Relations of isotope yields as an indicator of neutron fluxes in artificial rapid process

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Experimental investigation on creation of transuranium elements in intensive neutron fluxes

The first time the transuranium elements (up to A=255) were discovered in the yields of thermonuclear test "**Mike**" in 1952 (with **^{238}U**-target**)**.

Nuclear and thermonuclear explosions ensure an extreme neutron fluxes (10^{24} – 10^{25} neutron/cm^2) at the very short irradiation (~ <10^{-6} s – for reaction of neutron captures) and so they are the unique instrument for investigation in nuclear physics

For comparison:

The maximal flux obtained at the reactor **HFIR** – 5.5 * 10^{15} neutron/ (cm^{2}s);

In the neutron trap of the **PIK**-reactor(the project) – $4\cdot10^{15}$ neutron/ (cm^{2}s);

fluence during 1 year of the work - $1.2\cdot10^{23}$ neutron/ (cm^{2}s);

in the trap of **CM-2** – $5\cdot10^{15}$ neutron/ (cm^{2}s);

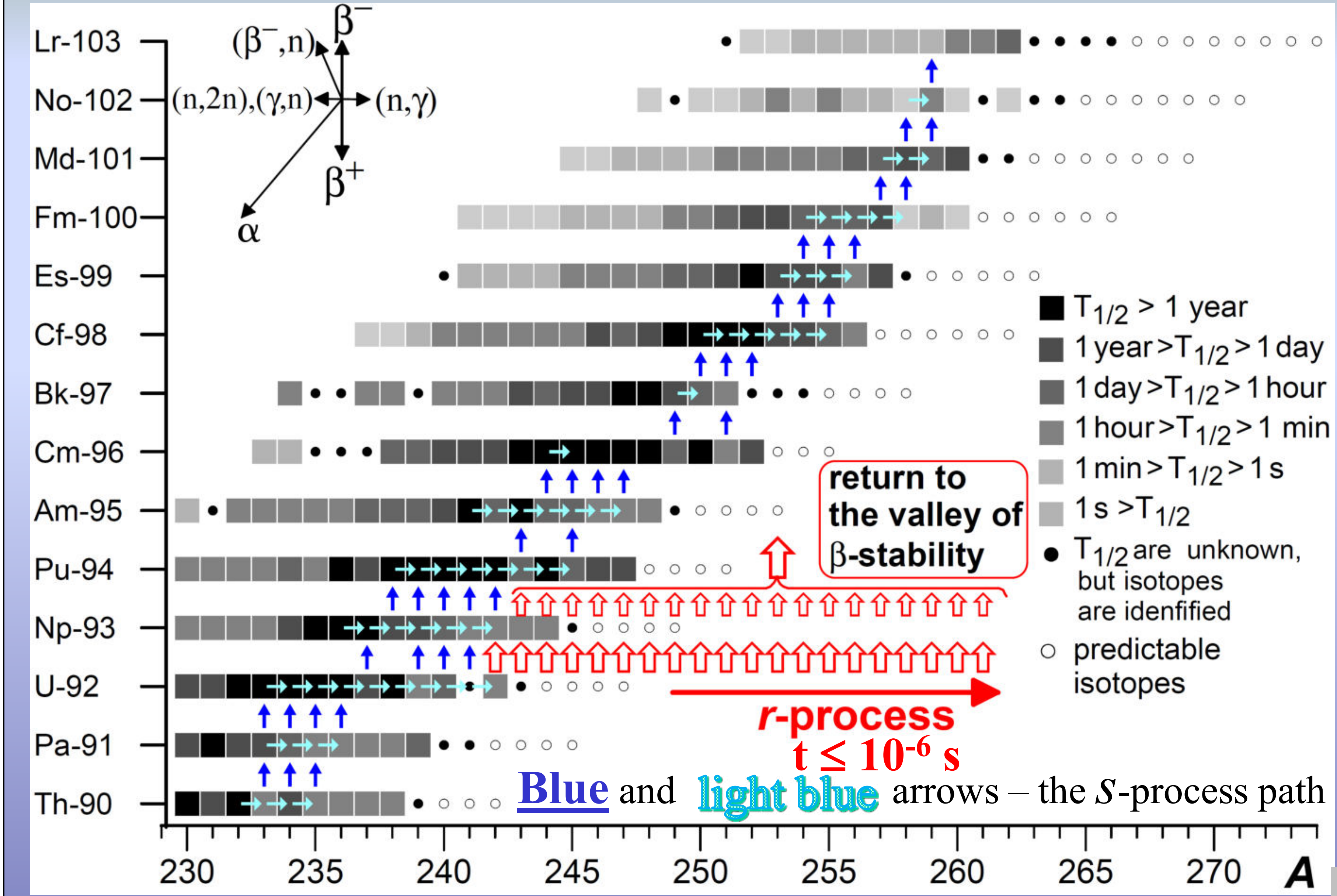
IGR (pulse graphite reactor) – the maximal flux – $1\cdot10^{18}$ neutron/ (cm^{2}s);

BIGR (the pulse reactor) - $1.2\cdot10^{16}$ neutron/ (см^{2}c) in the central channel;

GIDRA (the pulse liquid-type reactor) – $8\cdot10^{14}$ neutron/ (cm^{2}s);

JAGUAR (liquid-type pulse reactor) – $2.5\cdot10^{18}$ neutron/ (cm^{2}s) for pulse in the channel.

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



Blue and **light blue** arrows – the *S*-process path

Scheme of transuraniums creation

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

where z and n 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.

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

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.

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

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):

$$N_Z^{n+i} = \lambda_{n,\gamma}^{n}\lambda_{n,\gamma}^{n+1}\dots\lambda_{n,\gamma}^{n+i-1}N_Z^{n}(0)\sum_{k=n}^{n+i}\frac{\exp(-\lambda_{n,\gamma}^{k}t)}{\prod\limits_{j\neq k}(\lambda_{n,\gamma}^{j}-\lambda_{n,\gamma}^{k})}, \tag{4}$$

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

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

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

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

$\Delta t_1, \Delta t_2, \ldots, \Delta t_{\mathrm{m}}$.

