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TAKING INTO CONSIDERATION THE DYNAMICS AT CREATION OF TRANSURANIUM ISOTOPES UNDER THE CONDITIONS OF NUCLEAR EXPLOSION

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The creation of transuranium isotopes under the conditions of nuclear explosions (i. e. nucleosynthesis at pulsed intensive fluxes) is considered. The model description of multiple neutron captures takes into account variation of (n,γ)-reaction cross sections at the adiabatic expansion of a space (where the explosive nucleosynthesis process goes) during the time interval of multiple neutron captures $t \leq 10^{-6}$ c. The calculations performed for nuclear explosion "Par" show that the model taking into account adiabaticness allows to approach model results to experimental data on transuranium isotopes yields in the wide interval of mass number A=248÷257.

Fig. - 1, ref. - 15 name.

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For the first time the creation of transuranium isotopes up to ^{255}U was performed in the thermonuclear experiment "Mike" [1]. Following the Plowshare project (USA) the series of experiments (Anacostia, Par, Barbel, Tweed, Cyclamen, Kankakee, Vulcan, Hutch and other) on investigation of heavy isotopes creation were performed (see e.g. Ref. 2). The isotopes up to A=257 were registrated in the experiment "Par" and "Barbel" what is implies creation of transuranium isotopes up to ^{257}U.

Let us consider the creation of transuranium isotopes under the conditions of an artificial powerful pulsed nucleosynthesis, i.e. under the conditions of nuclear explosions. The general scheme of transuranium elements creation at nuclear explosion is described by equations in the form:

$$\begin{aligned}\frac{\partial N_z^n}{\partial t} &= (\lambda_\beta N)_{z-1}^{n+1} + (\lambda_\alpha N)_{z+2}^{n+2} + \\ &+ \int_0^\infty F(E,t)\left\{\left[\sigma_{n,\gamma} N\right]_z^{n-1} + \left[\sigma_{n,2n} N\right]_z^{n+1} + \left[\sigma_{n,3n} N\right]_z^{n+2}\right\} dE - \\ &- (\lambda_\beta N)_z^n - (\lambda_\alpha N)_z^n - (\lambda_f N)_z^n - \\ &- \int_0^\infty F(E,t)\left\{\left[\sigma_{n,\gamma} N\right]_z^n + \left[\sigma_{n,2n} N\right]_z^n + \left[\sigma_{n,3n} N\right]_z^n + \left[\sigma_{n,f} N\right]_z^n\right\} dE,\end{aligned} \tag{1}$$

where z and n - charge and number of neutrons of the nucleus at which the cosidered reaction occurs; λ_β, λ_α and λ_f - rate of β-, α-decays and

spontaneous fission; $\sigma_{n,\gamma}$, $\sigma_{n,2n}$, $\sigma_{n,3n}$, $\sigma_{n,f}$ - cross sections of the respective reactions; $F(E,t)$ - value of the flux.

The solving of this problem requires knowledge of reactions cross sections, rates of transuranium isotopes decays and it is necessary especially to know these values for neutron-rich isotopes of the starting element (or combination of several starting isotopes). It is necessary to know temporary dependence for an energy spectrum of neutrons. Basically the values of these nuclear constants are unknown.

Taking into account the features of an explosive nucleosynthesis admits simplification of an explosive nucleosynthesis model. The evolution of the chain reaction occurs in the time $\approx 10^{-7}$ *c* [3], duration of multiple neutron captures does not exceed 10^{-6} *c* [4, 5]. The rates of decays λ_β, λ_α и λ_f are much less than $\lambda_{n,\gamma}$, so the contributions of β–, α–decays and spontaneous fissions in the equation (1) can be neglected. Further, it is possible to neglect the reactions (*n*, *f*), (*n*, *2n*) and especially (*n*, *3n*) as the rate of (*n*, γ)-capture becomes significant at the energy below of energy thresholds of the given reactions. So, for the starting isotope of ^{238}U the thresholds for these reactions are following: $E_{n,f}^{threshold} = 0.42$ MeV, $E_{n,2n}^{threshold} = 6$ MeV [6].

As far as the function of a neutron source $F(E,t)$ in the given experiments is unknown then expediently to perform the convolution for a time and energy:

$$\int_0^{\Delta t}\int_E F(E,t)\,dE\,dt \doteq \Delta t \int_E \tilde{F}(E)\,dE = \Phi \quad , \qquad (2)$$

where Δt - the exposition time.

