RELATION OF TRANSURANIUM ISOTOPES YIELDS AS INDICATOR OF THE ACHIVED NEUTRON FLUENCES AT THE PULSE NUCLEOSYNTHESIS

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Importance of the topic (of the artificial nucleosynthesis):

1. An artificial nucleosynthesis is similar (in many features) to cosmological nucleosynthesis (which responsible for element abundance in the universe);
2. An artificial nucleosynthesis gives the chance to create an unknown transuranium isotope and investigate them properties (for example: the odd-even effect for yields at atomic mass $A > 250$; cross-sections);
3. An artificial nucleosynthesis is one of the possible way to reach the island of stability (in the periodic table of elements);
4. Investigating of the artificial nucleosynthesis helps to understand the values of the achieved neutron fluxes in different nuclear tests;

5 …etc.

Creation of transuranic elements under intensive neutron fluxes

For the first time the transuranic elements were discovered in 1952 in the debris of the thermonuclear test "**Mike**" with ^{238}**U**-target. Nuclear and thermonuclear explosions ensure a high neutron flux (from units of 10^{24} and up to ~10^{25} neutron/см^2) at the short time exposure (~ <10^{-6} s – for (n,γ)-reaction) that makes the tests a unique instrument for nuclear physics.

<u>For compare:</u> The maximal neutron flux achieved at the reactor **HFIR** (USA) – 5.5 * 10^{15} neutrons/(cm^{2}s), in the trap of the **PIK** (Russia) – the planned flux is – $4 \cdot 10^{15}$ neutron/(cm^{2}s), during 1 year operation – $1.2 \cdot 10^{23}$ neutron/cm^2 ;

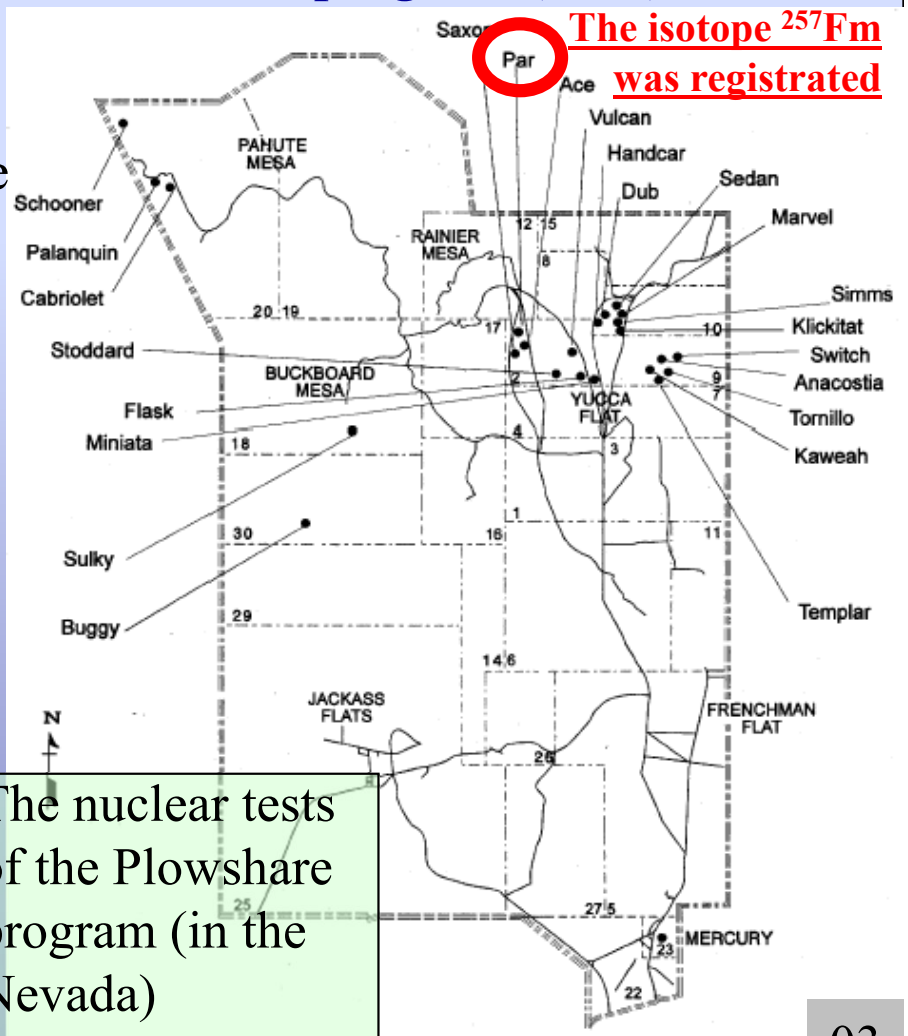
in the trap of **SM-2** (Russia) – $5 \cdot 10^{15}$ neutron/(cm^{2}s);

IGR (the pulse graphite reactor, Russia) – the max integral flux – $1 \cdot 10^{18}$ neutron/cm^2 ;

JAGUAR (Russia) (the burst liquid-type reactor) – $2.5 \cdot 10^{18}$ neutron/(cm^{2}s) in core for the pulse.