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 $t_2 \leq t_B$

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

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

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

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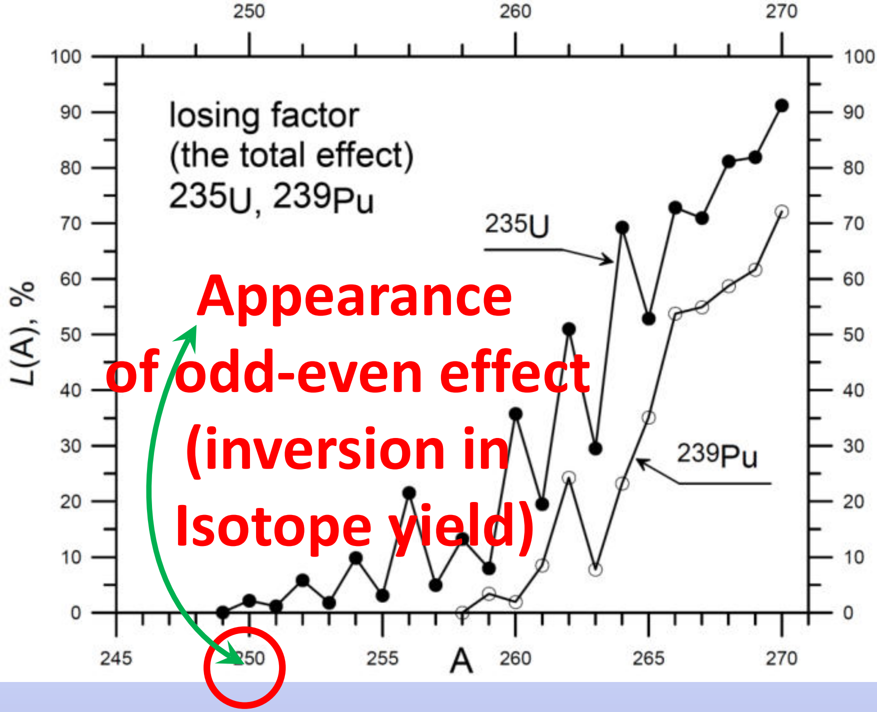
$$N_z^{n-i}(t_2) = \lambda^n \lambda^{n+1} ... \lambda^{n+i-1} N_z^n(t_1) \sum_{k=n}^{n+i} \frac{\exp(-\lambda^k \Delta t)}{\prod_{j \neq k} (\lambda^j - \lambda^k)} + ... + \lambda^{n+i-1} N_z^{n+i-1}(t_1)\left[\frac{exp(-\lambda^{n+i-1} \Delta t)}{\lambda^{n+i} - \lambda^{n+i-1}} + \frac{exp(-\lambda^{n+i} \Delta t)}{\lambda^{n+i-1} - \lambda^{n+i}}\right] + N_z^{n+i}(t_1)\, exp(-\lambda^{n+i} \Delta t)$$

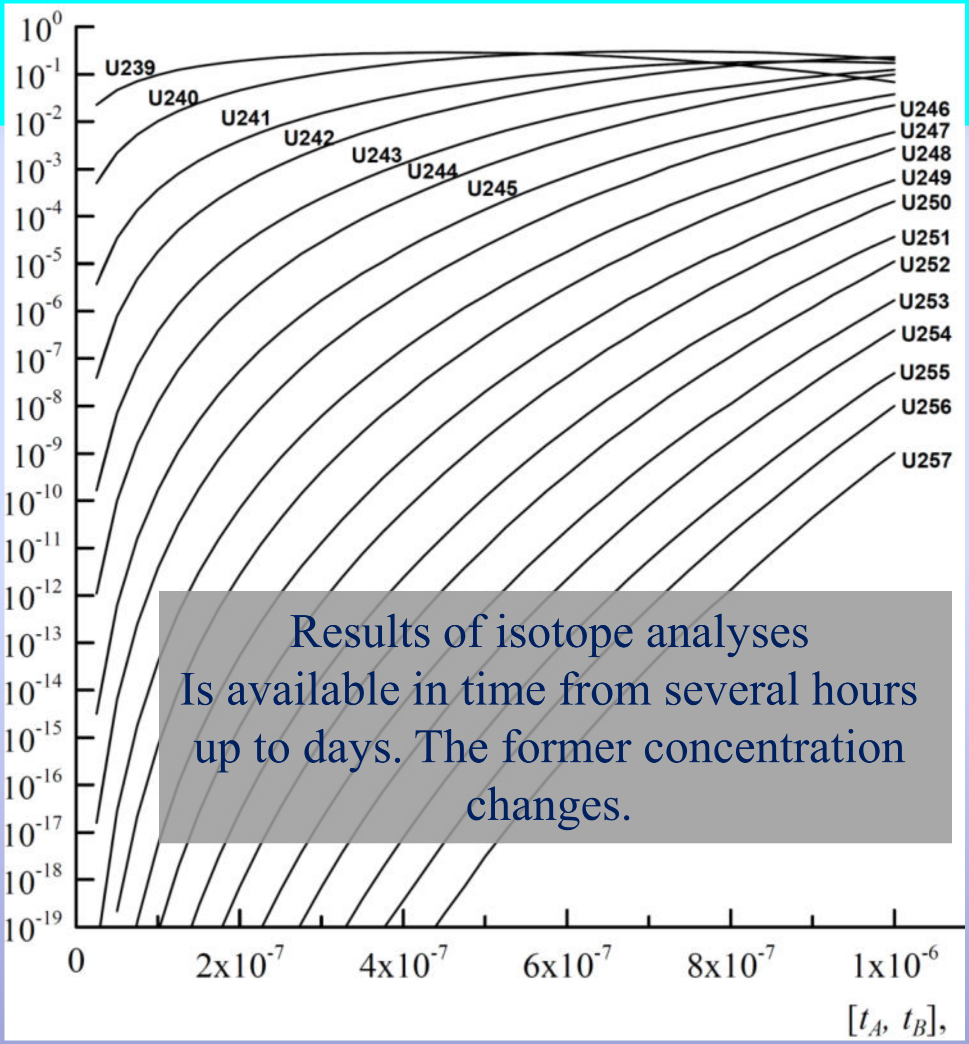
Creation of thransuraniums in the *r*-process and decrease of isotope concentration (losing-factor) in (U+Pu)-target (ABM-model)



Appearance of odd-even effect (inversion in Isotope yield)

Losing-factor: decrease of isotope concentration (created at *r*-process for 238U and 239Pu targets) due to decay

The specific approach: mixing of U-target and Pu during the explosive nucleusynthesis is inavitably.



