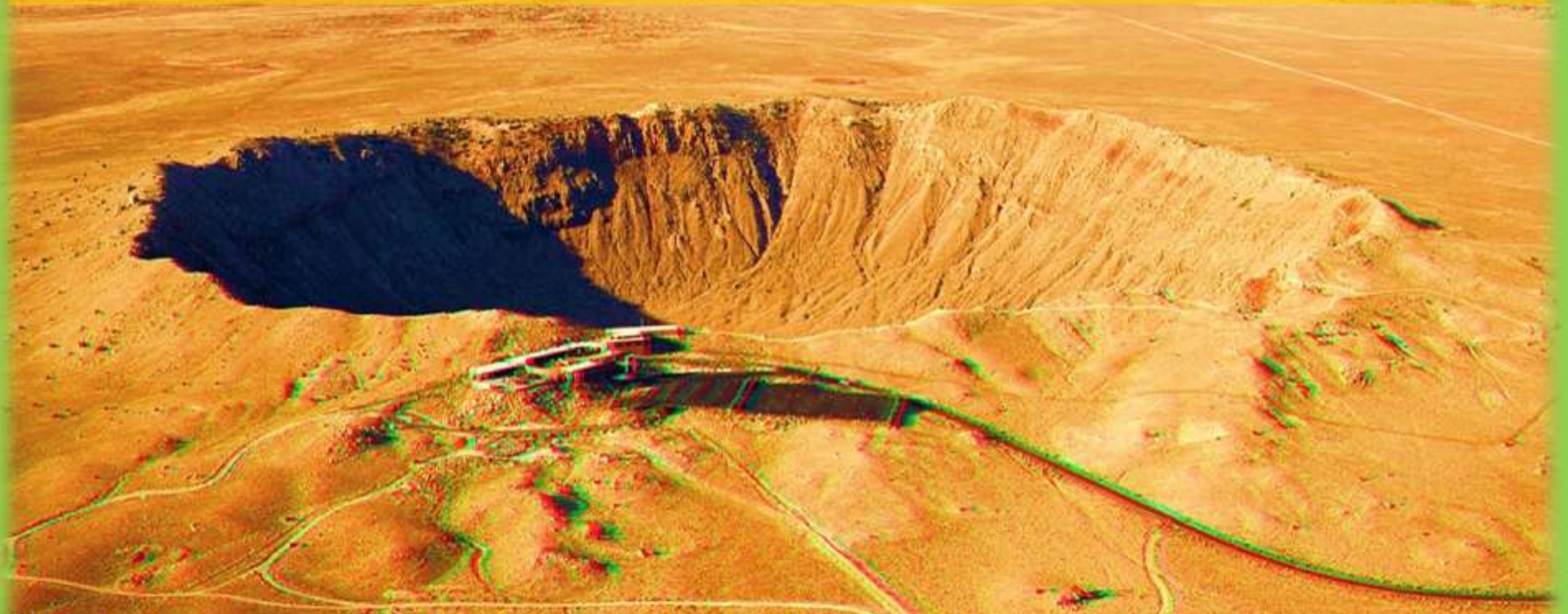
Creation of transuraniums under neutron fluxes of nuclear explosions



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

1)National Research Center "Kurchatov Institute", Moscow, Russia

2) Institute for Nuclear Research, Russian Academy of Sciences, Moscow, Russia

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Creation of transuranics under intensive neutron fluxes

Transuranium elements production in nature takes place in powerful neutron flux owing to reactions of multiple neutron capture (radiation capture) followed by β-decays. In the nature the process of rapid nucleosynthesis (the *r*-process) is realized in the explosions of supernova stars, where neutron density exceeds 10^{20} neutron/cm^3 at temperatures of ~10^9 °K.

Under artificial conditions, the *r*-process is realized in nuclear explosions, which produce neutron fluences above 10^{24} neutron/cm^2 in a time of ~10^{-6} s. Transuranium isotopes (up to ^{255}Fm) were found for the first time in the "Mike" thermonuclear explosion (Yield -10.4 Mt) in 1952.

After that a wide program for investigation of heavy and transuranium isotope production under neutron pulse process was realized (includung the PLOWSHARE Program for peaceful use of nuclear explosions).

Experiment	Date	Power (kt)	Isotope Yield up to	Comments
MIKE	November 31, 1952	10400	255	
GNOME	December 12, 1961	3.1 ± 0.5		Plowshare
ANACOSTIA	November 27, 1962	5.2	254	Plowshare
PAR	October 9, 1964	38	257	Plowshare
BARBEL	October 16, 1964	20	257	
CYCLAMEN	May 5, 1966	12	257	
KANKAKEE	June 15, 1966	20-200	257	
VULCAN	June 25, 1966	25	257	Plowshare
HUTCH	July 16, 1969	20-200	257	

Creation of transuranics under intensive neutron fluxes (2)

Nuclear and thermonuclear explosions ensure an extreme fluence ($10^{24} – 10^{25}$ neutron/cm^2) at the short exposition (~ < 10^{-6} s) and in so way is the unique instrument for investigation in nuclear physics. In the **"CYCLAMEN"** and **"HUTCH"** the obtained fluence: $1.2 \cdot 10^{25}$ and $4.5 \cdot 10^{25}$ neutron/cm^2

For compare, maximal flux for:

HFIR-reactor – $5.5 \cdot 10^{15}$ neutron/cm^2

PIK (in the central experimental channel) – $8.6 \cdot 10^{15}$ neutron/(cm^2·s)

SM-2 – $5 \cdot 10^{15}$ neutron/(cm^2·s)

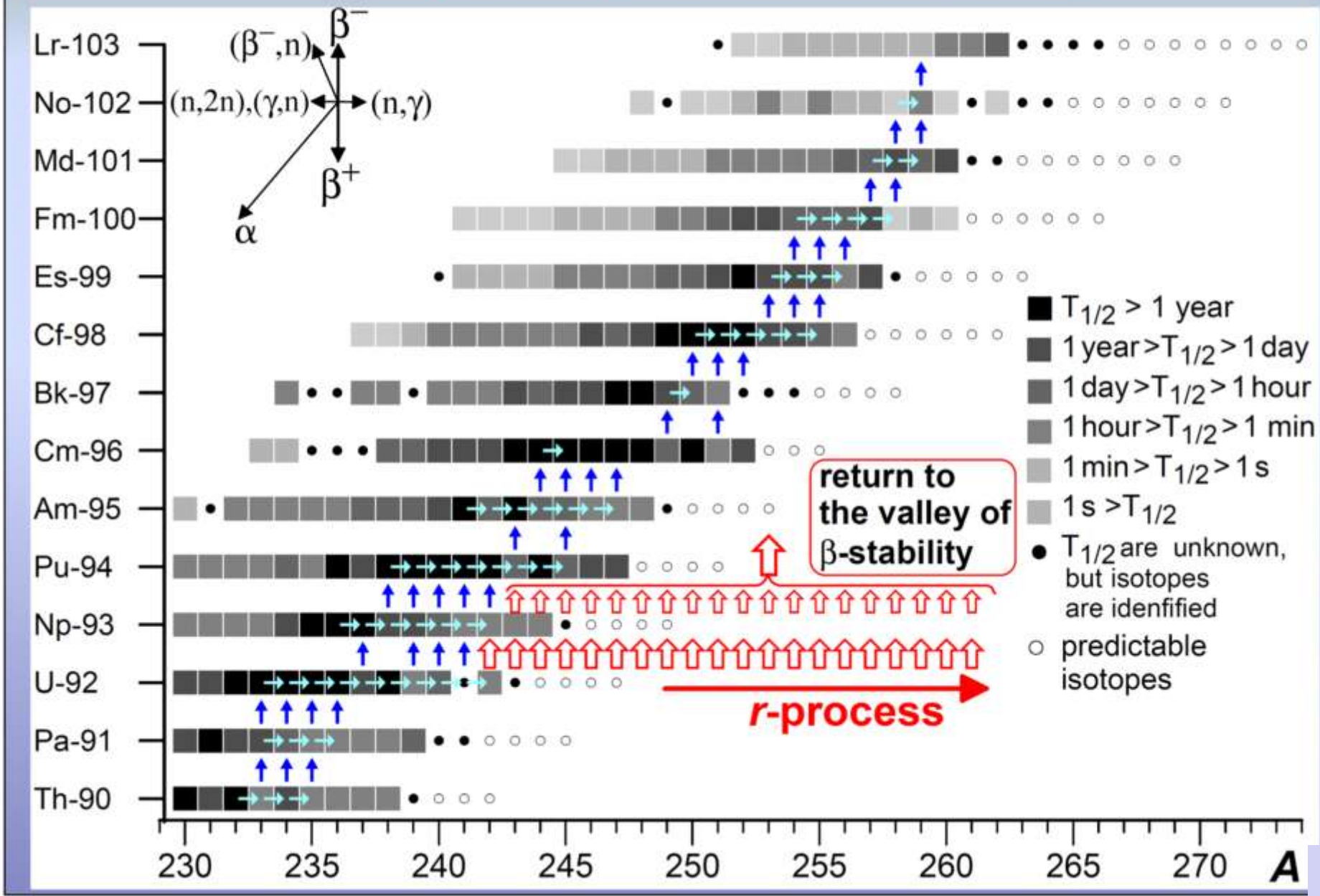
IGR (pulsed graphitic reactor) – maximal integral flux – $1 \cdot 10^{18}$ neutr./(cm^2·s)