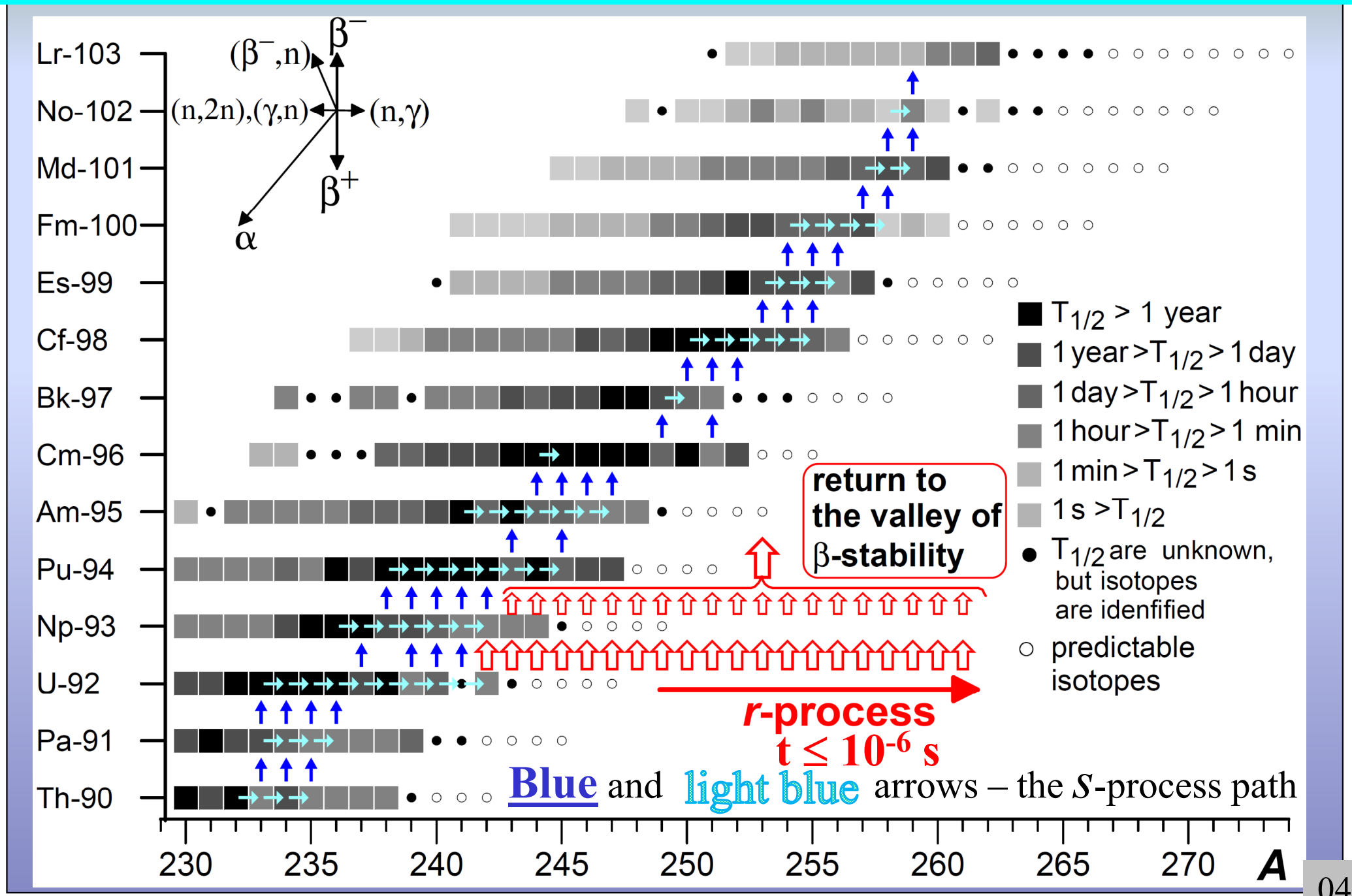
According evaluation in "Cyclamen" and "Hutch" experiments the fluxes obtained in the targets are $1.2 \cdot 10^{25}$ neutron/cm^2 and $4.5 \cdot 10^{25}$ neutron/см^2 , correspondingly.

The investigation of transuranium isotope creation under intensive neutron fluxes was included in the "Plowshare" – program (USA)

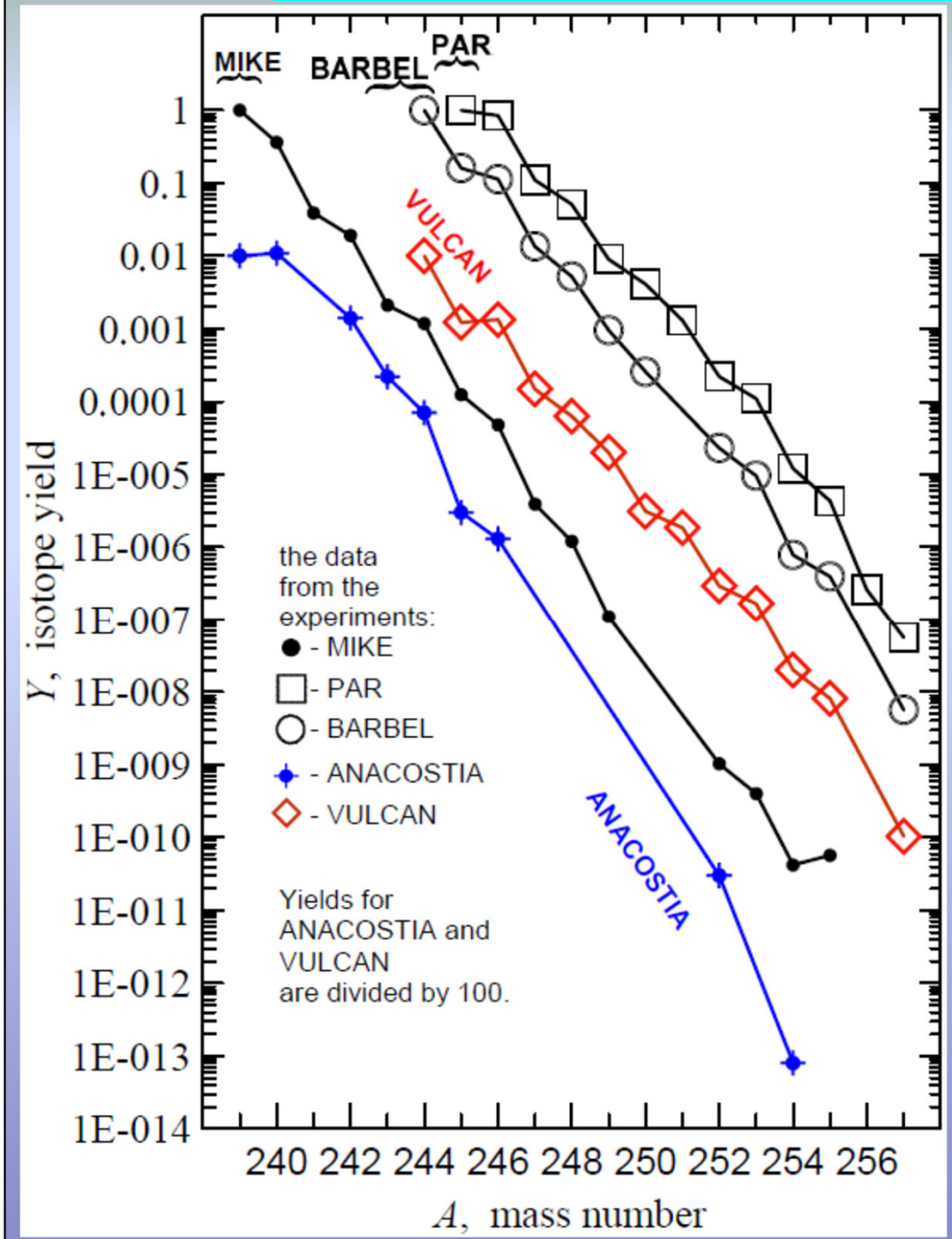


The nuclear tests of the Plowshare program (in the Nevada)

Multiple (n,γ)-captures under conditions of the pulse nucleosynthesis. r – process (rapid process)



Yields in experiments. Difficulties of identifications



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 . 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). As a result the short lived isotopes decay before isotope analysis in the laboratory.