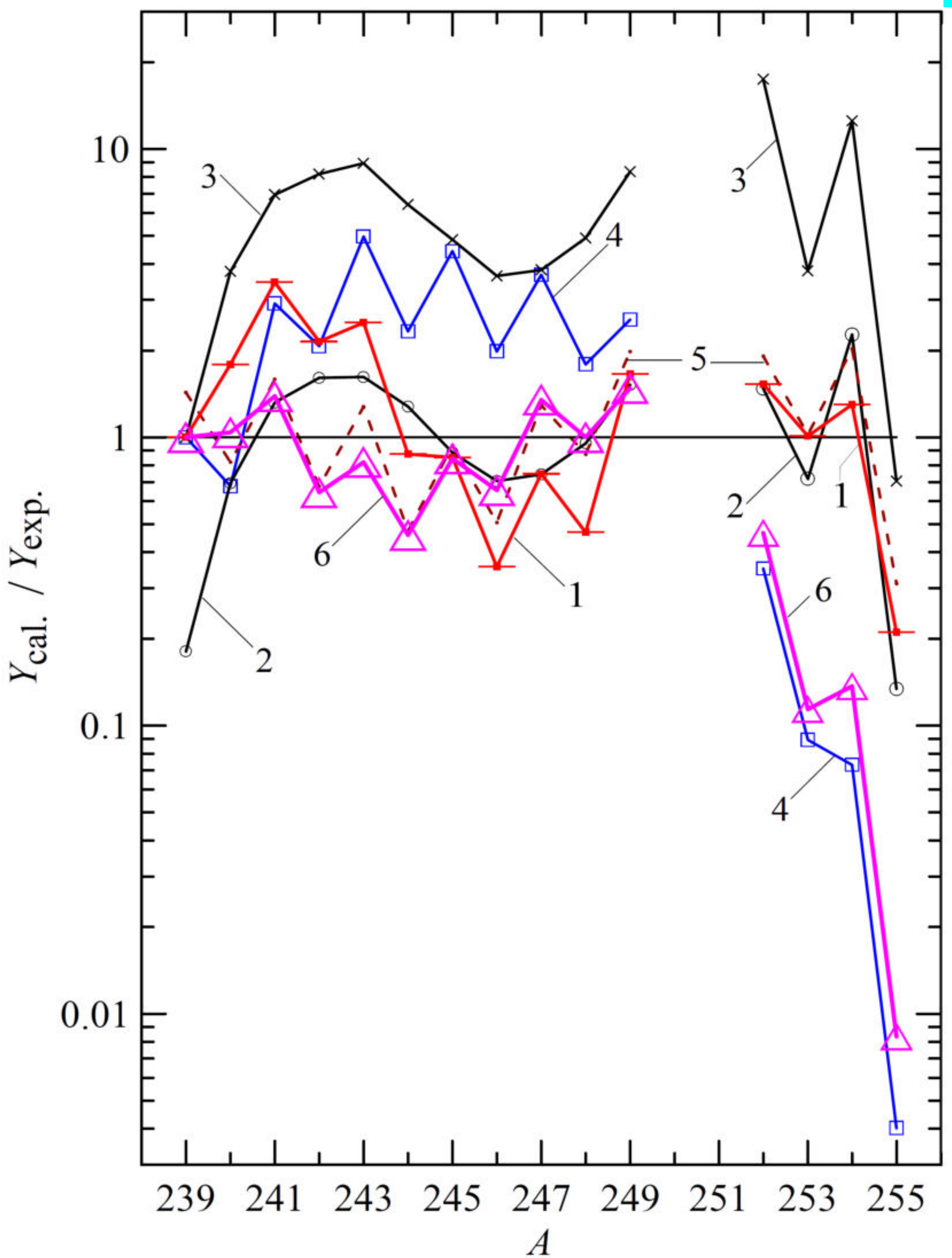
Results of isotope analyses Is available in time from several hours up to days. The former concentration changes.

Time dependence of isotope creation during the interval of nucleosynthesis , (s)

Yu.S. Lutostansky, and V.N. Tikhonov, Bull. Russ. Acad. Sci. Phys. 76, 534 (2012)

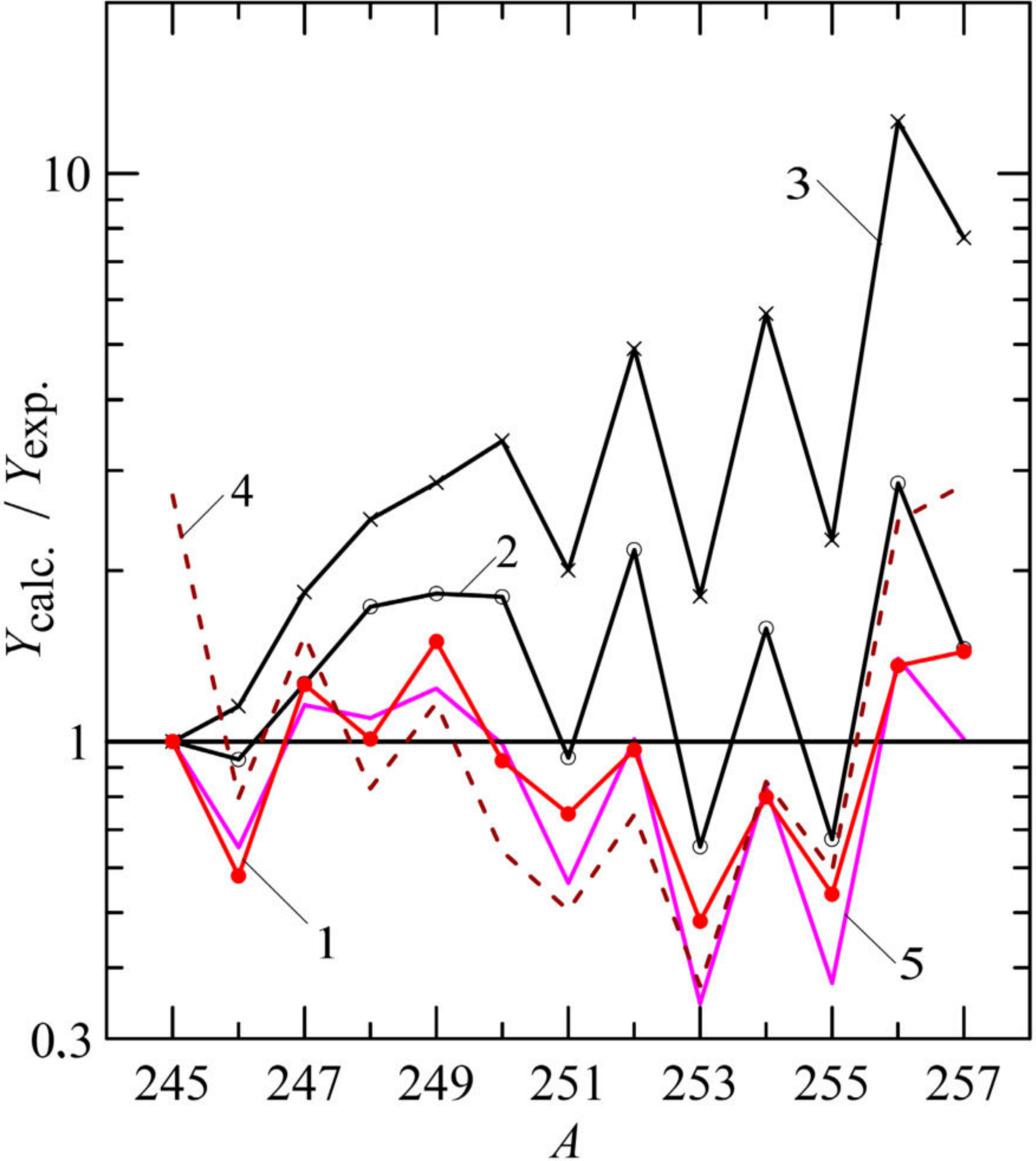
V. I. Lyashuk. // Bull.of Rus.Ac.of Sci. Phys, 2012, Vol. 76, No. 11, pp. 1182–1186.