Hereinafter, in the calculation it is used the neutron flux $\Phi[neutron / cm^2]$ integrated for a time and possessed the fixed energy in the

interval ≈20÷30 keV (i.e. one-group energy representation) according to the process temperature .

Then, in the given static model the equations system of transuranium elements creation (generated by equations (1)) becomes one-group and takes the form:

$$\begin{cases} \dfrac{\partial N_z^n}{\partial t} = (\lambda_{n,\gamma} N)_z^n \\ \dfrac{\partial N_z^{n+1}}{\partial t} = (\lambda_{n,\gamma} N)_z^n - (\lambda_{n,\gamma} N)_z^{n+1} \\ \cdot \\ \cdot \\ \cdot \\ \dfrac{\partial N_z^{n+i}}{\partial t} = (\lambda_{n,\gamma} N)_z^{n-1+i} - (\lambda_{n,\gamma} N)_z^{n+i} \end{cases} \qquad (3)$$

I.e. the given stage of the modeling is reduced to calculation of multiple neutron capture reactions.

The similar systems have the known solution for the *i*-th isotope at the single starting isotope (*z*, *n*) [2]:

$$N_z^{n+i} = \lambda_{n,\gamma}^n \lambda_{n,\gamma}^{n+1} \dots \lambda_{n,\gamma}^{n+i-1} N_z^n(0) \sum_{k=1}^{i} \frac{exp(-\lambda_{n,\gamma}^k t)}{\prod_{j \neq k} (\lambda_{n,\gamma}^j - \lambda_{n,\gamma}^k)}, \qquad (4)$$

where $N_z^n(0)$ - number of nuclei of the starting isotope at $t = 0$; $\lambda_{n,\gamma}^{n+i}$ - the rate of (n,γ)- reaction for the isotope $(z, n+i)$; $\prod_{j \neq k}$ - product of all combinations $(\lambda_{n,\gamma}^j - \lambda_{n,\gamma}^k)$, excepting $j = k$.

At calculation of the radiative capture rate $\lambda_{n,\gamma}^{n+i}$ the cross section $\sigma_{n,\gamma}(A+i, Z)$ for neutron-rich isotopes is extrapolated as follows. It was

assumed that variation of cross sections ralative to the known cross section $\sigma_{n,\gamma}(A,Z)$ of the previous isotope is proportional to the variation of binding energy for neutron:

$$\sigma_{n,\gamma}(A+i,Z)=\frac{B_n(A+i+1,Z)}{B_n(A+1,Z)}\sigma_{n,\gamma}(A,Z)\,, \qquad (5)$$

где B_n- binding energy of neutron in the $(A+1,Z)$ and $(A+i+1,Z)$ compound nuclei for the (n,γ)- reaction with known and calculated cross sections, respectively.

The cross sections for even and odd nuclei are calculated separately. At the even mass number $(A+i)$ the cross section $\sigma_{n,\gamma}(A+i,Z)$ is calculated on the basis of reliable data ($\sigma_{n,\gamma}(A,Z)$ and $B_n(A+1,Z)$) of the A-even isotope. At $(A+i)$-odd it is used the reliable data for A-odd isotope.

Let us note that in the the given scheme the exposition time Δt (see the expression (2)) of the target (i.e.some mass of the starting isotope placed in an exploded device with the purpose of transuranium isotopes creation) is excluded from calculations (that is quite justified as the nucleosynthesis rate is much more than decay rates of creating isotopes). As a result the product of the neutron flux (integrated with respect to time) into the cross section enter to the exponent of the expression (4):

$$\lambda^k_{n,\gamma}\,t=\Phi\sigma^k_{n,\gamma}\quad, \qquad (6)$$

where the upper index k in a cross section similarly means number of neutrons in a nucleus.

The results of calculations for transuranium isotopes creation in the experiments "Par" and "Barbel" are presented in Ref. [7,8]. In these explosions the starting isotope was ^{238}U. The multiple neutron captures