Scheme of transuraniums creation

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

where z and n are the charge and the number of neutrons of the considered nucleus; λ_β, λ_α, and λ_f are the rates of β^- and α-decays and spontaneous fission; $\sigma_{n,\gamma}$, $\sigma_{n,2n}$, $\sigma_{n,3n}$ and $\sigma_{n,f}$ are the corresponding cross-sections of the reactions; and $F(E, t)$ is the neutron flux.

Simplification of the calculation scheme (1):

the rates of λ_β, λ_α, and λ_f are much smaller than the rate of n-capture $\lambda_{n,\gamma}$. We can ignore the contribution from reactions (n, f); (n, 2n), which have a higher energy threshold with respect to the (n, γ) reaction.

Simplification of the calculation scheme (2)

The source function $F(E, t)$ in the present experiments is unknown. So, we perform a convolution for the time and energy.
Here Δt is the exposition time.

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

The applied neutron flux Φ (neutron/cm2) is integrated with respect to time with fixed energy in the interval of ≈20 keV (i.e., a singlegroup energy representation) in accordance with the temperature of the process used in the calculations.

The system of equations for transuranium element creation generated by Eq. (1) then becomes single group in the present static model and takes the form
and this stage of modeling is reduced to calculating the neutron multiple capture reactions.

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

Yu.S. Lutostansky, V. I. Lyashuk, and I. V. Panov. Calculation of Transuranium Element Synthesis in Intensive Neutron Fluxes under Adiabatic Conditions//Bulletin of the Russian Academy of Sciences: Physics, 2010, V.74, No.4, p.504.

Solution for single –group representation

Yield for the *i*-th isotope at the sigle start isotope (z, n) is given by Eq (4):

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

where $N_Z^n(0)$ is the number of nuclei of an initial isotope at $t = 0$; $\lambda_{n,\gamma}^{n+i}$ - designates the rate of the (n,γ)-reaction for the isotope of (z,n+i); and $\prod_{j\neq k}$ denotes the product of all combinations $(\lambda_{n,\gamma}^{j}-\lambda_{n,\gamma}^{k})$ excluding $j = k$.

For calculation of the $\lambda_{n,\gamma}^{n+i}$ the cross section $\sigma_{n,\gamma}(A+i, Z)$ for neutron–rich isotopes was extrapolated in relation to the known cross section $\sigma_{n,\gamma}(A, Z)$ of a preceding isotope in proportion to variations of the neutron binding energy:

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

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

Adiabatic approximation (1)

The model was extended by the process of dynamics which comprises variations of the (n, γ)–reaction cross section upon an environmental T- temperature drop during the adiabatic expansion.

A rough determination of the functional dependency of the temperature decrease in the given region (including a target mass made of the initial isotope 238U) upon the adiabatic expansion can be performed as follows:

1) to specify an interval of $(T_1 - T_2)$ for the mean energy decrease of the captured neutrons (i.e., a description by singlegroup energy of neutrons) upon substance cooling due to the adiabatic expansion within the relevant time interval $t_A - t_B$;

2) to assume that the linear velocity of the explosive expansion of substance $\upsilon = const$ at $t \in [t_A, t_B]$ and

3) To specify the adiabatic index γ for the adiabatic expansion of the volume V:

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

The algorithm for solving the problem of the yields of transuranium isotopes is reduced to partitioning the time interval of multiple captures$[t_A, t_B]$ into m intervals and the sequential solution of nucleosynthesis equations (3) for each given time step $\Delta t_1, \Delta t_2, \ldots, \Delta t_m$.

Yu.S. Lutostansky, V. I. Lyashuk, I. V. Panov. Bulletin of the Russian Academy of Sciences: Physics, 2010, V.74, No.4, p.504.
V. I. Lyashuk. Bulletin of the Russian Academy of Sciences: Physics, 2012, V.76, No.11, p.1182.

Adiabatic approximation (2)

System of equations (3) has the following solution for the time interval $\Delta t = [t_1 , t_2]$ 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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$$N_z^{n+i}(t_2) = \lambda^n \lambda^{n+1} \ldots \lambda^{n+i-1} N_z^n(t_1) \sum_{k=n}^{n+i} \frac{\exp(-\lambda^k \Delta t)}{\prod_{j \neq k}(\lambda^j - \lambda^k)} + \ldots +$$

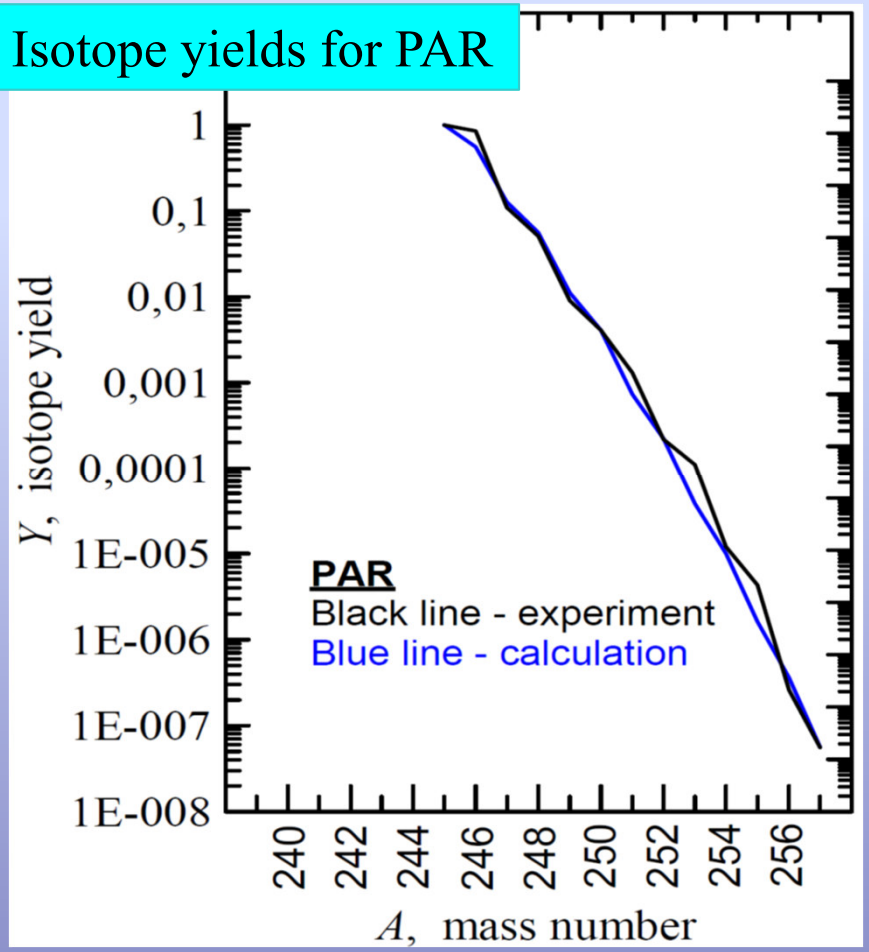
$$\lambda^{n+i-1} N_z^{n+i-1}(t_1)\left[\frac{\exp(-\lambda^{n+i-1}\Delta t)}{\lambda^{n+i}-\lambda^{n+i-1}} + \frac{\exp(-\lambda^{n+i}\Delta t)}{\lambda^{n+i-1}-\lambda^{n+i}}\right] + N_z^{n+i}(t_1)\exp(-\lambda^{n+i}\Delta t)$$

