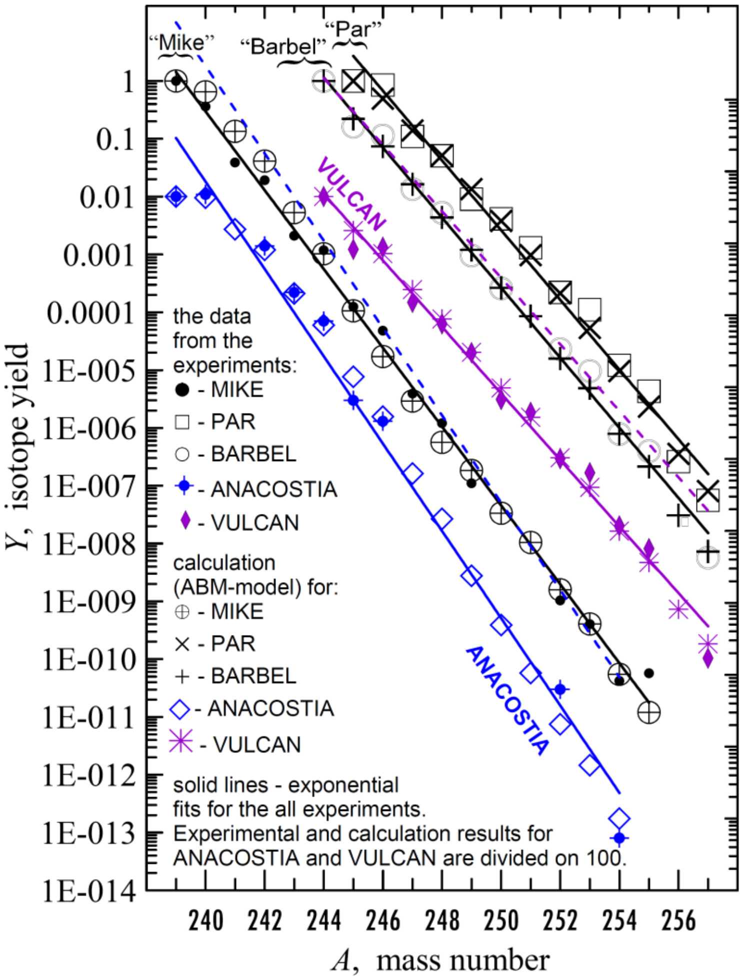

Figure 1. Results of isotope yield calculations in ABM-model compare to the experimental yields in tests "Mike", "Par", "Barbel", “Anacostia” and “Vulcan”. An exponential fitting of the experimental data are shown by the solid lines. The experimental data and calculation results for “Anacostia” and “Vulcan” are divided on 100. The dashed lines are exponential fitting for “Anacostia” and “Vulcan” without dividing on 100.

The standard deviations of this approximation are: δ_1 = 56% ("Mike"), δ_2 = 60.2% ("Barbel"), δ_3 = 86.8% ("Par"), δ_4 = 357% ("Anacostia"), δ_5 = 86.4% ("Vulcan"), which are comparable to the accuracy of the known calculations. The results of isotope yield calculations on the base ABM model (Adiabatic Binary Model [13, 12]) are presented in the figure 1 to compare to the experimental data and its exponential fitting (1, 1a-1e). The details of calculation for "Mike", "Barbel" and "Par" are discussed in [14, 15].

For the *pr*-process modeling of (thermo)nuclear explosions the serious simplification were made owing to the fact that neutron captures and decays of the nuclides are separated in time. So the system of equations for the time dependence of concentrations $N(A; Z; t)$ of nuclei with the mass number A and the charge Z have the form:

$$\mathrm{d}N(A, Z, t)/\mathrm{d}t = -\lambda_{n\gamma}(A, Z, t)\cdot N(A, Z, t) + \lambda_{n\gamma}(A-1, Z, t)\cdot N(A-1, Z, t) +$$
$$\lambda_{n,2n}(A+1, Z, t)\cdot N(A+1, Z, t) - \lambda_{n.2n}(A, Z, t)\cdot N(A, Z, t) - \lambda_{n.f}(A, Z, t)\cdot N(A, Z, t) -$$
$$\Phi[\lambda_{\beta}, \lambda_{\beta n}, \lambda_{\beta f}, \lambda_{\alpha}, \lambda_{sf}], \qquad (2)$$