calculations were performed being based on the cross sections $\sigma_{n,\gamma}$ for ^{235}U and ^{238}U (data of the library *ENDL-83* [9]) and using the binding energy of neutron from Ref. [10]. For "Barbel" the second variant of the pair (as reference cross sections) - ^{239}U and ^{240}U [9] (as more detailed and complete files) was considered in addition. But use of ^{239}U and ^{240}U cross sections does not give the more good agreement with the experiment and in all probability one of the reasons is a large reliability of the data for ^{235}U and ^{238}U isotopes. The data analysis is complicated by absence of the experimental information about the integral flux value that results in significant spread in evaluation of the flux at these experiments modeling. So two different flux evaluations are presented in Ref. [11] and [12]: $(4.2 \div 4.8) \cdot 10^{24}$ and $7 \cdot 10^{24}$ *neutron/cm*^{2}, respectively. Flux decreasing gives improvement of agreement with experiments at the mass number $A < 250$ but at $A>250$ yields decrease nonlinear and rapidly. Taking into account multiple capture only the yield calculations (in the $A \geq 250$ range) give characteristic sawtooth dependence with the steady tendency to overestimation for $A=252, 254, 256$ and underestimation for odd isotopes. We emphasize especially that only multiple neutron captures are discussed in the given work as distinct from Ref. [7,8] where the subsequent processes redistributing the isotope yields after multiple neutron captures are considered also: delayed fission (i.e. (β^-, f)–process) and delayed neutrons emission (i.e. (β^-, n)–process).

These calculations were performed at characteristic temperature of the process - 30 keV. The distribution of transuranium isotopes (after multiple neutrons captures) is stable and it principally does not vary with respect to the shape at variations of the mean process temperature (that determines the (n, γ)-cross section) in the temperatures interval $T=20 \div 30$ keV . The increase of the mean temperature T in this interval results in lowering of the graph

without essential changes in its sawtooth shape. The processing of results with various paired variants of reference isotopes (^{235}U, ^{237}U, ^{238}U, ^{239}U, ^{240}U) show the stability of the given model.

The discussed model description of the transuranium isotopes creation at multiple neutron captures under the conditions of a pulsed nucleosynthesis can be extended by consideration of the process dynamics taking into account variations of (n,γ)-cross sections at fall of the medium temperature T during an adiabatic expansion (i.e. after termination of the chain reaction). It is possible to determine crudelly the functional dependence of the temperature for the considered volume (including the target made from the starting isotope ^{238}U) at adiabatic expansion if: 1) to specify the interval $T_1 \div T_2$ of the fall for the mean energy of captured neutrons (i.e. description by one-group energy of neutrons) during the medium cooling due to adiabatic expansion for the time interval $t_A \div t_B$; 2) to assume that a linear speed of an explosive expansion of a matter $\vartheta = const$ at $t \in [t_A, t_B]$ and; 3) to specify the adiabat index γ at adiabatic expansion of the volume V:

$$T = \left(\frac{const}{V}\right)^{\gamma-1} \qquad (7)$$

In a given scheme the interval $T_1 \div T_2$ is parameter. This value can and must be tested with respect to the linear speed ϑ (of the explosive expansion of the matter) that is determined from the interval fall $T_1 \div T_2$ and taking into account the initial sizes R_0 of the volume with a fissile material [13]. The assumption about the constantness for the speed ϑ is justified with the low external opposite pressure p for the considered small time interval $[t_A, t_B]$ (see Ref. [14]).

In the model calculations it was assumed: the multiple captures proceed up to the time moment $t=10^{-6}$ s [4,5]; the chain reaction lasts $3 \cdot 10^{-7}$ s; the

volume with fissile matter has spherical shape with initial radius R_0=5 *cm*; the possible interval of the adiabat index variation γ =1.5÷2.0 [15]. The algorithm of the transuranium isotopes problem solving is reduced to the partition of the multiple capture time interval $[t_A, t_B]$ into *m* intervals and sequential solution of nucleosynthesis equations (3) for the each of these time steps $\Delta t_1, \Delta t_2, \ldots, \Delta t_m$ at the initial conditions: $N_z^n(t_A) = 1$ and $N_z^{n+i}(t_A) = 0$ — for the first step, i.e. the accumulation begins with the single starting isotope; for the subsequent steps the initial conditions for the all isotopes are nonzero and are equal to accumulation of these nuclides by the moment of the previous step ending. Under these conditions the system of equations (3) has the following solution for the interval $[t_1, t_2]$ at $t_1 > t_A$ and $t_2 \le t_B$:

$$N_z^n(t_2) = N_z^n(t_1)\exp(-\lambda^n \Delta t)$$

$$N_z^{n+1}(t_2) = \lambda^n N_z^n(t_1)\left(\frac{\exp(-\lambda^n \Delta t)}{\lambda^{n+1}-\lambda^n} + \frac{\exp(-\lambda^{n+1}\Delta t)}{\lambda^n-\lambda^{n+1}}\right) + N_z^{n+1}(t_1)\exp(-\lambda^{n+1}\Delta t)$$