BIGR (pulsed reactor) - $1.2 \cdot 10^{16}$ neutron/(cm^2·s) in the central channel

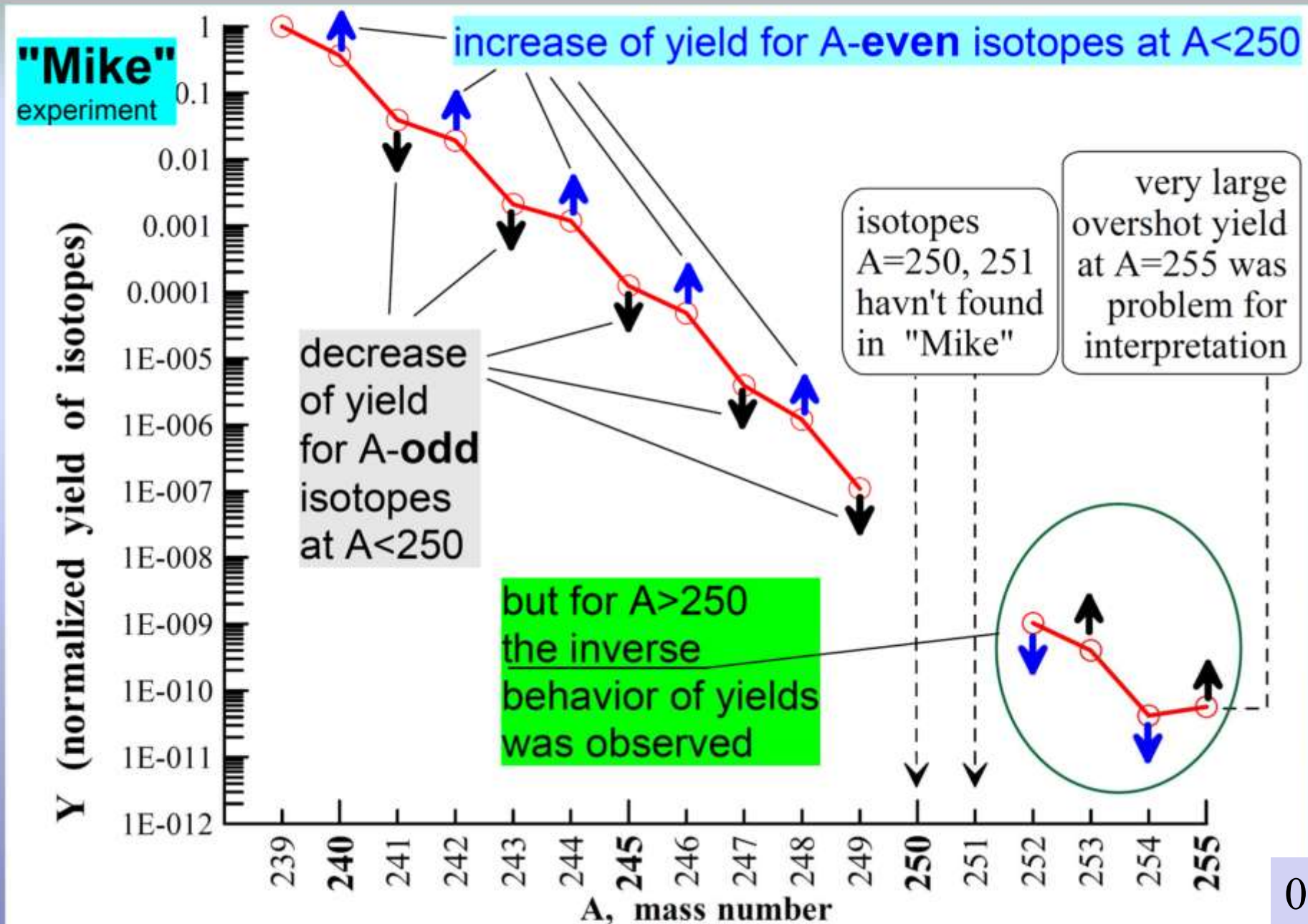
GIDRA (pulsed solution reactor) – $8 \cdot 10^{14}$ neutron/(cm^2·s)

JAGUAR (pulsed solution reactor) – $2.5 \cdot 10^{18}$ neutron/(cm^2·s) in pulse in the output channel

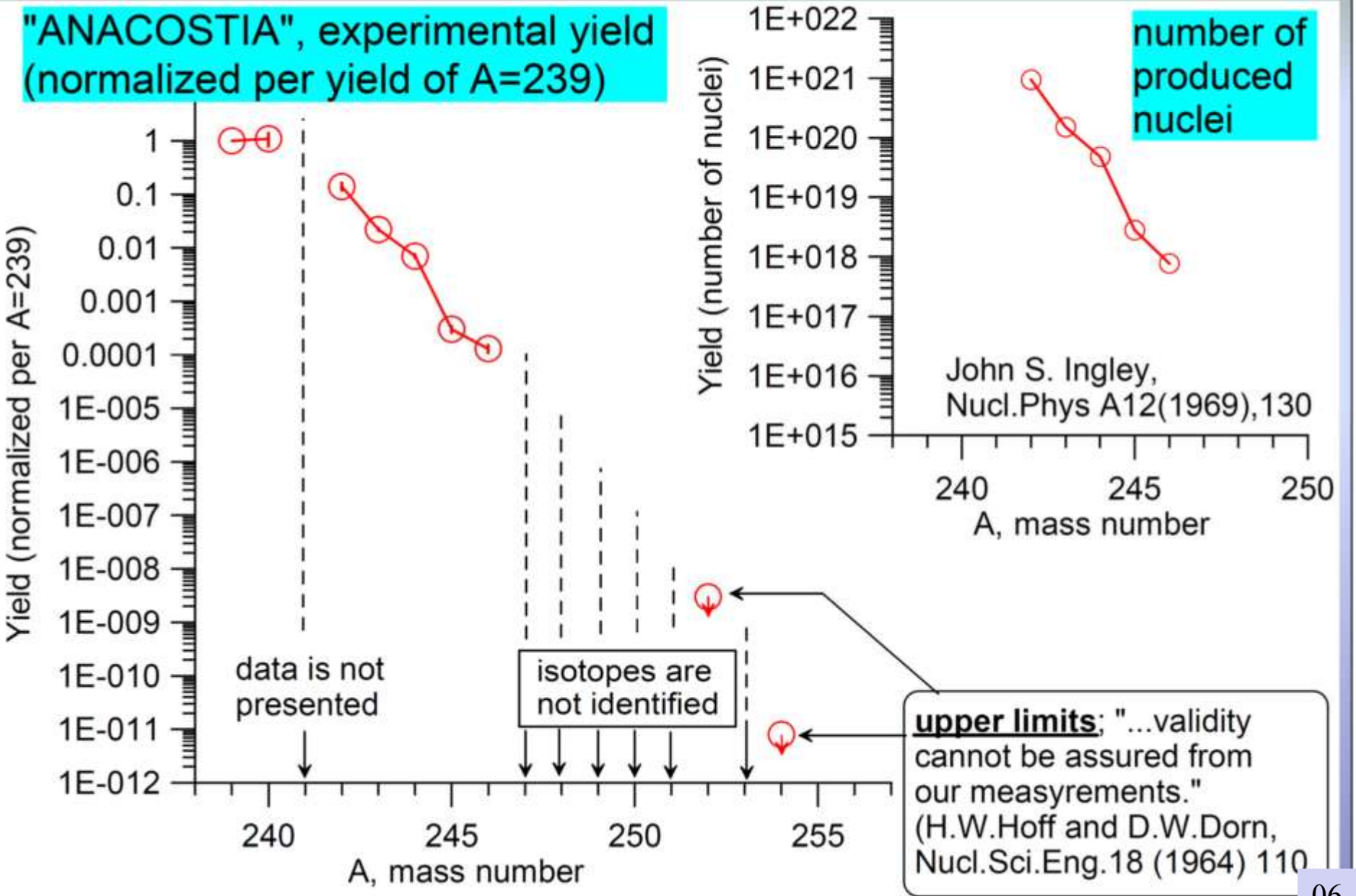
r – process (rapid-process) under condition of explosive nucleosynthesis (time interval $t \leq 10^{-6}$ s)