where $\lambda_{n\gamma}$ – is the capture rate of neutrons in the (n, γ)-reaction, $\lambda_{n,2n}$ is the same for (n, 2n) reaction, and $\lambda_{n,f}$ is the neutron fission rate. The reactions with γ-quantum were not taken into account because of lower temperatures in comparison with astrophysical processes. The term $\Phi[\lambda_{\beta}, \lambda_{\beta n}, \lambda_{\beta f}, \lambda_{\alpha}, \lambda_{sf}]$ in the system of equations (2) does not depend on time since it includes the processes occurring after the active phase of the explosion: β-decay processes, (β,n)-emission of delayed neutrons (DN), α-decay, (β, f)-delayed (DF) and (s, f)-spontaneous fission. The DF and DN probabilities were calculated basing on the microscopic theory of finite Fermi systems [16]. The effect of the resonant structure of the β-decay strength function, including the pigmy resonances, was taken into account [17].

The time-dependent part of the system of equations (2) was solved using the adiabatic binary model (ABM) [18] where numerical simulation is performed by dividing duration of *pr*-process on small nanosecond time steps with calculations of isotope yields in succession for each step. The step initial conditions are also determined by the isotope composition of the target and yields of the preceding isotopes in the previous time step. In view of the binary, two-stage character of the thermonuclear explosion: the nuclear explosion (the first stage with the fission reaction) and the second stage associated with the thermonuclear reaction, two neutron fluxes and two sets of initial concentrations were used in the calculations.

2. RESULTS OF ISOTOPE YIELDS CALCULATIONS

For model calculations a unified approach was adopt within the framework of the adiabatic binary model (ABM) – it was assumed that there was an admixture of ^{239}Pu in the primary ^{238}U target. The specificity of the binary, two-stage explosion process also allowed to the irradiation of the uranium-plutonium target by two different fluxes. In accordance with the experimental data the all model yields of the isotopes $Y(A)_{\mathrm{calc}}$ are normalized (see (1)): note that for “Anacostia” the yields are normalized on $Y(A{=}239)$ which in [5] is considered as unit; for “Vulcan” the experimental yields for every A-value are absolute numbers of nuclei which produced from starting $3.4\cdot10^{22}$ nuclei of ^{238}U-target. The calculated yields and experimental data for “Anacostia” and “Vulcan” tests are presented in the table 2, where the standard (r.m.s.) deviations δ are also given for ABM calculations and for approximation (1).

Results for “Anacostia” are obtained at the conditions: ^{238}U (99.994% in the target) was irradiated with neutron fluence $1.3 \cdot 10^{24}$ neutron/cm^2; ^{239}Pu (0.006% in the target) was irradiated with fluence $2.7 \cdot 10^{24}$ neutron/cm^2.

Results for “Vulcan” are obtained at the conditions: ^{238}U (99.3% in the target) was irradiated with neutron fluence $4.21 \cdot 10^{24}$ neutron/cm^2; ^{239}Pu (0.7% in the target) was irradiated with fluence $7.55 \cdot 10^{24}$ neutron/cm^2.

Table 2. Experimental and calculated yields of isotopes in “Anacostia” and “Vulcan” tests.

"Anacostia"			"Vulcan"		
A	$Y(A)_{exper}$ [5]	$Y(A)_{calc}$	*A*	$Y(A)_{exper}$ [10]	$Y(A)_{calc}$
239	1±0,3	1.00	244	2.7E+20	2.56E+20
240	1,09±0,22	9.51E-1	245	3.3E+19	6.60E+19
241		2.71E-1	246	3.6E+19	2.64E+19
242	(1,4±0,2)E-1	1.20E-1	247	4.0E+18	6.30E+18
243	(2,2±0,1)E-2	2.12E-2	248	1.7E+18	1.95E+18
244	(7,0±0,4)E-3	5.95E-3	249	5.4E+17	5.16E+17
245	(3,0±0,4)E-4	7.64E-4	250	8.4E+16	1.26E+17
246	(1,3±0,2)E-4	1.56E-4	251	≤5E+16	3.88E+16
247	–	1.62E-5	252	7.9E+15	7.77E+15
248	–	2.65E-6	253	4.5E+15	2.39E+15
249	–	2.76E-7	254	5.4E+14	4.20E+14
250	–	3.87E-8	255	2.2E+14	1.20E+14
251	–	5.73E-3	256	–	1.86E+13
252	<3.E-9	7.45E-10	257	2.8E+12	4.69E+12
253	–	1.45E-10			
254	<8.E-12	1.75E-11			
δ %	357 (1d)	70		86 (1e)	45

To illustrate the degree of agreement between calculations and experiments "Anacostia" and "Vulcan" the calculated yields are normalized on experimental data (figures 2 and 3).