$$N_z^{n+2}(t_2) = \lambda^n \lambda^{n+1} N_z^n(t_1)\left[\frac{\exp(-\lambda^n \Delta t)}{(\lambda^{n+1}-\lambda^n)(\lambda^{n+2}-\lambda^n)} + \frac{\exp(-\lambda^{n+1}\Delta t)}{(\lambda^n-\lambda^{n+1})(\lambda^{n+2}-\lambda^{n+1})} + \frac{\exp(-\lambda^{n+2}\Delta t)}{(\lambda^n-\lambda^{n+2})(\lambda^{n+1}-\lambda^{n+2})}\right] + \lambda^{n+1} N_z^{n+1}(t_1)\left[\frac{\exp(-\lambda^{n+1}\Delta t)}{\lambda^{n+2}-\lambda^{n+1}} + \frac{\exp(-\lambda^{n+2}\Delta t)}{\lambda^{n+1}-\lambda^{n+2}}\right] + N_z^{n+2}(t_1)\exp(-\lambda^{n+2}\Delta t)$$

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$$N_z^{n+i}(t_2) = \lambda^n \lambda^{n+1} \dots \lambda^{n+i-1} N_z^n(t_1) \sum_{k=1}^{i} \frac{exp(-\lambda^k \Delta t)}{\prod_{j \neq k}(\lambda^j - \lambda^k)} + \dots +$$

$$\lambda^{n+i-1} N_z^{n+i-1}(t_1) \left[\frac{exp(-\lambda^{n+i-1} \Delta t)}{\lambda^{n+i} - \lambda^{n+i-1}} + \frac{exp(-\lambda^{n+i} \Delta t)}{\lambda^{n+i-1} - \lambda^{n+i}} \right] +$$

$$N_z^{n+i}(t_1) exp(-\lambda^{n+i} \Delta t) \ , \qquad (8)$$

where $\Delta t = t_2 - t_1$.

At solving of the nucleosynthesis equations system (8) the (n,γ)-reaction rate λ is recalculated (for the each isotope at transition to the next time step) according to a mean energy decrease of the captured neutrons at a matter cooling during the step Δt.

In calculations based on this dynamic model it is obtained small but obvious improvement of correspondence for calculated results with the experimental data. The ratios of the calculated transuranium isotopes yields (namely, neutron-rich uranium isotopes after the termination of multiple captures) to the experimental yields (for mass number in the interval A=245÷257) for the nuclear explosion "Par" are presented in fig. 1. The calculated yields are presented as with the account of the dynamics for the process, as without the account of the dynamics. The calculated yields are normalized on the ^{245}U yield according to the form of experimental data representation [10]. The error intervals on fig. 1 correspond to the experimental errors at measuring of transuranium nuclides [10]. It is important to note the improvement of agreement at $A \leq 250$ does not cause deterioration of the agreement at $A>250$ as it takes place in the model without the account of the dynamics.

The yields at γ =2.0 and 1.5 are obtained for $T_1 \div T_2$=*(60÷1)* and *(60÷6)* keV and integral fluxes $3.02 \cdot 10^{24}$ and $5.69 \cdot 10^{24}$ *neutron/cm*^{2},

respectively. The yields obtained without account of the dynamics correspond to the energy T=25 keV and integral flux 7.0 $\cdot 10^{24}$ *neutron/cm*^{2}. The data of fig. 1 are presented also in table 1. The experimental isotopes yields used in given work are presented in table 2. The model yields are sensitive as to the flux value as to the mean energy of captured neutrons by the moment of termination for multiple neutron captures.

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Table 1.

Calculated isotope yields for the experiment "Par" *			
Mass number	Yields without considering the dynamics	Yields based on the dynamics	
		$\gamma = 1.5$	$\gamma = 2.0$
245	1	1	1
246	0.784±0.037	0.730±0.034	0.713±0.034
247	1.513±0.069	1.533±0.070	1.535±0.070
248	1.690±0.066	1.553±0.061	1.521±0.060
249	1.969±0.875	1.916±0.852	1.918±0.852
250	1.705±0.083	1.559±0.076	1.529±0.075
251	≥0.866	≥0.890	≥0.890
252	1.654±0.150	1.583±0.144	1.553±0.141
253	0.460±0.042	0.491±0.045	0.490±0.045
254	1.165±0.097	1.128±0.094	1.105±0.092*
255	0.359±0.017	0.393±0.018	0.392±0.018
256	1.397±0.215	1.360±0.209	1.332±0.205
257	0.619±0.111	0.659±0.118	0.656±0.117

* All the yields are normalized to the yield of ^{245}U similar to the representation of the result in the Ref.14.