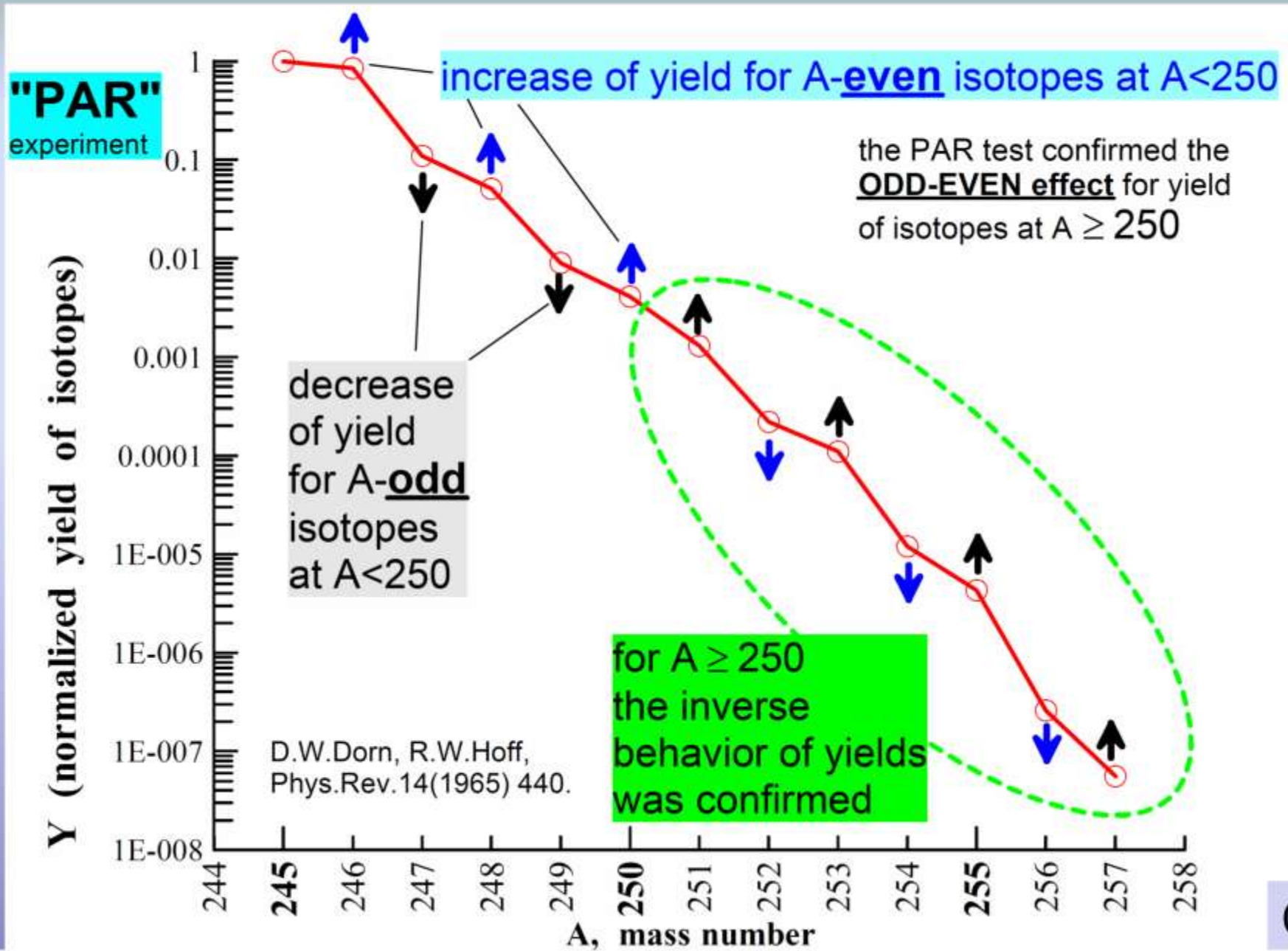
Odd-even effect in yields of transuranics under explosive nucleosynthesis



Experiment Anacostia for production of transuranics



Experiment PAR for production of transuranics



Scheme of transuranium isotopes creation

$$\partial N_z^n / \partial t = (\lambda_\beta N)_{z-1}^{n+1} + (\lambda_\alpha N)_{z+2}^{n+2} +$$

$$+ \int_0^\infty F(E,t)\left\{\left[\sigma_{n,\gamma} N\right]_z^{n-1} + \left[\sigma_{n,2n} N\right]_z^{n+1} + \left[\sigma_{n,3n} N\right]_z^{n+2}\right\} dE -$$

$$- (\lambda_\beta N)_z^n - (\lambda_\alpha N)_z^n - (\lambda_f N)_z^n -$$

$$- \int_0^\infty F(E,t)\left\{\left[\sigma_{n,\gamma} N\right]_z^n + \left[\sigma_{n,2n} N\right]_z^n + \left[\sigma_{n,3n} N\right]_z^n + \left[\sigma_{n,f} N\right]_z^n\right\} dE,$$

where z and n are the charge and the number of neutrons of the 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 respective cross-sections of the reactions; and $F(E, t)$ is the time-depended neutron flux.

V. I. LYASHUK. TAKING INTO CONSIDERATION THE DYNAMICS AT CREATION OF TRANSURANIUM ISOTOPES UNDER THE CONDITIONS OF NUCLEAR EXPLOSION. Preprint ITEP-7-97. Moscow, 1997;
http://lss.fnal.gov/archive/other/itep-7-97.pdf
Yu. S. Lutostanskii, V. I. Lyashuk, and I. V. Panov, Calculation of Transuranium Element Synthesis in Intensive Neutron Fluxes under Adiabatic Conditions. Bull. Russ. Acad. Sci.: Phys. 74, 504 (2010).
V. I. Lyashuk, Bull. Russ. Acad. Sci.: Phys. 76, 1182 (2012).

Simplification of the scheme

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 (Δ_t -the exposition time)

$$\int_0^{\Delta t}\int_E F(E,t)\,dE\,dt = \Delta t \int_E \widetilde{F}(E)\,dE = \Phi \quad ,$$

Hereinafter. in the calculation it is used the neutron flux Φ [*neutron* /cm^2] integrated for a time and possessed the fixed energy in the interval ~20+30 keV (ie., one-group energy representation) according to the process temperature.

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

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

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

One group approximation

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

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

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 in calculating the rate of radiative capture $\lambda_{n,\gamma}^{n+i}$

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

where A is the mass number; 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.