The table with experimental and calculated yields for "Mike", "Par" and "Barbel", the relations of normalized data ($Y_{calculation}$ / $Y_{experimental}$) and comparison with known simulation results are given in [14,15].

The most successful setup of nucleosynthesis experiment for production of transuraniums was the "Par" [7], where all nuclides with mass numbers up to A = 257 were detected. In the very similar test "Barbel" [9] the yields had considerable differences compare to "Par". The most high yields were obtained in the “Hutch” test where it was reached the record fluence $(2.1–2.4) \cdot 10^{25}$ neutron/cm^2. It is need to underline that the values of obtained fluences are strongly device-dependent ones.

The ABM model applied to yields simulation in several explosive experiments have demonstrated the adequate algorithm for describing of *pr*-process with good or (at least) satisfactory agreement.

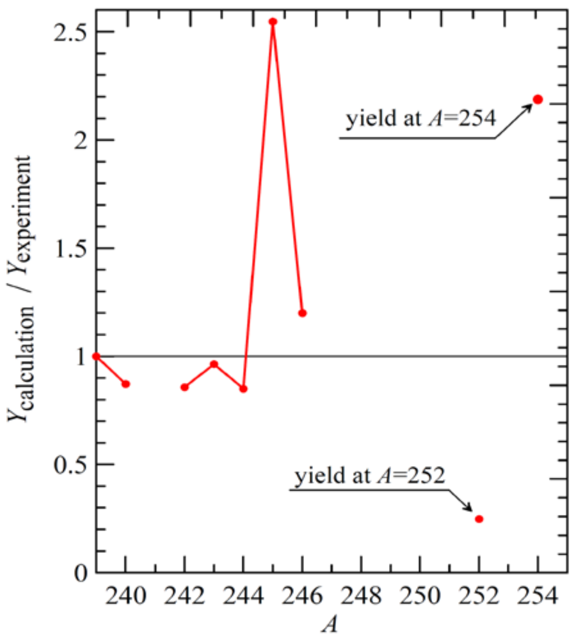

Figure 2. Relation of normalized (on $Y(A=243)_{calc}$) calculated yields to normalized (on $Y(A=243)_{exp}$) experimental yield of "Anacostia".

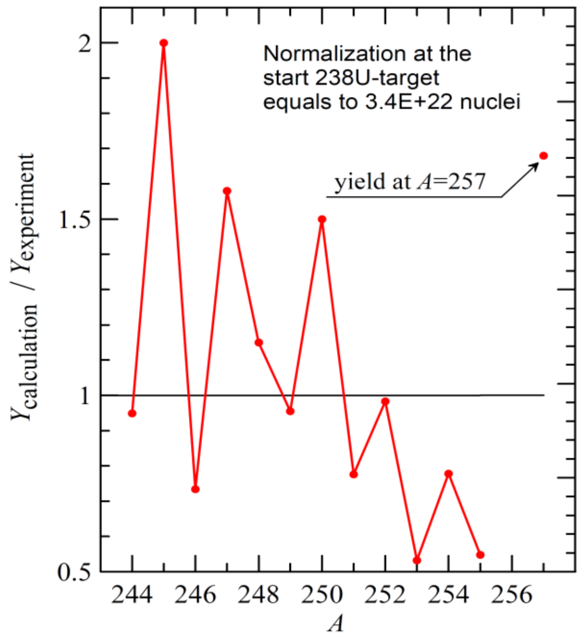

Figure 3. Relation of calculated yields to the experimental yield $Y(A=244)_{exp}$ (in number of nuclei) of "Vulcan" for ^{238}U-target (with number of nuclei 3.4E+22).

3. CONCLUSION

The article devoted to simulation of artificial *pr*-process of nucleosynthesis in which the neutron fluences reach the values up to ~10^{25} neutrons/cm^2 during the short exposition – about 10^{-6}s. The results of calculation on the base of the developed ABM model are compared with the experimental date for all mass numbers in the region A = 239 – 257. The Adiabatic Binary Model has confirmed the adequacy of used algorithm. The model is confirmed as in previous our calculations (the δ -values of standard r.m.s. [root mean square] deviation for “Mike”, “Par” and “Barbel” experiment are: 91%, 33% and 29% [14, 15]) as in the current results: δ –values for “Anacostia” and “Vulcan” – 70% and 45% correspondingly.