Table 2.

Experimental isotope yields for the experiment "Par" *			
Mass number	Relative number of atoms	Mass number	Relative number of atoms
245	1.000	252	$(2.2\pm0.2)\times10^{-4}$
246	$(8.5\pm0.4)\times10^{-1}$	253	$(1.1\pm0.1)\times10^{-4}$
247	$(1.1\pm0.05)\times10^{-1}$	254	$(1.2\pm0.1)\times10^{-5}$
248	$(5.1\pm0.2)\times10^{-2}$	255	$(4.3\pm0.2)\times10^{-6}$
249	$(9\pm4)\times10^{-3}$	256	$(2.6\pm0.4)\times10^{-7}$
250	$(4.1\pm0.2)\times10^{-3}$	257	$(5.6\pm1.0)\times10^{-8}$
251	$\leq1.3\times10^{-3}$		

* The experimental data are normalized to the yield of Cm^{245} [11].

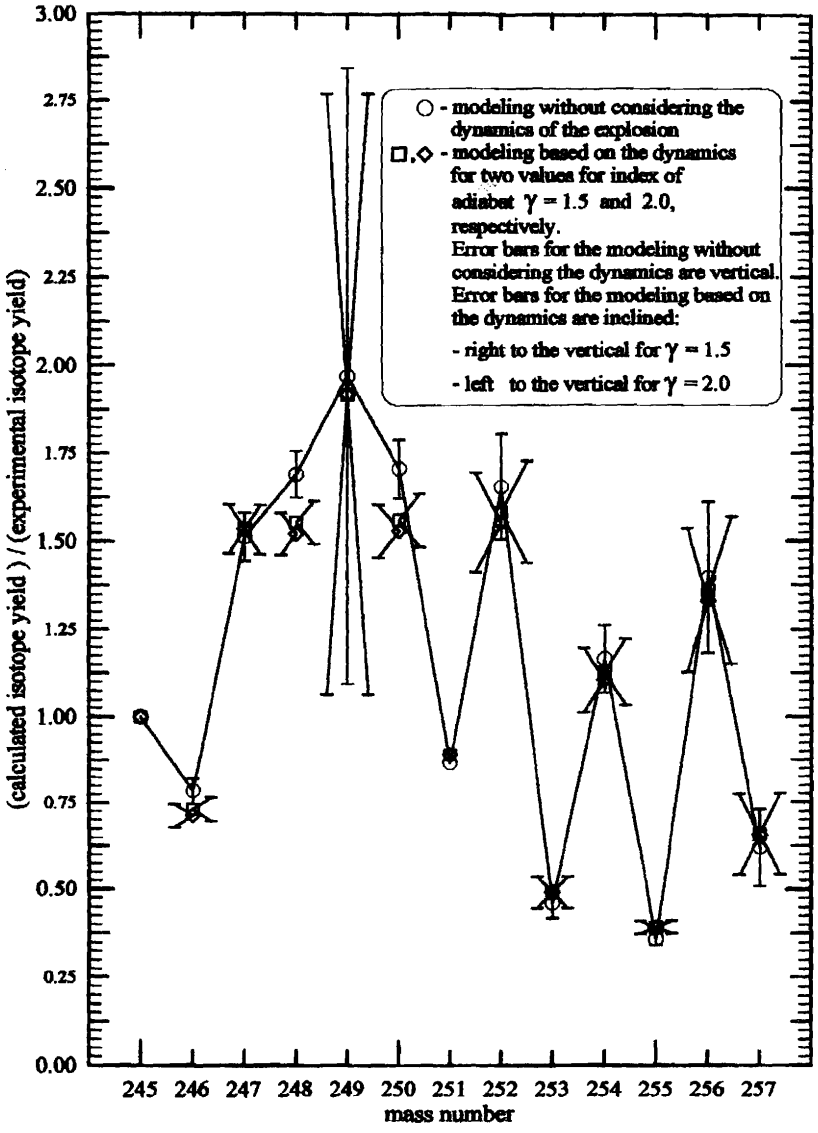

Fig 1. The ratio of calculated values to experimental isotope yields for explosion 'Par' at mass number A=245-257. Calculated values are given for: 1) modeling without considering the dynamics of the explosion and 2)modeling based on the dynamics for two values for index of adiabat $\gamma = 1.5$ and 2.0

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