Adiabatic approximation

The described model of transuranium isotope creation upon multiple neutron captures based on pulsed nucleosynthesis can be extended by the process dynamics, which comprises variations of the (n, γ)–reaction cross section upon an environmental T-temperature drop during the adiabatic expansion that follows termination of the chain reaction. 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 (239Pu or another target) 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 single-group 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 υ remains constant for any moment of time $t \in [t_A, t_B]$;

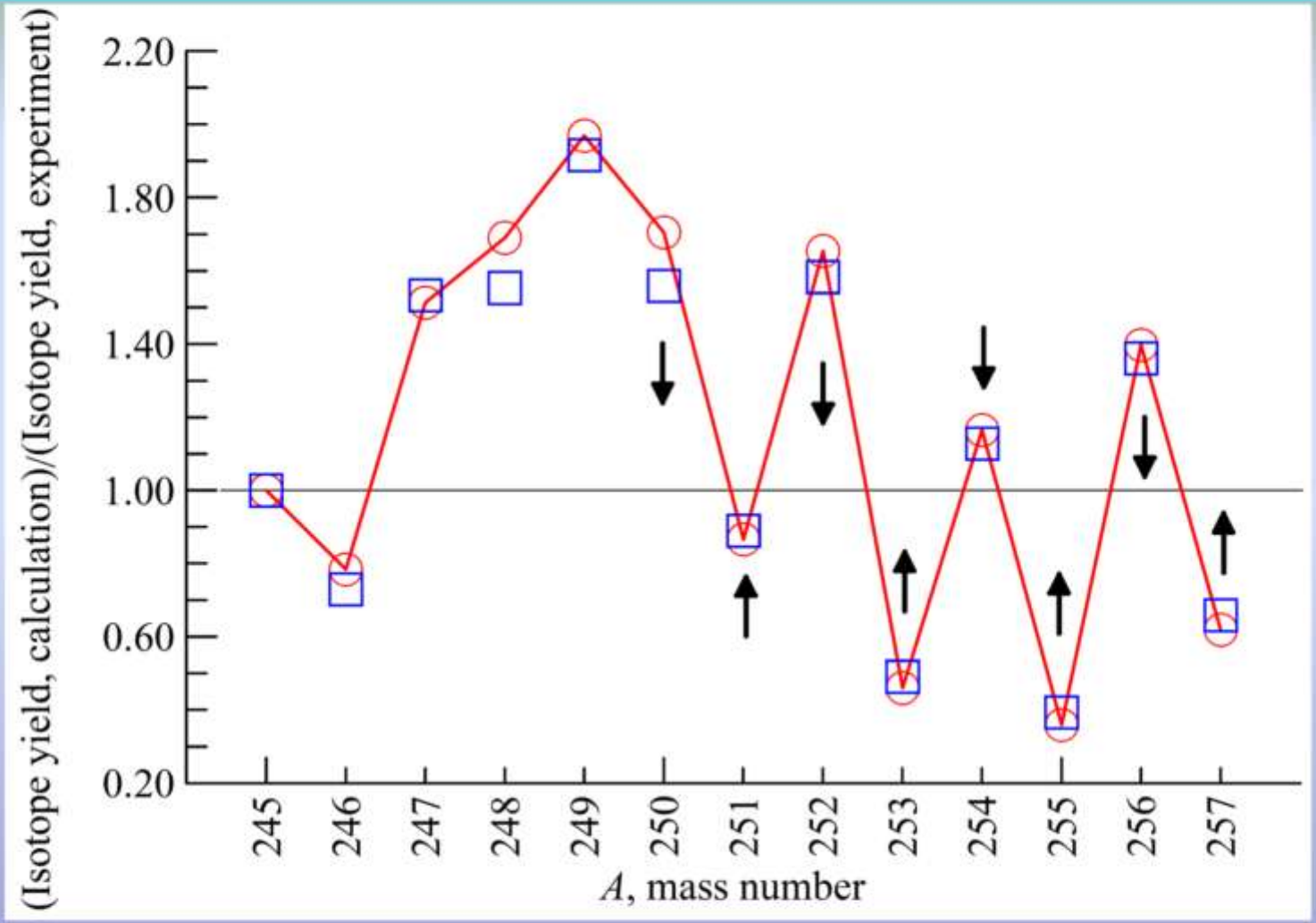
(3) to specify the adiabatic index γ upon the adiabatic expansion of V-volume:

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

It was assumed that the multiple captures proceed up to the moment $t = 10^{-6}$ s, and the chain reaction lasts 3×10^{-7} s; the area with fissile material is spherical in shape with an initial radius R_0 and the possible interval of adiabatic index: γ=1.5-1.6.

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 for each given time step $\Delta t_1, \Delta t_2, \ldots, \Delta t_m$.

Adiabatic approximation. Experiment PAR



Red line – simulation without adiabatic approximation (static model) for the 238U-target; root mean square value(rms) relative to the experiment δ=53.3%

☐ - simulation with adiabatic approximation: γ = 1.5, δ=48.5%

Adiabatic approximation with slow neutron flux component

The presence of hydrogen bearing substances (e.g., $C_6H_2(NO_2)_3CH_3$) in the experimental facility leads to the rapid moderation of neutrons; a considerable part of the neutron flux irradiating a target made of the initial isotope (or mixer of isotopes) will therefore consist of a soft component.

Calculations for the neutron leakage spectrum in our experimental facility were performed for various points of time in the range of $t = (0–3.6) \times 10^{-7}$ s (H.A. Sandmair, S.A. Dupree, G.E. Hansen. Nucl. Sci.Eng. V48 (1972) 142). By the time $t = 3.6 \times 10^{-7}$ s, the neutron leakage in the range $E_n = (9–26)$ keV over a time interval of 10^{-8} s was ≥≈0.3 in the total leakage flux.

In the model it is assumed that the total neutron flux in the target (containing the start isotope) consists of two fluxes (slow and fast) – F_{sl} and F_{fa} (**gross model for two groups**).

For two group system we have:

$$\lambda_{sl} \equiv \lambda_{n,\gamma}(T_{sl}) \quad \text{and} \quad \lambda_{fa} \equiv \lambda_{n,\gamma}(T_{fa})$$

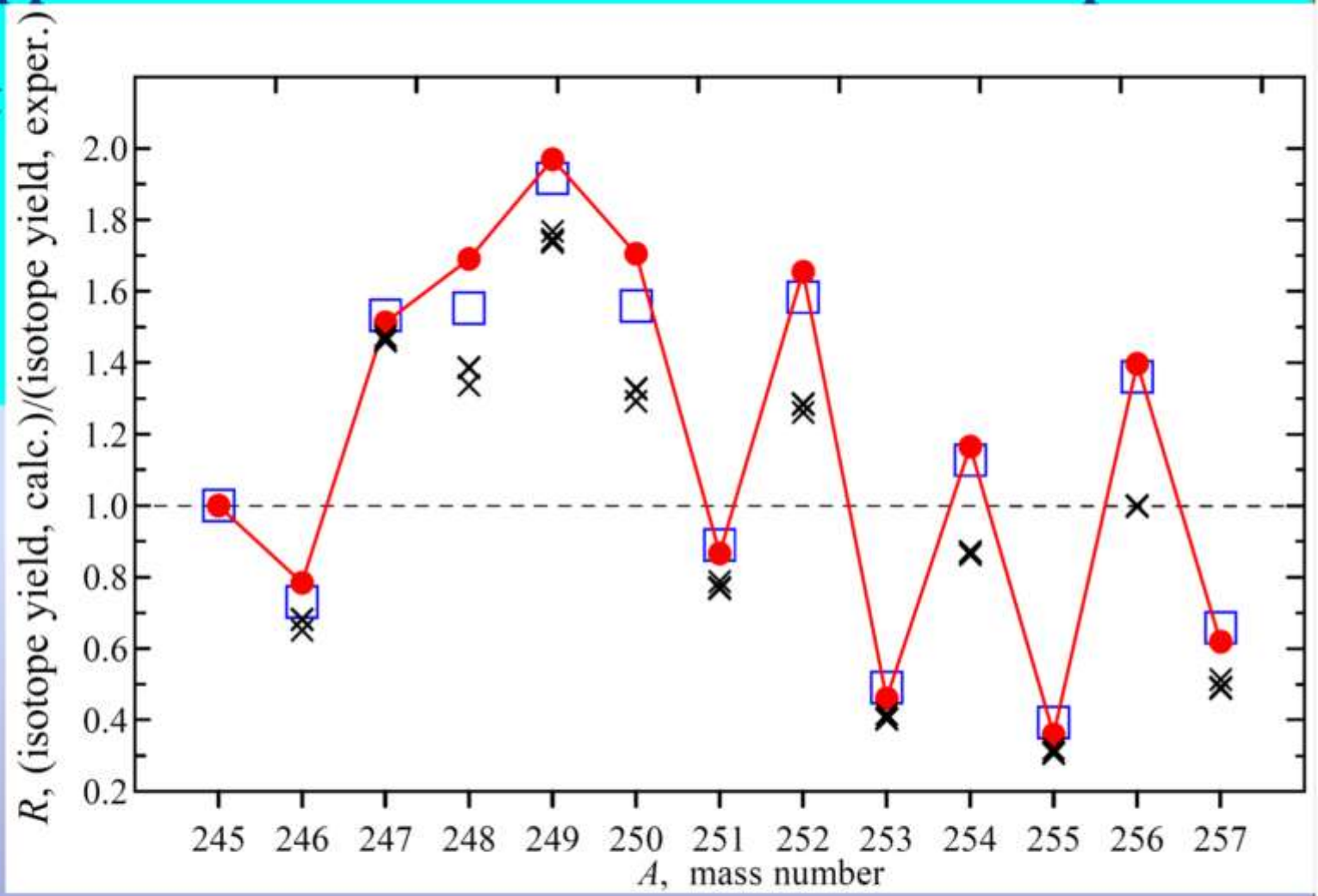
$$\begin{cases} \dot{N}_z^{\,n} = -[(\lambda_{sl}+\lambda_{fa})N]_z^n \\ \dot{N}_z^{\,n+1} = [(\lambda_{sl}+\lambda_{fa})N]_z^n - [(\lambda_{sl}+\lambda_{fa})N]_z^{n+1} \\ \cdot \\ \cdot \\ \cdot \\ \dot{N}_z^{\,n+i} = [(\lambda_{sl}+\lambda_{fa})N]_z^{n+i-1} - [(\lambda_{sl}+\lambda_{fa})N]_z^{n+i} \end{cases}$$