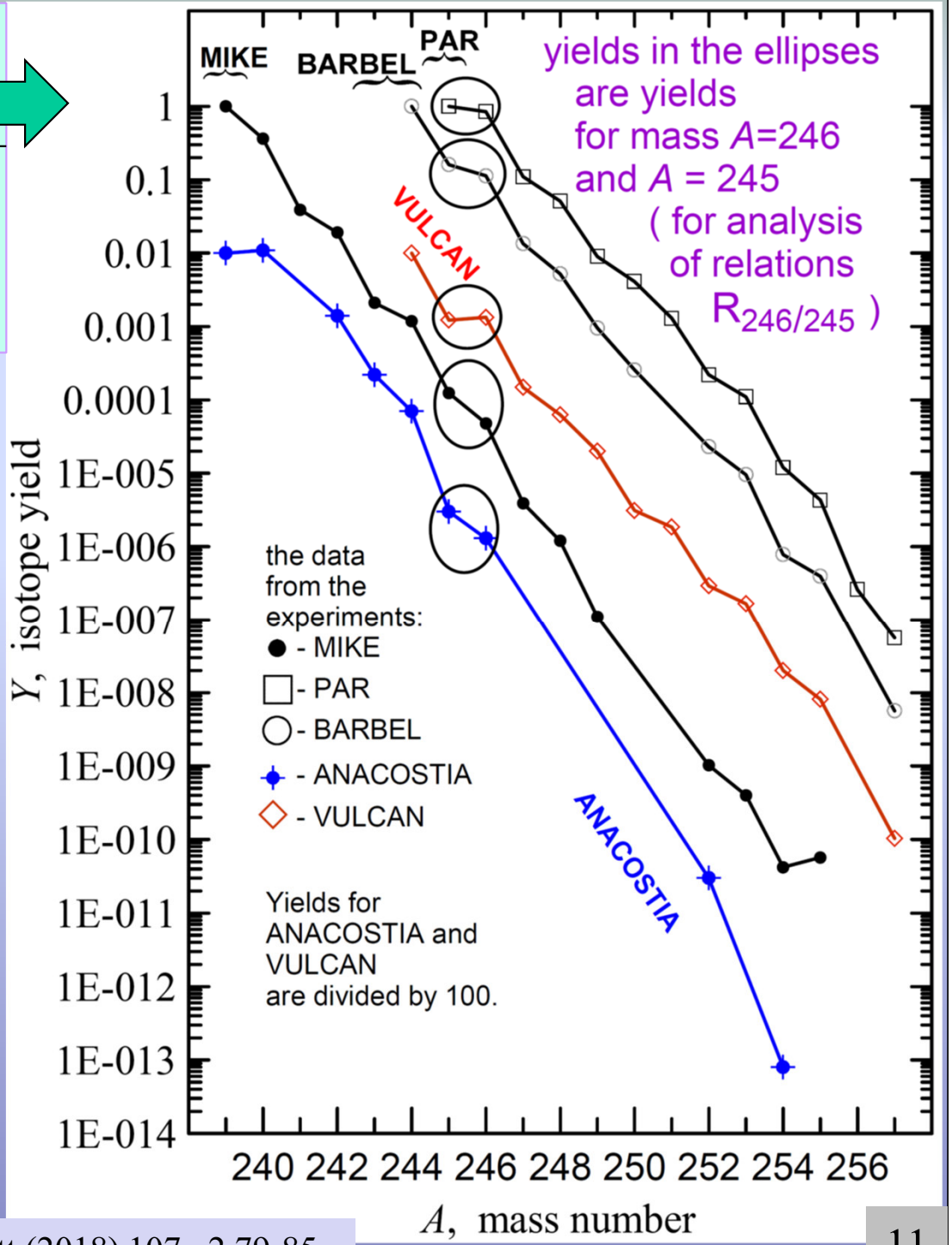
Application of the dynamical model for analysis: Relations of isotope yields

Illustration for relations of yields with mass A=246 and A=245.

Below for dynamical model we consider three three pairs: (245 and 244), (246 and 245), (247 and 246).

Isotope yields for PAR





See yields also in: Lutostansky Yu.S., Lyashuk V.I. JETP Lett.(2018) 107,n2,79-85.

Relation R of isotope yields Y_1 and Y_2 (1)

Static model (i.e., constant cross sections taken at the energy ~20 keV according to the initial temperature of the media) for the time interval [A, B]

$$R = Y_2(A_2) / Y_1(A_1) = N_z^{n+i}(t_B) \Big/ N_z^{n+i-1}(t_B) =$$

$$\lambda^{n+i-1} \times F(\lambda^n \lambda^{n+1} \ldots \lambda^{n+i}, t_B - t_A)$$

Here: z and n – charge and neutron number of (n,γ)-activated isotope, i – number of captured neutrons; A_2 and A_1 – neighboring atomic mass, A_2=A_1+1.

$$N_z^{n+i}(t_B) = \lambda^n \lambda^{n+1} \ldots \lambda^{n+i-1} N_z^n(t_A) \sum_{k=n}^{n+i} \left[\exp(-\lambda^k (t_B - t_A)) \Big/ \prod_{j \neq k} (\lambda^j - \lambda^k) \right],$$

$\lambda^n, .., \lambda^{n+i}$ - rate of (n, γ)-activation of (z,n),..,(z,n+i)-isotopes,

$N_z^n(t_A)$ - starting number of irradiating (z,n)-isotope nuclei,

F - relation sums of $\sum_{k=n}^{n+i}$ and $\sum_{k=n}^{n+i-1}$.