Experiment MIKE. Relation of calculated (ABM-model) yields to experimental data



1. Lutostansky YuS, Lyashuk VI, **(RED),** δ %=91%. JETP Lett.(2018) 107,n2,79-85
2. Dorn D. W. *Phys*. *Rev*. 1962. V. 126. p. 693; without normalization on $Y(\mathrm{A}=239)_{\mathrm{calc}}$;
3. Dorn D. W. *Phys*. *Rev*. 1962. V. 126. p. 693; with normalization on $Y(\mathrm{A}=239)_{\mathrm{calc}}$; δ %=681%.
4. Zagrebaev V. I., Karpov A. V., Mishustin I. N., Greiner W. *Phys*. *Rev*. C. 2011. V. 84. 044617; δ %=180%.
5. Экспериментальный фит $Y(A_i)/\ Y(A=239) = \exp\{-\ b\cdot A_i + c)$, b=1.395, c=340.584; δ %=60.2%;
6. **results of this work**, δ = 51 %, $\Phi = 0.42\times10^{24}$ н/см^2 и 1.12×10^{24} н/см^2 для изотопов мишени – ^{238}U(99.6%) и ^{239}Pu(0.4%).

Experiment PAR. Relation of calculated (ABM-model) yields to experimental data



1. Lutostansky YuS, Lyashuk VI, **(RED)**
δ =33%, JETP Lett.(2018) 107,n2,79.

2. Dorn D W and Hoff R W 1965
Phys. Rev. Lett. 1965 **14** 440;
U-target,
fluence = 4.2E+24 neutr./cm^2;
δ =75.9%.

3. Dorn D W and Hoff R W 1965
Phys. Rev. Lett. 1965 **14** 440;
U-target,
fluence = 4.8E+24 neutr./cm^2;
δ =417%.

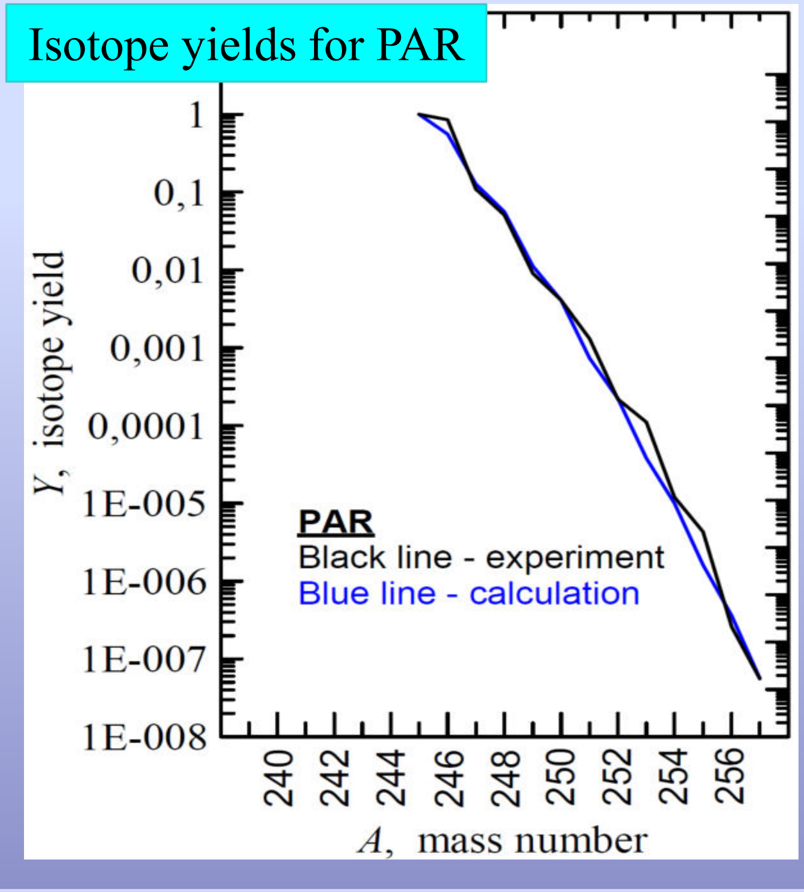
4. Экспериментальный фит
$Y(A_i)/ Y(A=245) = \exp\{- b \cdot A_i + c)$,
b=1.388, c=341.015;
δ =87%.

5. **results of this work**, δ=33 %,
Φ=2.28×10^{24} н/см^2 и 2.44×10^{24} н/см^2
для мишени с составом:
^{238}U(99.6%) и ^{239}Pu(0.4%).

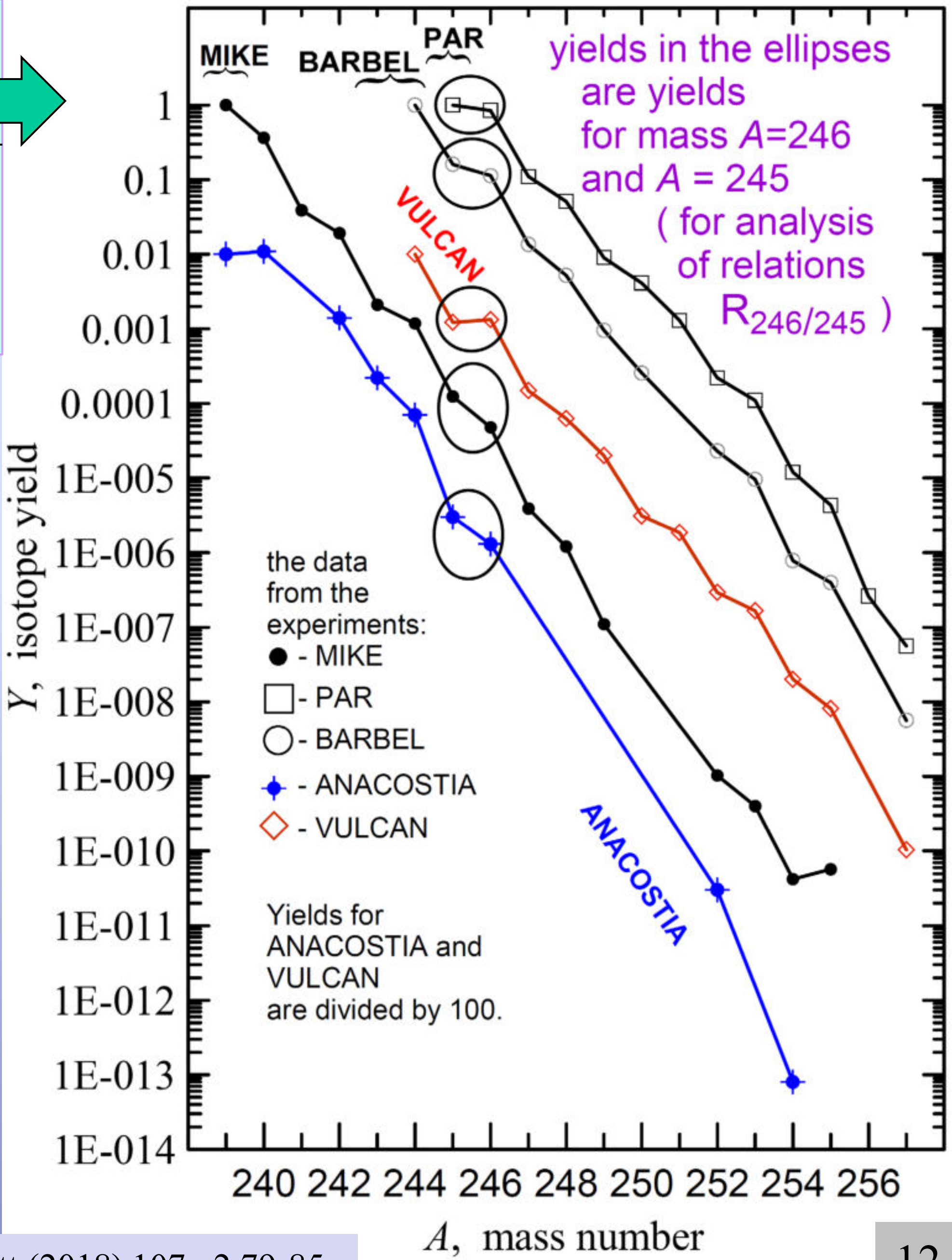
Application of the dynamical model for analysis: Relations of isotope yields