Lyashuk, V.I., Preprint ITEP-46-98

V. I. Lyashuk, Simulating Transuranium Isotope Yields upon Explosive Nucleosynthesis with Allowance for Elements of Process Dynamics. **Bull. Russ. Acad. Sci.: Phys. 76, 1182 (2012).**

Adiabatic approximation with slow neutron flux component.

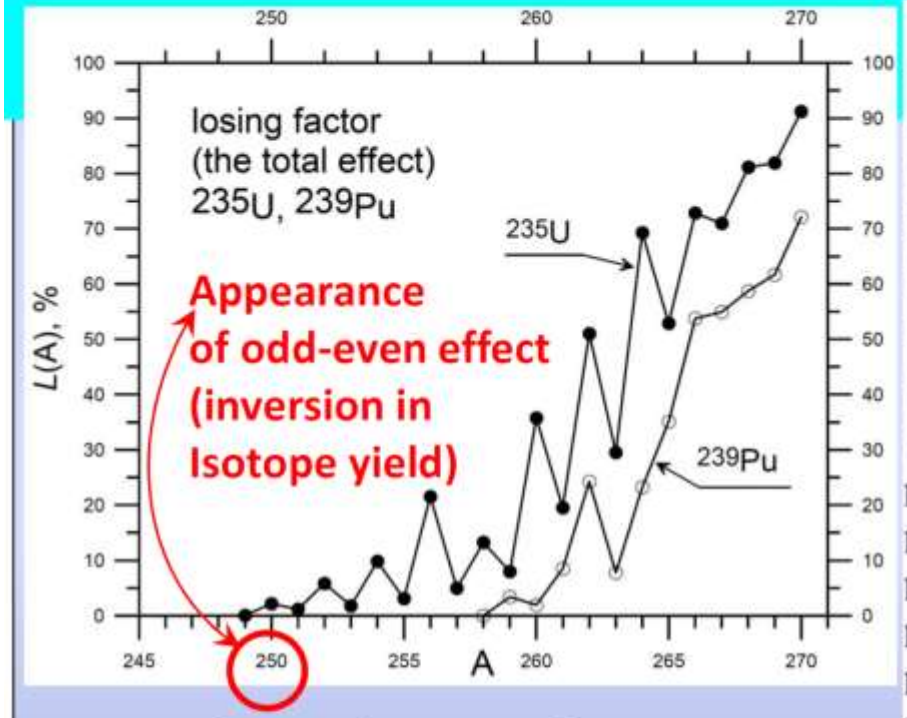
Experiment PAR



Red line – simulation without adiabatic approximation (static model) for the 238U-target; root mean square value(rms) relative to the experiment δ=53.3%; □ - simulation with adiabatic approximation (one-group approximation), γ = 1.5, δ=48.5%.

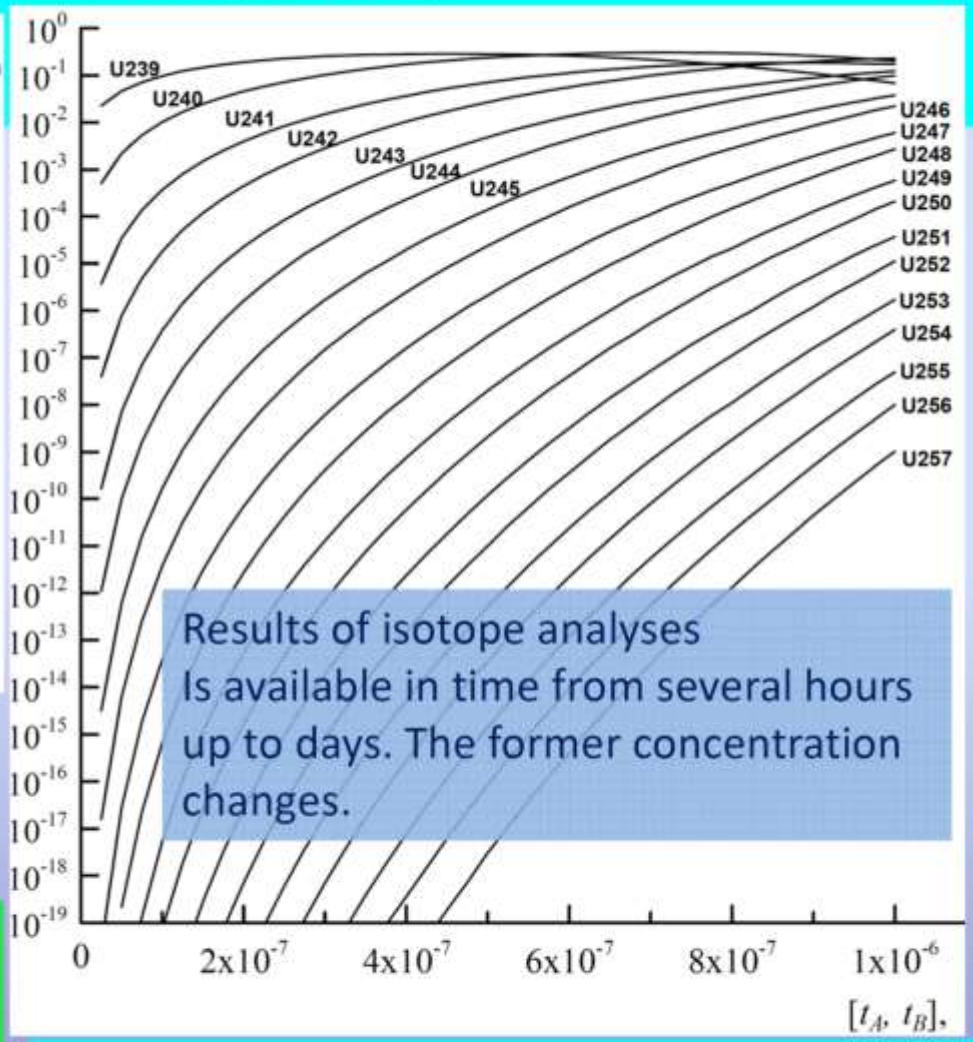
× - adiabatic approximation for two group model, γ = 1.5, 1.6; δ=42.6%.

Creation in *r*-process and decrease of isotope concentration (losing-factor). (U+Pu)-target ABM model (Adiabatic Binary Model)



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.