The calculations include the processes of delayed fission (DF) and the emission of delayed neutrons (DN), which determine the "losing factor" – the total loss of isotope concentration in the isobaric chains. The DF and DN probabilities were calculated in the microscopic theory of finite Fermi systems [8]. Thus, it was possible to describe the even-odd anomaly in the distribution of concentrations $N(A)$ in the mass number region A = 251 – 257. It is shown qualitatively also that the odd-even anomaly may be explained mainly by DF and DN processes in very neutron-rich uranium isotopes.

The results confirms that the assumptions [14, 15] (**1.** emission of other isotopes [^{239}Pu in our calculations] into the target volume at stage of plasma ball and **2.** irradiation of admixture [^{239}Pu in this case] and the main target under different fluences at the stage of plasma ball) are reasonable.

Acknowledgements

We are grateful to L.B. Bezrukov, B.K. Lubsandorzhiev, I.V. Panov, E.E., V.N. Tikhonov, I.I. Tkachev and S.V. Tolokonnikov for stimulating discussions and assistance in the work. The work is supported by the Russian Foundation for Basic Research Grants no.18-02-00670_a.

References

1. Burbridge B. M., Burbridge G., Fowler W., Hoyle P., *Rev. Mod. Phys.* **29** (1957) 547.
2. Ghiorso A., Thompson S. G., Higgins G. H. *et al.*, *Phys. Rev.* **99** (1955) 1048.
3. Diamond H., Fields P. R., Stevens C. S. *et al.*, *Phys. Rev.* **119** (1960) 2000.
4. United States Nuclear Tests. July 1945 through September 1992. DOE/NV-209-REV 16 September 2015.
5. Hoff R.W., Dorn D.W., *Production of Transuranium Elements in a Thermonuclear Explosion – Anacostia. Nuclear Science and Engineering*, **18**, (1964) 110–112.
6. Becker, Stephen A., *Approximating the r-Process on Earth with Thermonuclear Explosions*. Lessons Learned and Unanswered Questions (2012). LA-UR-12-25146
7. Dorn D.W., Hoff R.W., *Spontaneous fission in very neutron-rich isotopes*, Phys. Rev. Lett. **14** (1965) 440.
8. Hoff. R.W., *Production einsteinium and fermium in nuclear explosions*. Lawrence Livermore Laboratory. UCRL-81566. August 21, 1978.
9. Los Alamos Radiohemistry Group. *Production of very heavy elements in thermonuclear explosion - test Barbel*. Phys. Rev. Lett.**14** (1965) 962–964.
10. John S. Ingley, Nucl. Phys. A **124** (1969) 130–144.
11. Hoff. Richard W., *Beta decay of neutron-rich transuranic nuclei*. UCRL-94252. June 6, 1986.
12. Lutostansky Yu.S., Lyashuk V.I., Panov I.V., *Calculation of Transuranium Element Synthesis in Intensive Neutron Fluxes under Adiabatic Conditions.* Bull. Russ. Acad. Sci. Phys. **74** (2010) 504.
13. Lyashuk V.I., *Taking into consideration the dynamics at creation of transuranium isotopes*. Preprint of of Alikhanov Institute for Theoretical and Experimental Physics, Moscow: ITEP, 1997, no.7; http://lss.fnal.gov/archive/other/itep-7-97.pdf
14. Lutostansky Yu.S., Lyashuk V.I., *Production of Transuranium Nuclides in Pulsed Neutron Fluxes from Thermonuclear Explosions*. JETP Letters, **107**, No.2 (2018) 79–85.
15. Lutostansky Yu.S., Lyashuk V.I., *Nucleosynthesis of Heavy Elements in Thermonuclear Explosions "Mike", "Par" and "Barbell"*, in *the 3rd International Conference on Particle Physics and Astrophysics*, KnE Energy & Physics, pages 57–64, DOI 10.18502/ken.v3i1.1723.
16. Migdal A.B., *Theory of Finite Fermi Systems and Applications to Atomic Nuclei* (1983) Nauka Moscow; 1967 Inter-Sci. New York.
17. Lutostansky Yu.S., JETP Lett. **106** (2017) 7.
18. Lyashuk V.I., *Simulating Transuranium Isotope Yields upon Explosive Nucleosynthesis with Allowance for Elements of Process Dynamics*, Bull. Russ. Acad. Sci. Phys. **76** (2012) 1182.