Illustration for relations of yields
with mass *A*=246 and *A*=

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, $A_2 = A_1 \pm 1$.

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

$\lambda^n, \ldots, \lambda^{n+i}$ - 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

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

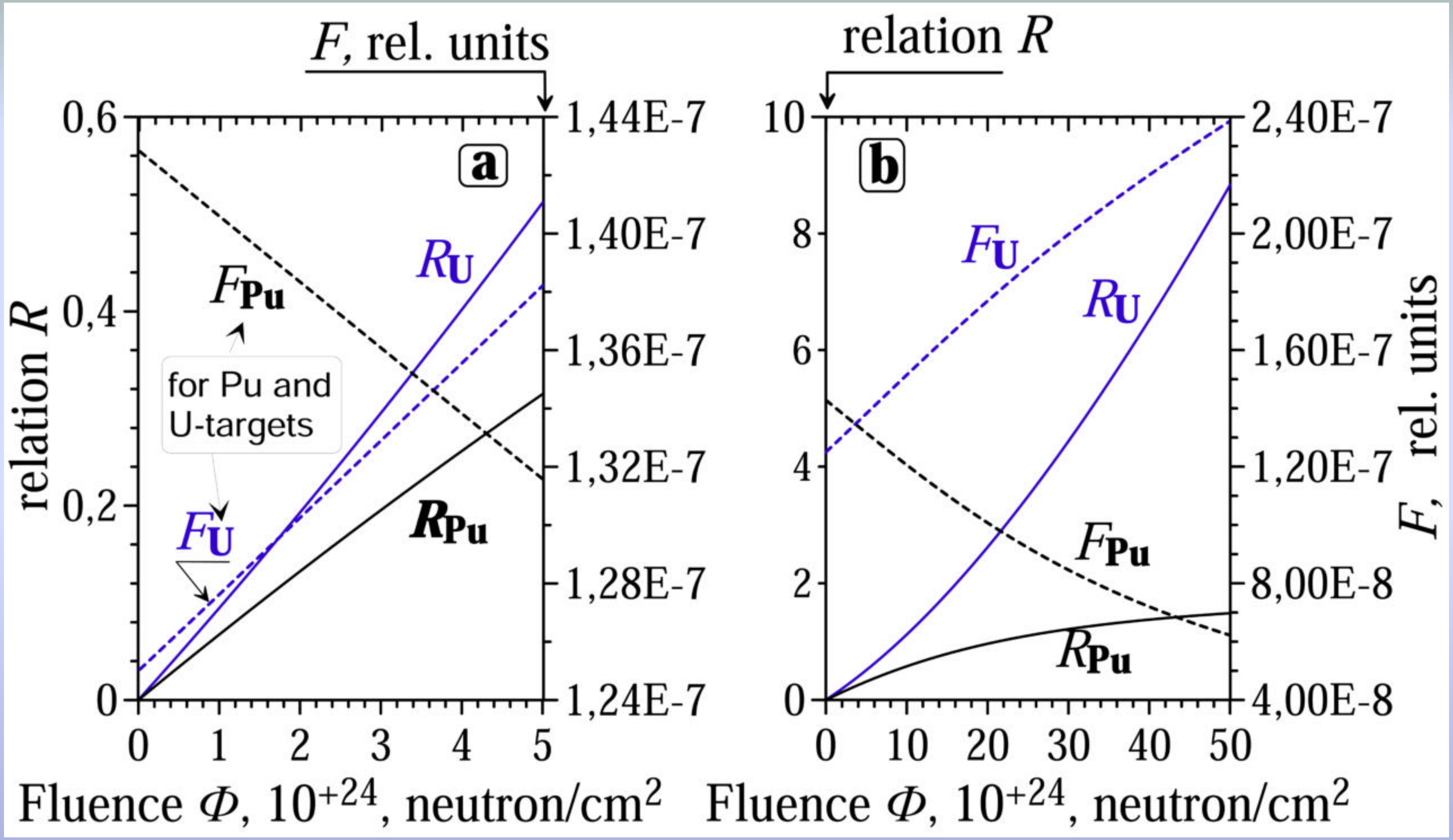
$$\sigma_{n,\gamma}^{n+i-1} \Big/ (t_B - t_A) \times \Phi \times F(\lambda^{n} \lambda^{n+1} \ldots \lambda^{n+i}, t_B - t_A).$$

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.