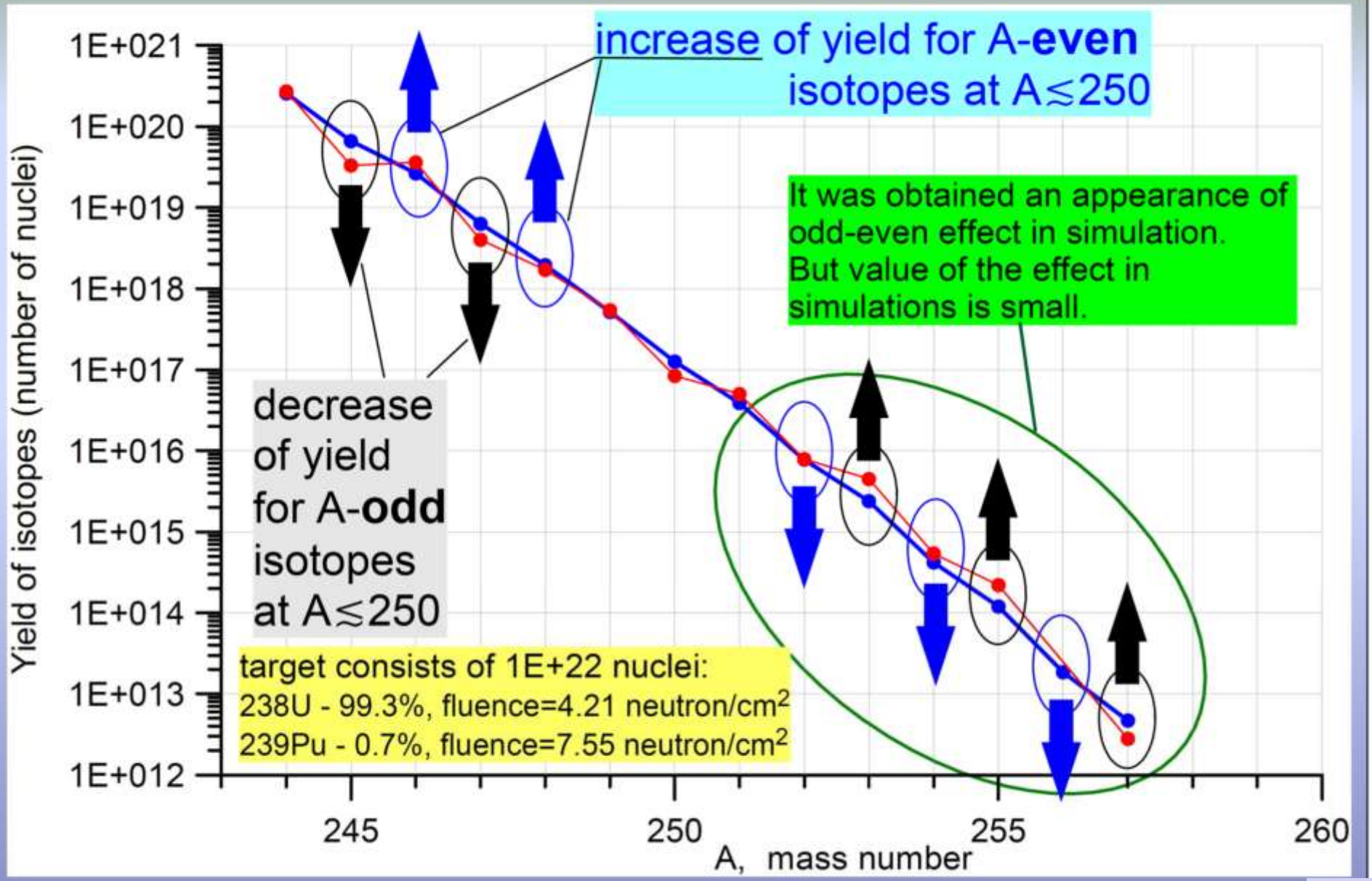


Time dependence of isotope creation during the interval of nucleosynthesis , (s)

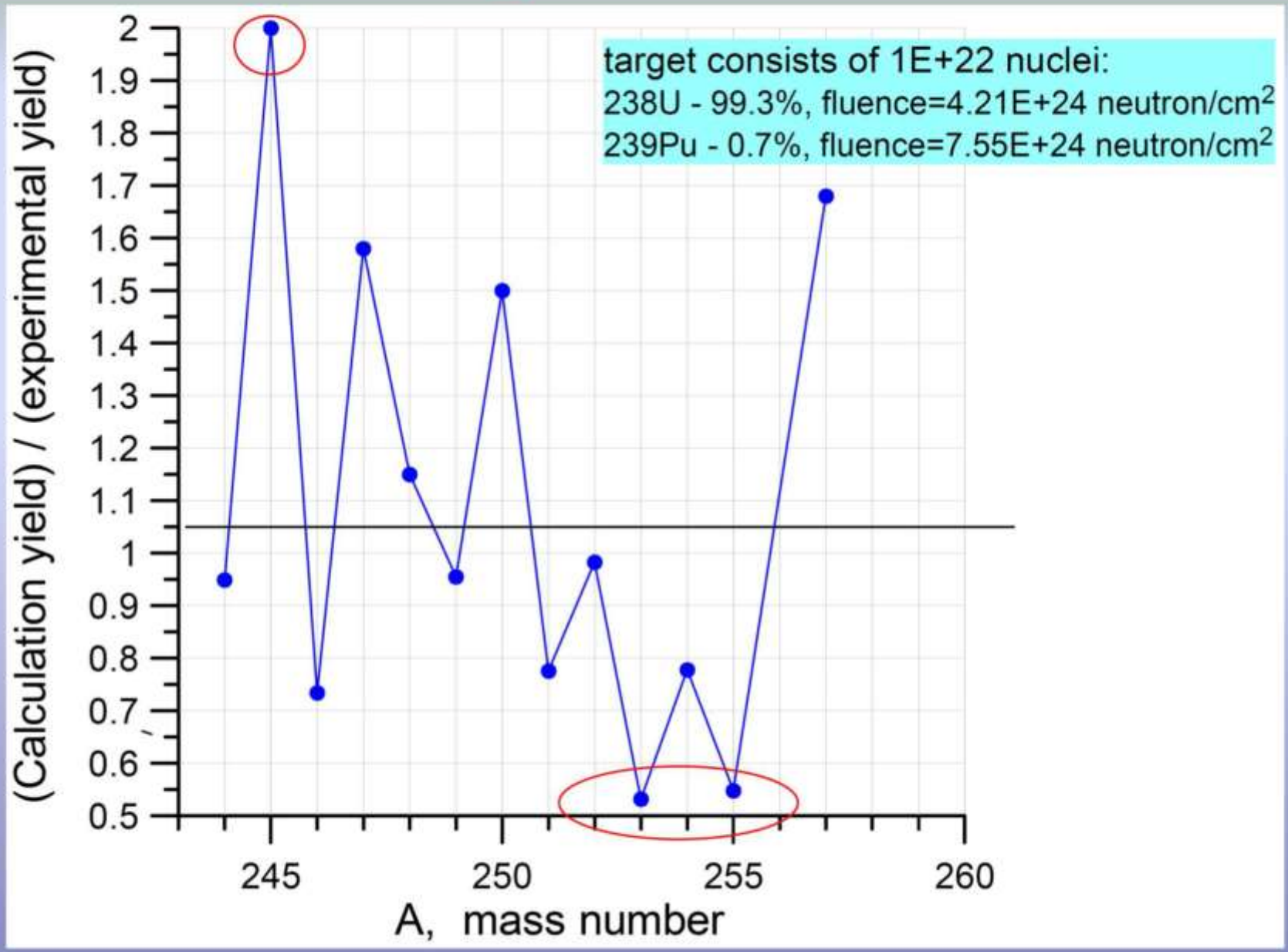
Yu.S. Lutostansky, and V.N. Tikhonov, Bull. Russ. Acad. Sci. Phys. 76, 534 (2012)