If the cross sections are constant in time (static model), then $\lambda_{n,\gamma}^k (t_B - t_A) = \Phi \sigma_{n,\gamma}^k$, where Φ – is neutron fluence during [A, B] interval of the nucleosynthesis.

Relation R of isotope yields Y_1 and Y_2 (2)

For the **Static** model we obtaine

$$R = Y2(A2) / Y1(A1) =$$

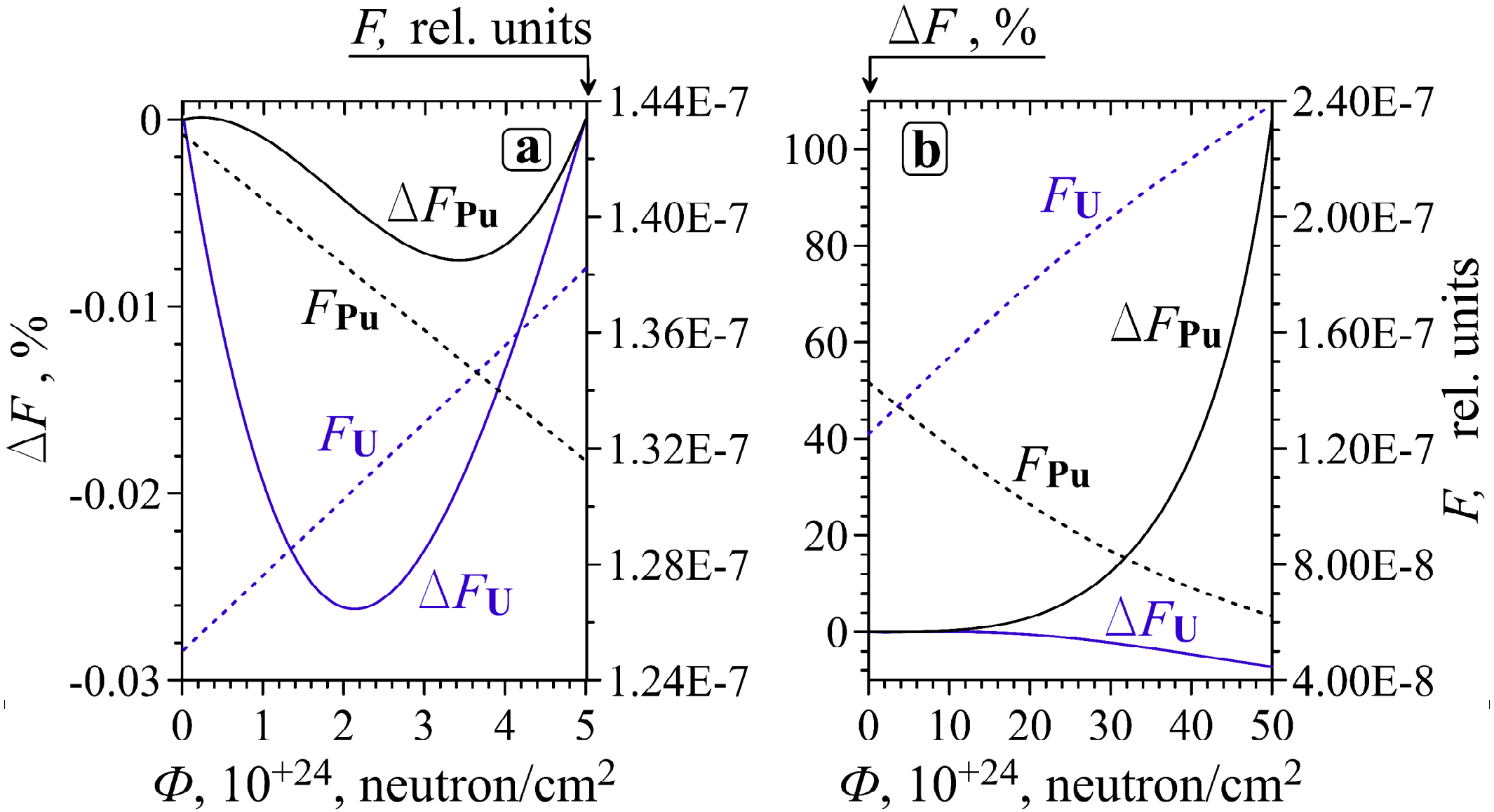
$$\sigma_{n,\gamma}^{n+i-1} \Big/ (t_B - t_A) \times \Phi \times F(\lambda^n \lambda^{n+1} \ldots \lambda^{n+i}, t_B - t_A).$$

Let us demonstrate that for the static model the ***F***-functional is proportional to the fluence ***Φ*** with satisfactory precision. If the hypothesis is right it will mean that the relation depends linear on the -fluence with satisfactory precision too. For yields of isotope mass A_2=246 and A_1=245 (obtained in all nuclear tests) the dependences of ***F*** on neutron fluence for possible fluence ***Φ*** interval (up to < 10^{25} neutrons/cm^2) can be considered as linear.

The feature of the executed experiments that possible to consider the presence of ^{239}Pu isotope as admixture (for initiating of the first burning stage of the thermonuclear device) in the target manufactured from the ^{238}U.

So, we need to consider the the dependences of ***F*** on neutron fluence for possible fluence ***Φ*** for ^{238}U as ^{239}Pu in the starting mixture of the irradiated target.

Dependence of relation R (of isotope yields Y_1 and Y_2) on the neutron fluence Φ



The dependence of F-functional from the neutron fluence for cases of ^{238}U-mono isotope target and for ^{239}Pu-mono isotope targets. The results are given for the static model. ΔF-values (on the left vertical axes indicate the deviation of F from linear trend (in the interval $\Phi < 5\ 10^{24}$ n/cm^2). The approximately linear dependence $F(\Phi)$ indicates on the $R(\Phi)$ close to the linear one. For extreme Φ-fluences the F-values are approximated by parabola (see the b-part).

Relation R of isotope yields Y_1 and Y_2 for dynamic model

Test of the Linear Hypothesis for Dynamical Model. The model realize the time dependence of cross sections and ensures good or satisfactory agreement with experimental isotope yields. The dynamical algorithm is reduced to partition of the r-process time interval $[t_A, t_B]$ into small ($\Delta t \sim 10$ ns) steps and for every isotope N_z^{n+i} we calculate (at each current time step) the yields produced by starting isotope of the target and then add the yields from next isotopes (as "new"-targers: the nuclei $N_z^{n+1}, \quad N_z^{n+2}, \ldots, N_z^{n+i-1}$).