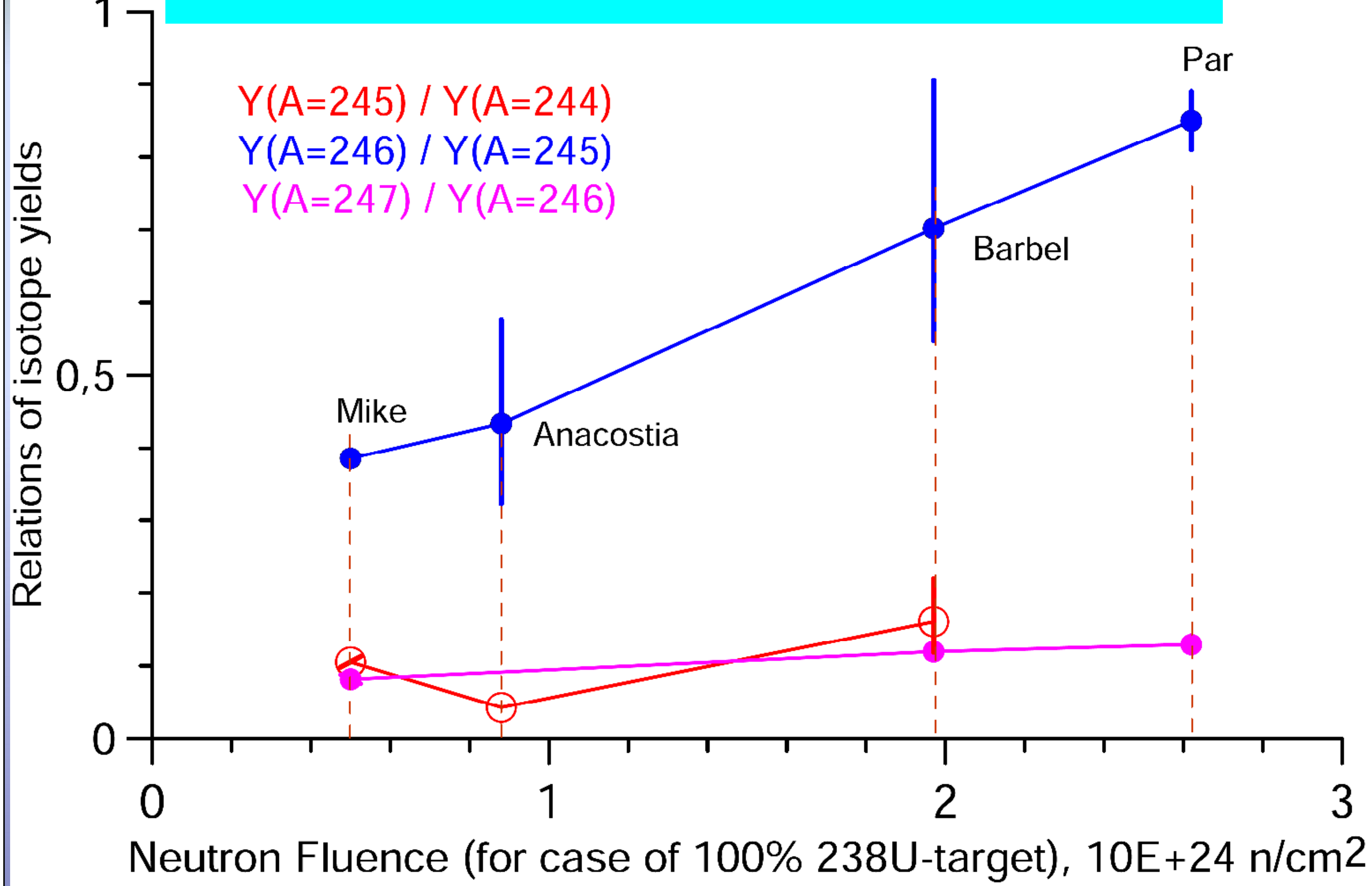
So, we need to consider the the dependences of ***F*** on neutron fluence for possible fluence ***Φ*** for ^{238}U as ^{239}Pu in the starting mixture of the irradiated target.

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



The dependence of F-functional and R-value from the neutron fluence for cases of ^{238}U-mono isotope target and for ^{239}Pu-mono isotope targets for two scales of the fluence. The results are given for the static model). The approximately linear dependence *F*(*Φ*) (in the interval Φ< 5E+24 n/cm^2 –see left a-part) indicates on the *R*(*Φ*) close to the linear one. For extreme *Φ*-fluences (see right part) the *F*-values are approximated by parabola (b-part).

Relation of Isotope Yields. 1



Experimental and Calculated Yields

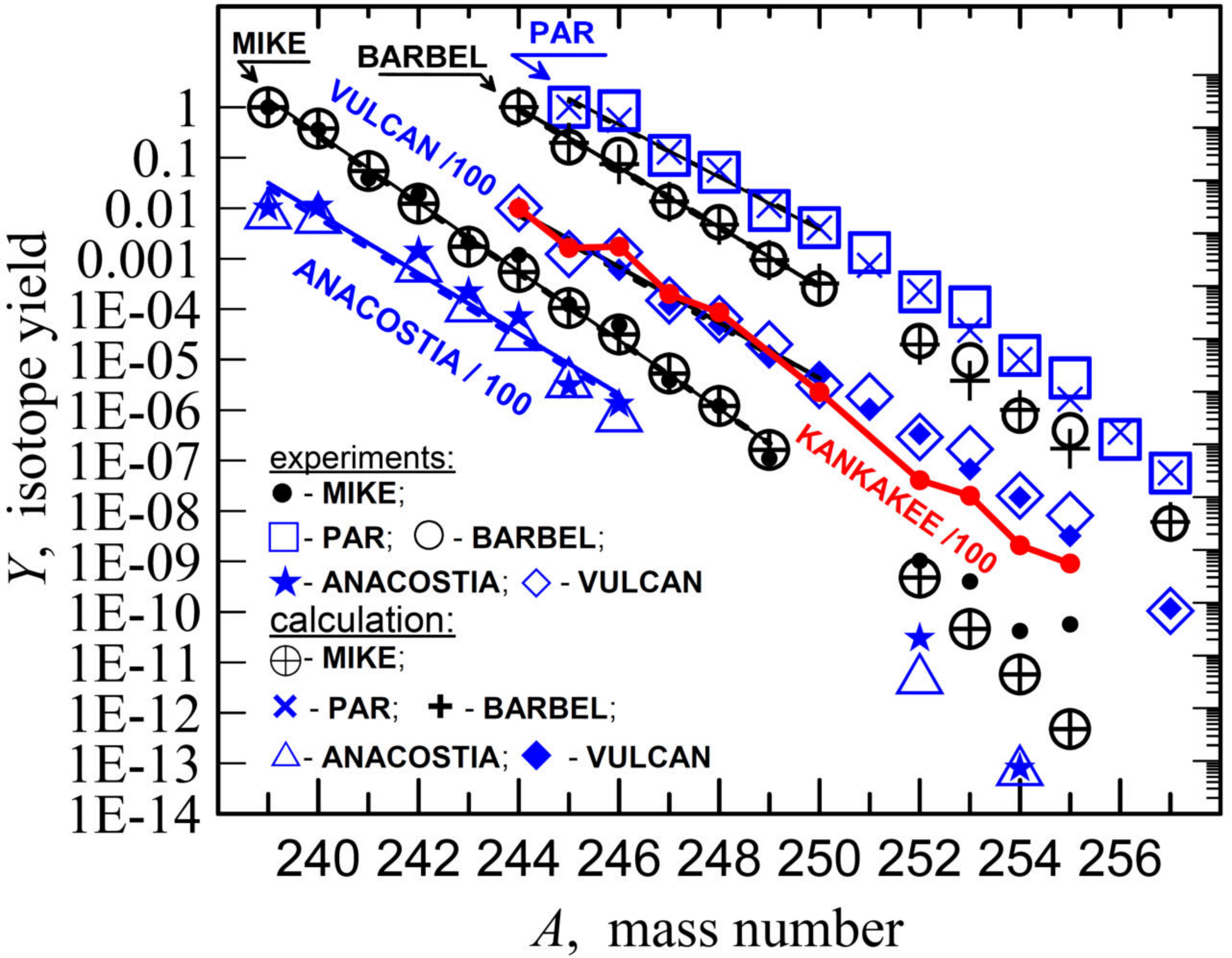
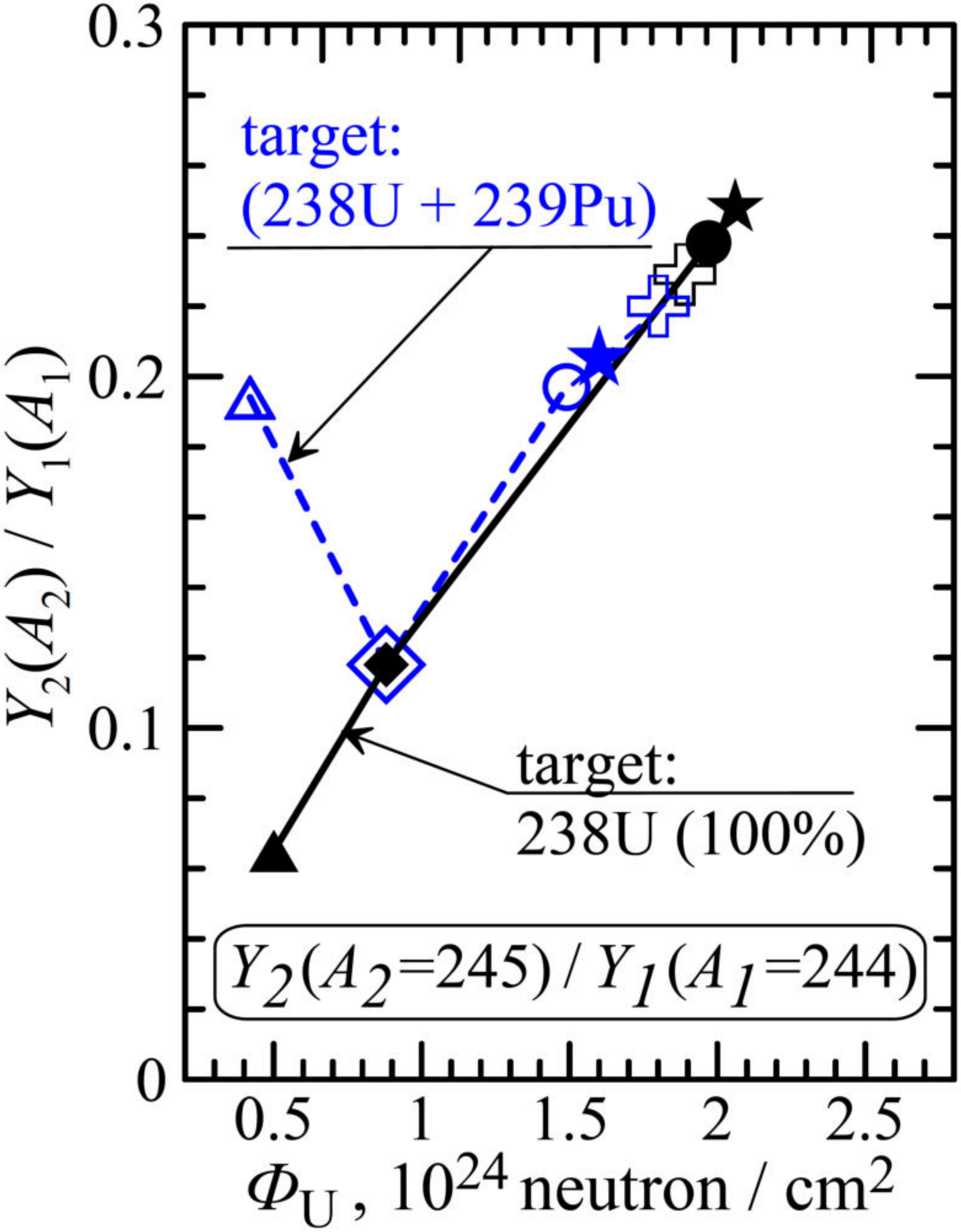