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

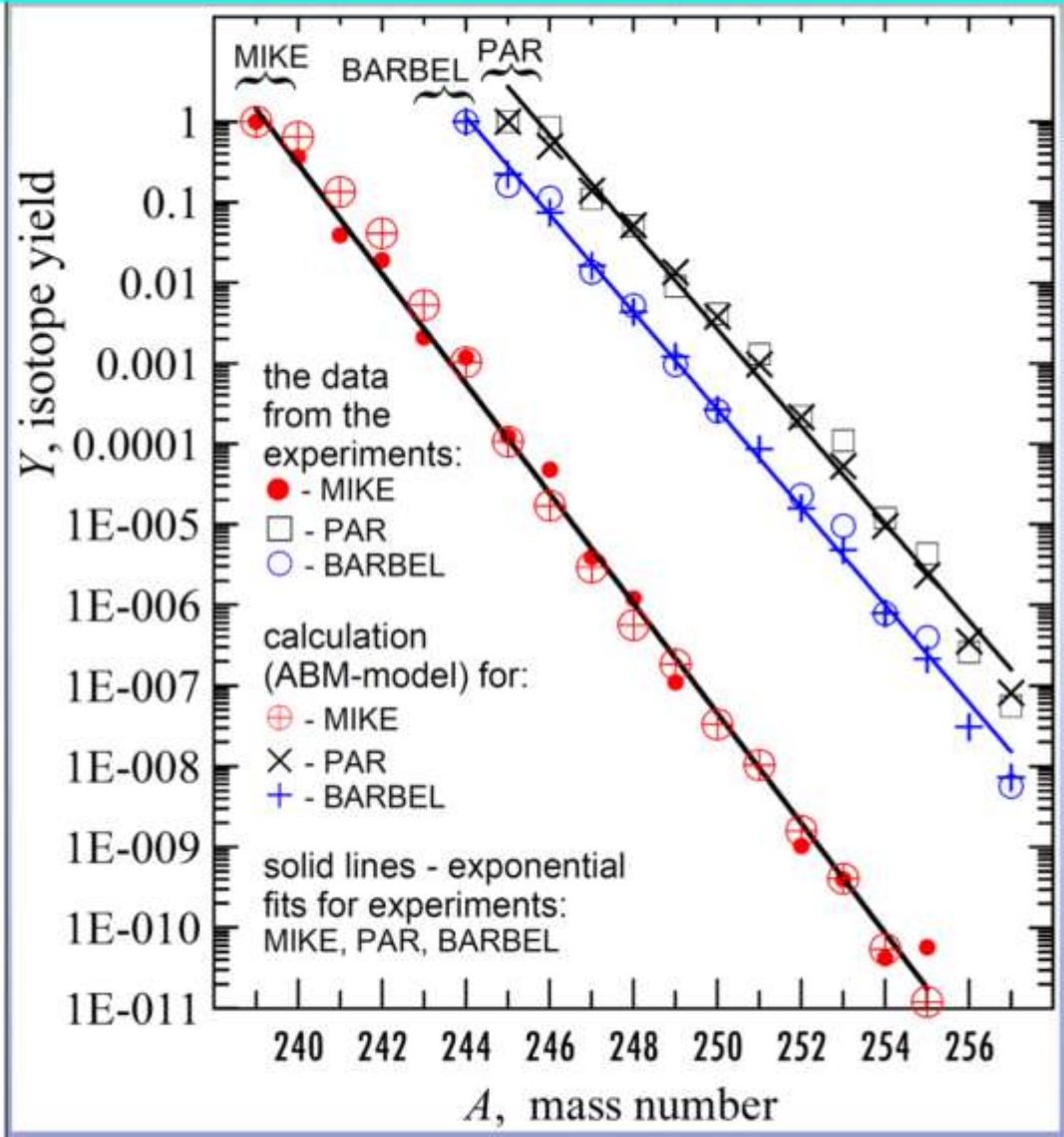
Experiment Vulcan. Odd-even effect



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



Isotope yields in MIKE, PAR and BARBEL experiments. Simulation results



The decreasing dependence of $Y(A)$ is fitted as follows:

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

$i = 1$ ("Mike") $A_1 = 239$,

$b_1 = 1.570$, $c_1 = 375.491$ (1a)

$i = 2$ ("Barbel") $A_2 = 244$,

$b_2 = 1.395$, $c_2 = 340.584$ (1b)

$i = 3$ ("Par") $A_3 = 245$,

$b_3 = 1.388$, $c_3 = 341.015$ (1c)

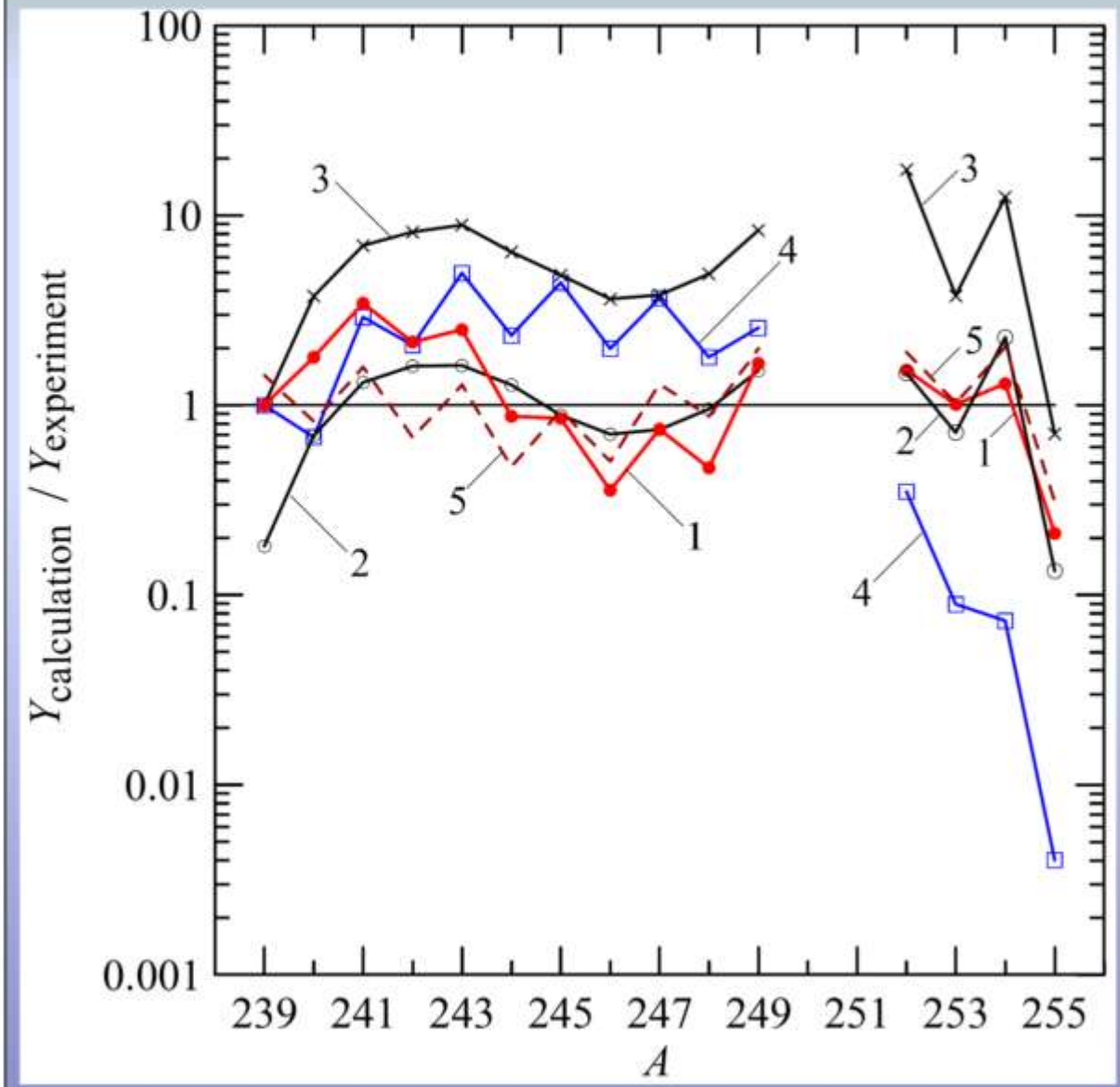
The standard deviations of this approximation are: $\delta_1 = 56\%$ ("Mike"), $\delta_2 = 60.2\%$ ("Barbel"), $\delta_3 = 86.8\%$ ("Par"),

which are better than many previous calculations and comparable to the accuracy of our calculations of the presented ABM model (Adiabatic Binary Model)

(see Table for Mike, Par, Barbel yield)

Yu. S. Lutostansky, and V. I. Lyashuk. Production of Transuranium Nuclides in Pulsed Neutron Fluxes from Thermonuclear Explosions. *JETP Letters, 2018, Vol. 107, No. 2, p. 79.*

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



1. RED line (ABM model)
Target:238U - 99.993%,
fluence – 1.55E+24 neutr/cm^2;
239Pu – 0.007%,
fluence=4.94 neutr./cm^2;
δ %=91%.