For dynamic model the relation is:

$$R = Y_2 / Y_1 = N_z^{n+i}(t_2) = \lambda^n \lambda^{n+1} \ldots \lambda^{n+i-1} N_z^n(t_1) \sum_{k=n}^{n+i} \left[\exp(-\lambda^k \Delta t) \Big/ \prod_{j \neq k} (\lambda^j - \lambda^k) \right] + \ldots +$$

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

Here $\Delta t = [t_1, t_2]$; $t_1 > t_A$ and $t_2 \leq t_B$.

V.I. Lyashuk, Results in Physics, V.56, 2024, 107234.

Dynamical model. Results of the calculations (1)

The parameters of the dynamical model and important features :

- the nuclesynthesis start at the temperature 20 keV;
- adiabatic index γ =1.5;
- during the nucleosynthesis the accepted decrease of the temperature is 20 times (that roughly correspond to the expansion of the matter);
- cross section for irradiated target – JEFF-3 library;
- calculation were realized for cases of 238U-target with different 239Pu-admixtures and for the case of pure 238U-target;
- the model with 239Pu-admixture indicates that uranium part and plutonium part in the target are irradiated by different neutron fluences;
- the obtained results for isotope yields and realtion of isotope yields are correspond to the minimal values of r.m.s (root mean square) deviation compare to the experimental yields;
- most of the (considered) experiments are normalized on the A=244 or 245 yields;
- the errors of the yields are significant and increase for larger A-mass;
- the odd-even effect in yields of transuraniums appears at mass A>250;
- the number of “candidates” for considered A-mass are limited.

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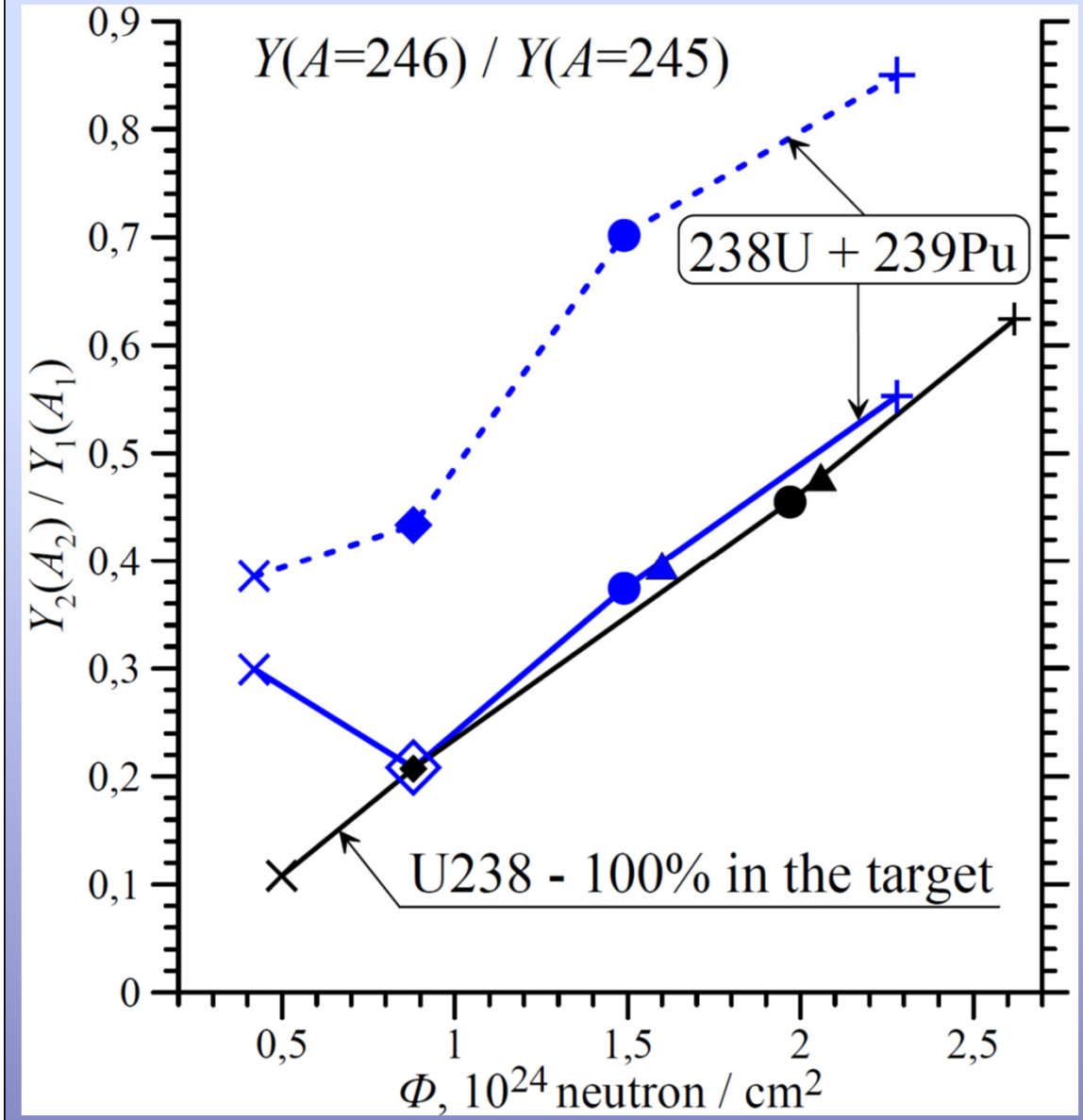
Dynamical model. Results (2). Relation of Yields for Isotopes A=246 and A=245