Table. Experimental and calculated parameters for the exponential fit of normalized yields (in the form $Y(A)/Y(_{\max}) = \exp(b \times A + c)$ in nucleosynthesis experiments.

	Mike, $A_{\max}$= 249		Anacostia, $A_{\max}$= 246		Kankakee, $A_{\max}$= 250		Barbel, $A_{\max}$= 250		Vulcan, $A_{\max}$= 250		Par, $A_{\max}$= 250	
	b	*c*	*b*	*c*	*b*	*c*	*b*	*c*	*b*	*c*	*b*	*c*
Exp.	-1.570	375.491	-1.376	329.938	-1.347	338.770	-1.362	332.255	-1.268	309.244	-1.197	293.676
Calc.	-1.557	372.358	-1.372	328.838	-1.412	344.742	-1.343	327.690	-1.266	303.924	-1.144	280.666
Δ (error)	0.013		0.004		0.048		0.019		0.002		0.053	

Relation of Yields Y(A=245) / Y(A=244)



Target (238U + 239Pu):

△ - **MIKE**; ◇ - **ANACOSTIA;**

○ - **BARBEL**;

★ - **VULCAN;** ✙ - **KANKAKEE**

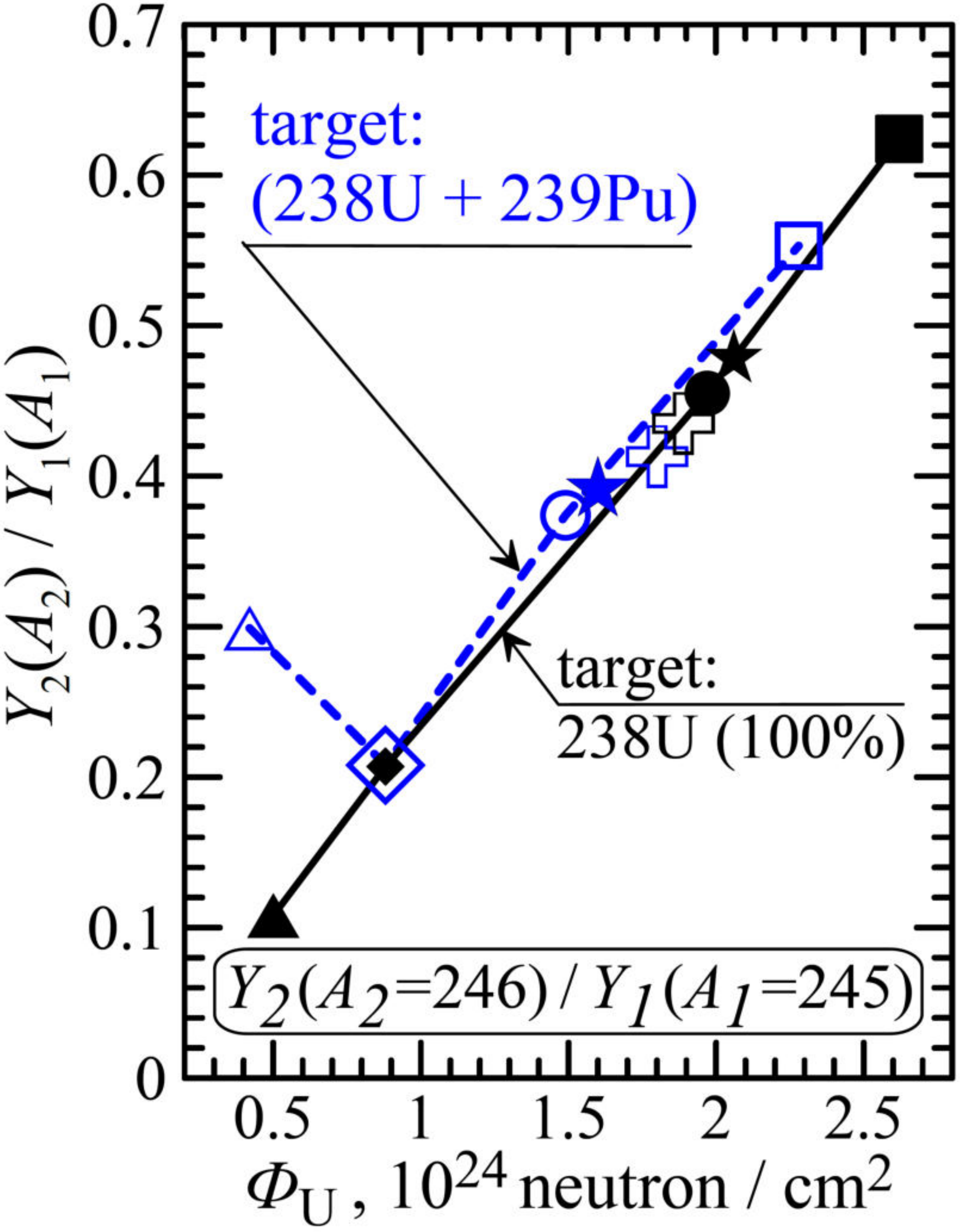
Target (100% - 238U):