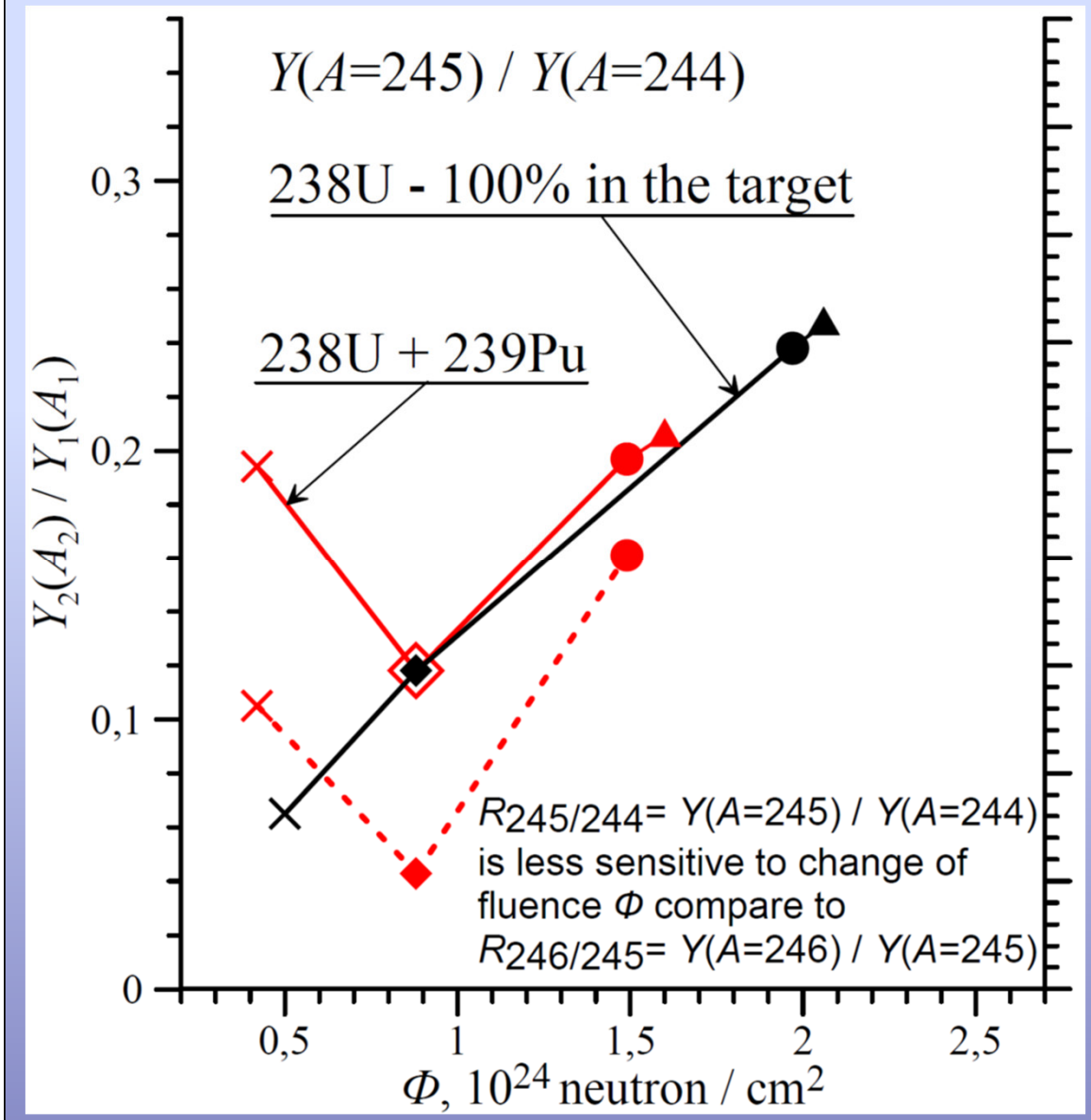


✕ -Mike;
◇, ◆ -Anacostia;
● -Barbel;
✚ -Par;
▲ -Vulcan;

1) dashed blue line - experiment;
2) solid blue lines - calculations for model with (238U+239Pu)-targets; admixtures of 239Pu (in %) were irradiated by fluences (n/cm^2):
Mike – (0.4%), 1.1×10^{24} ;
Anacostia – (0.1%), 1.3×10^{24} ;
Barbel – (0.3%), 2.3×10^{24} ;
Par – (2.0%), 2.4×10^{24} ;
Vulcan – (0.2%), 2.7×10^{24} ;
3) solid black lines - calculations for model with mono isotope target (100% of 238U).

Relation R= $Y(A$=246) / $Y(A$=245) is the most strong confirmation of approximately linear dependence $R(\Phi)$.

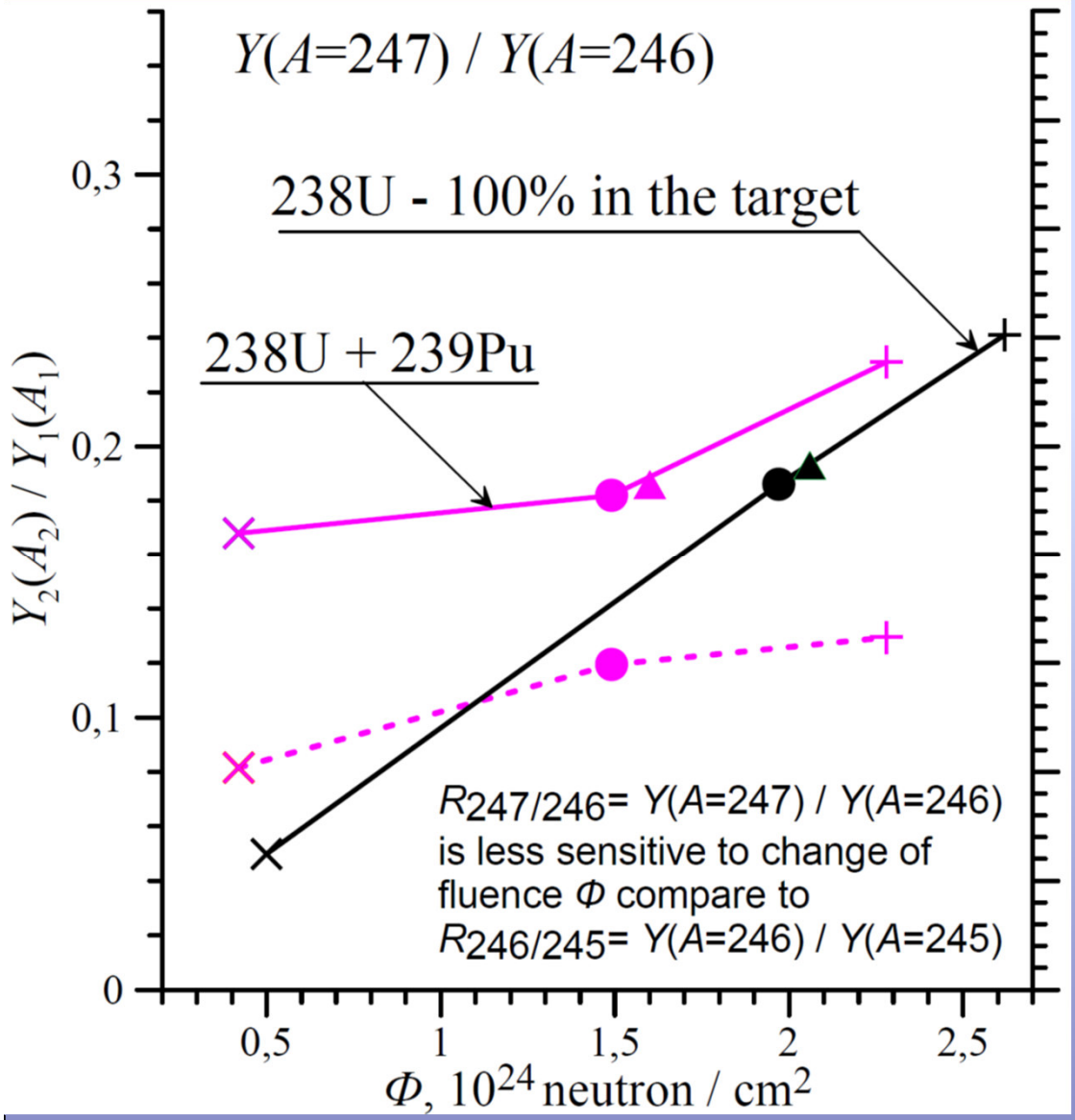
Dynamical model. Results (3). Relation of Yields for Isotopes A=245 and A=244



✕ -Mike;
◇, ◆ -Anacostia;
● -Barbel;
✚ -Par (data are absent);
▲ -Vulcan;

1) dashed red line - experiment;
2) solid red lines - calculations for model with (238U+239Pu)-targets; admixtures of 239Pu (in %) were irradiated by fluences (n/cm^2):
Mike – (0.4%), 1.1×10^{24} ;
Anacostia – (0.1%), 1.3×10^{24} ;
Barbel – (0.3%), 2.3×10^{24} ;
Vulcan – (0.2%), 2.7×10^{24} ;
3) solid black lines - calculations for model with mono isotope target (100% of 238U).

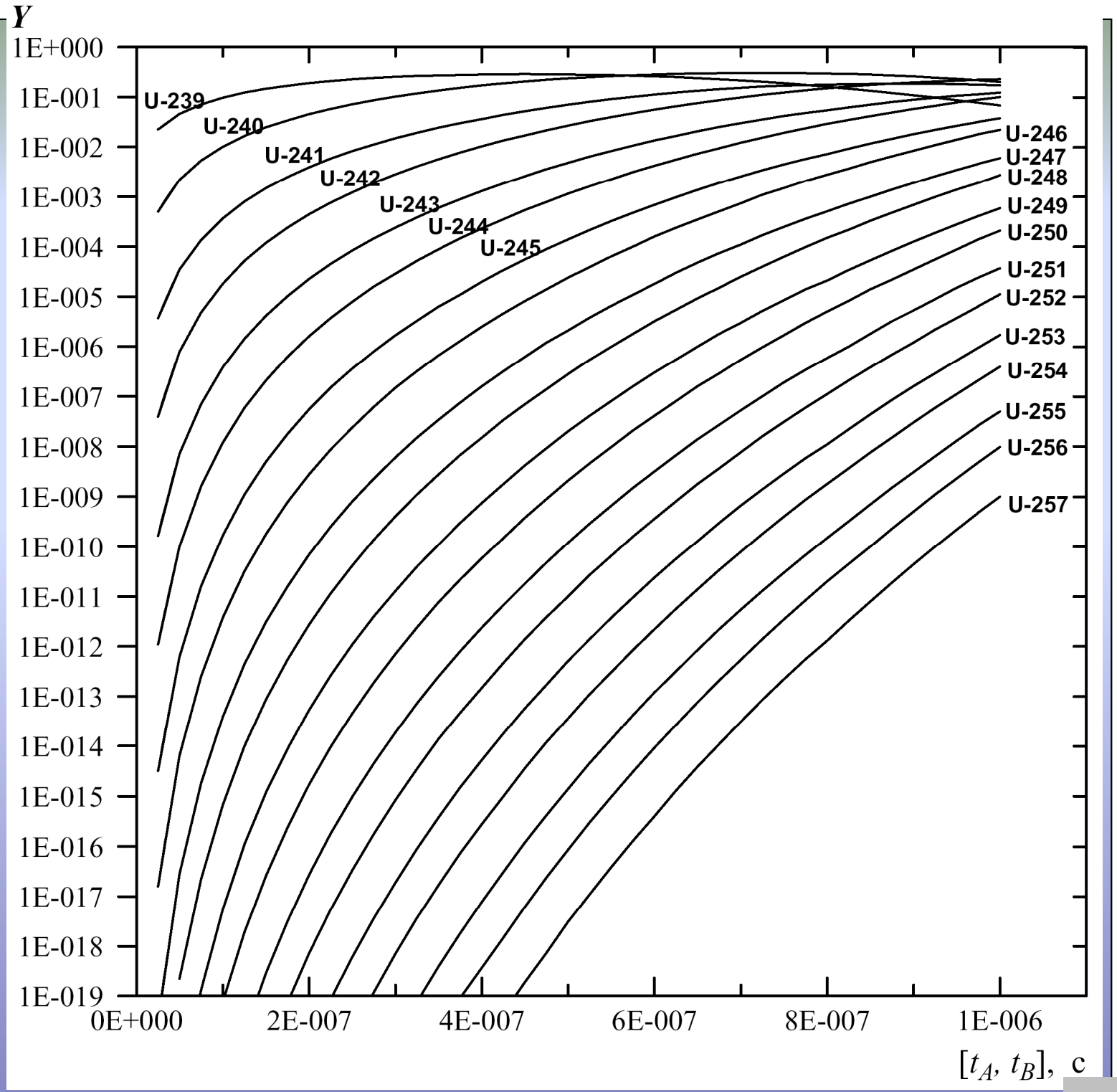
Dynamical model. Results (4). Relation of Yields for Isotopes A=247 and A=246



✕ -Mike;
◇, ◆ -Anacostia (data are absent) ;
● -Barbel;
✚ -Par;
▲ -Vulcan;
1) dashed violet line - experiment;
2) solid violet lines - calculations for model with (238U+239Pu)-targets; admixtures of 239Pu (in %) were irradiated by fluences (n/cm^2):
Mike – (0.4%), 1.1×10^{24} ;
Barbel – (0.3%), 2.3×10^{24} ;
Vulcan – (0.2%), 2.7×10^{24} ;
Par – (2.0%), 2.4×10^{24} ;
3) solid black lines - calculations for model with mono isotope target (100% of 238U).

Calculation of isotop yields for "Par"

The dynamical model allows to obtaine the time dependence of isotope accumulation during the nucleosynthesis interval.



CONCLUSION

1. The proposed dynamical model ensure satisfactory or good agreement with experimental data.
2. The results obtained for relation of isotope yields depending on the neutron fluence indicate on the approximately linear dependence. The linear dependence reveals itself the most clear for relation of isotope yields with mass A=246 and A=245: $R_{246/245} = Y(A=246) / Y(A=245)$.
3. Analysis of isotope relations is limited by small number of date from nuclear test.

Dear colleagues,
thank you a lot for attention !