▲ - **MIKE**; ◆ - **ANACOSTIA;**

● - **BARBEL**;

★ - **VULCAN;** ✙ - **KANKAKEE**

Relation of Yields Y(A=246) / Y(A=245)



Target (238U + 239Pu):

△ - **MIKE**; ◇ - **ANACOSTIA**;

○ - **BARBEL**; ★ - **VULCAN**;

✚ - **KANKAKEE**; □ - **PAR**

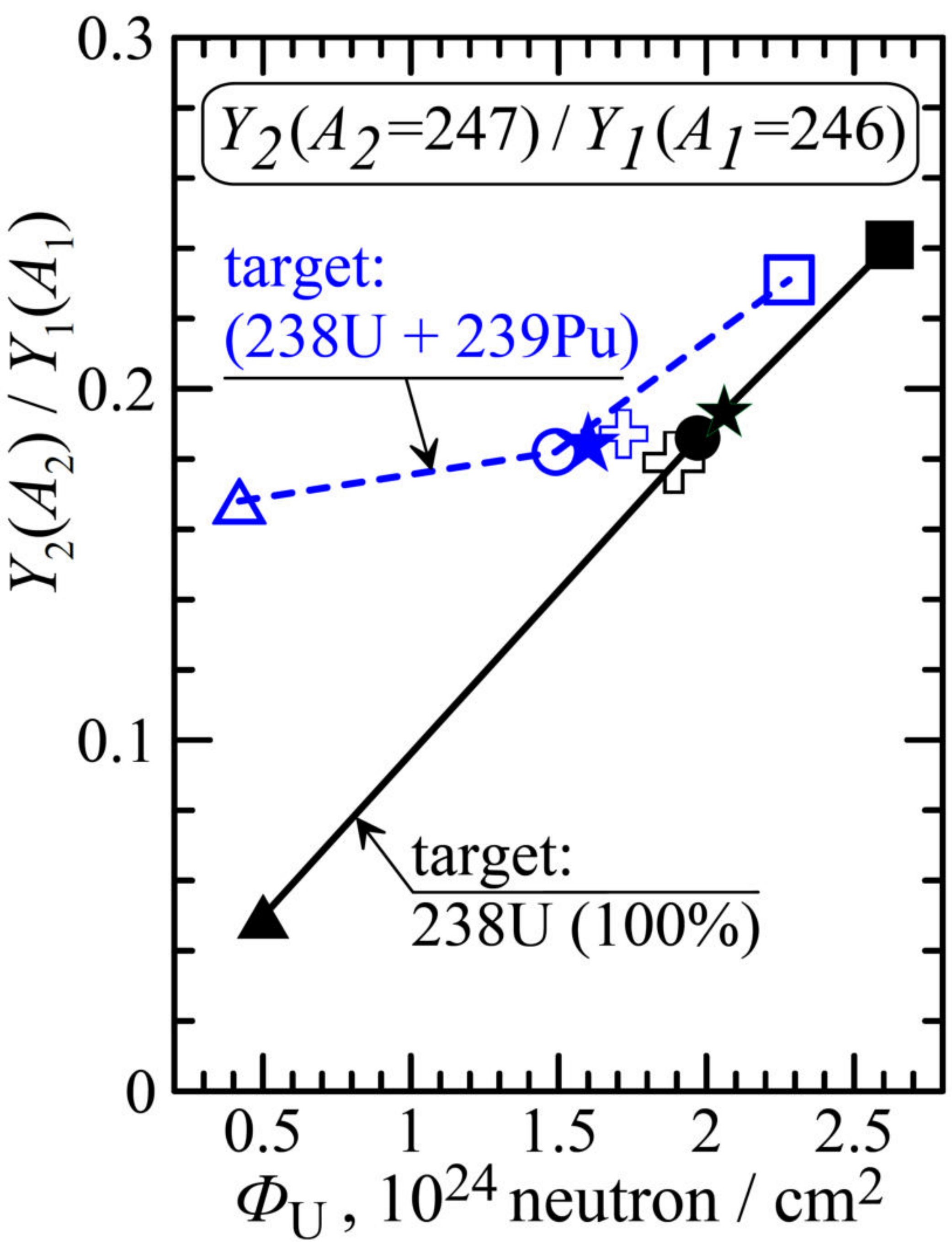
Target (100% - 238U):

▲ - **MIKE**; ◆ - **ANACOSTIA**;

● - **BARBEL**; ★ - **VULCAN**;

✚ - **KANKAKEE**; ■ - **PAR**

Relation of Yields Y(A=247) / Y(A=246)



Target (238U + 239Pu):

△ - **MIKE**;

○ - **BARBEL**; ★ - **VULCAN;**

✙ - **KANKAKEE;** □ - **PAR**

Target (100% - 238U):

▲ - **MIKE**;

● - **BARBEL**; ★ - **VULCAN;**

✙ - **KANKAKEE;** ■ - **PAR**

CONCLUSION

It were analyzed the relations of isotope yields Y(A=245)/Y(A-244), Y(A=246)/Y(A-245), Y(A=247)/Y(A-246) for the six large scale experiments : MIKE, ANACOSTIA, PAR, BARBEL, VULCAN и KANKAKEE. It were obtained the results for the model with uranium (238-U) target and for the target with addition of plutonium (239-Pu). The relations of isotope yields are approximately directly proportional to the neutron fluence at the values up to ~5E+24 n/cm^2. The most clear the dependence reveal itself for the uranium 238U-target. The such relation can be indicator of the achieved neutron fluence.

DEAR COLLEAGUES !
THANK YOU A LOT
FOR ATTENTION !