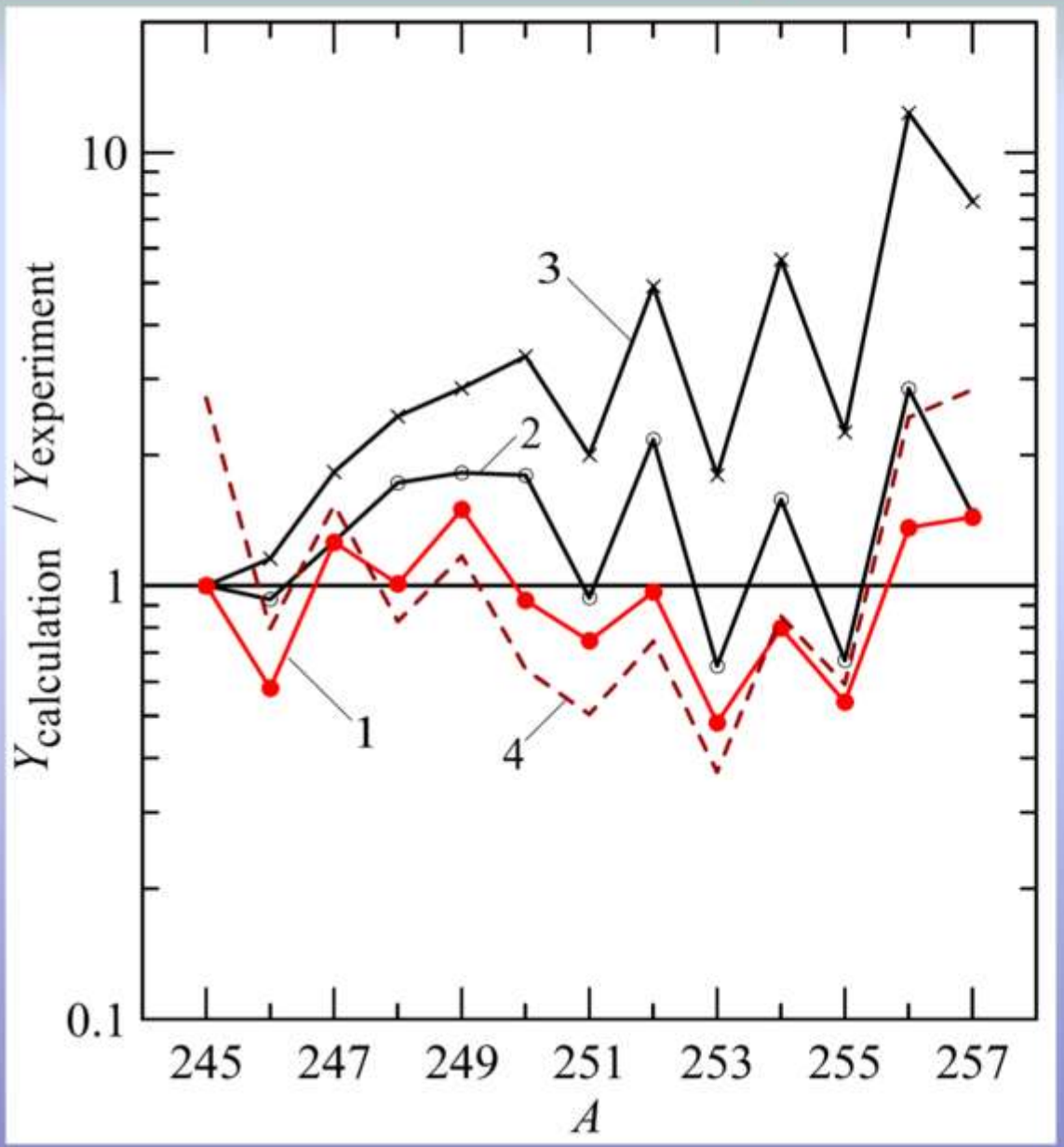
2. Dorn D. W. *Phys. Rev.* 1962. V. 126. p. 693; without normalization on $Y(A=239)_{calc}$;

3. Dorn D. W. *Phys. Rev.* 1962. V. 126. p. 693; with normalization on $Y(A=239)_{calc}$;
δ %=681%.

4. Zagrebaev V. I., Karpov A. V., Mishustin I. N., Greiner W. *Phys. Rev.* C. 2011. V. 84. 044617;
δ %=180%.

5. $Y(A_i)/ Y(A=239) = \exp\{- b \cdot A_i + c\}$,
b=1.395, c=340.584;
δ %=60.2%.

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



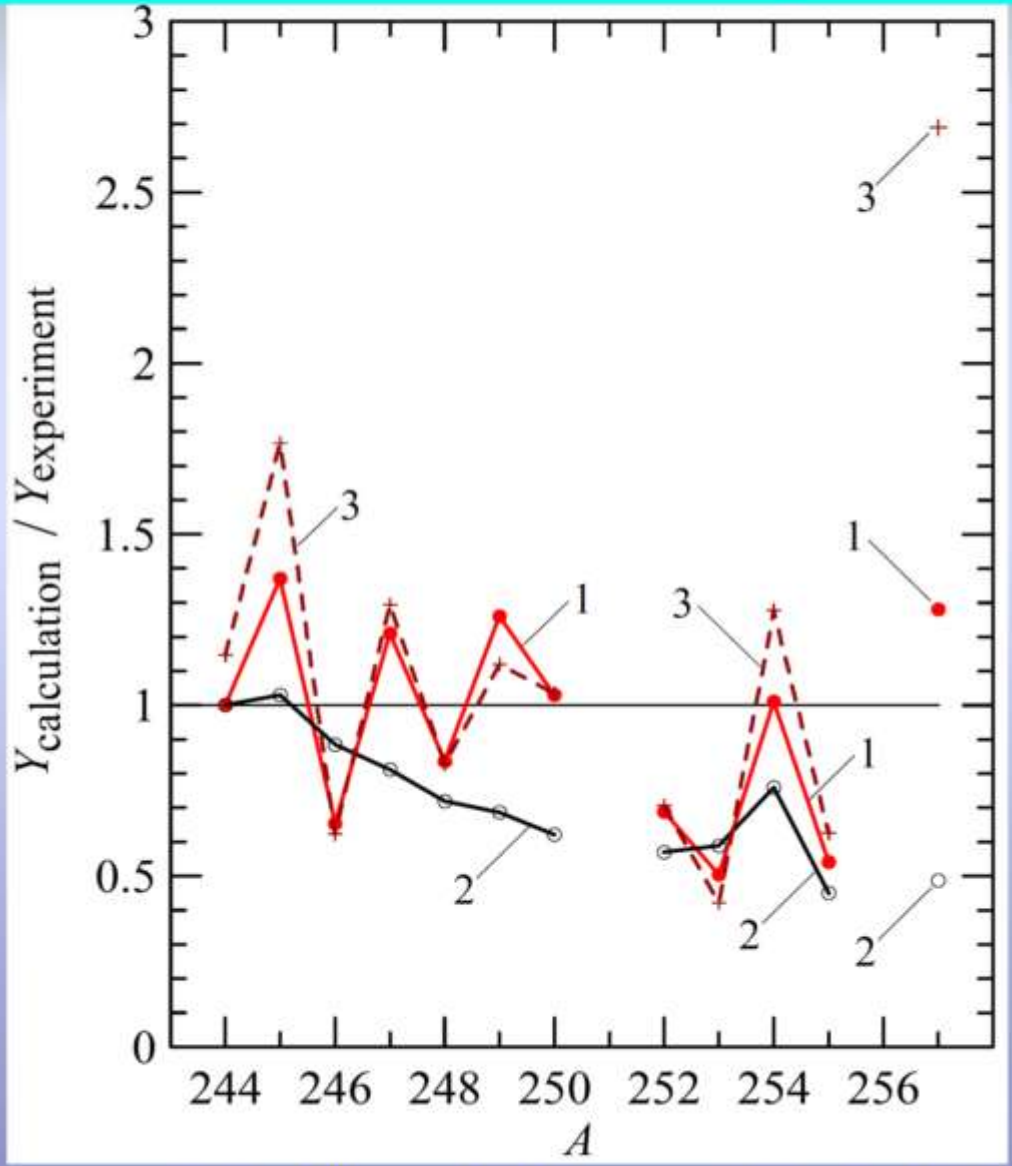
1. RED line (ABM model)
Target:238U - 97%,
fluence – 5.31E+24 neutr/cm^2;
239Pu – 3%,
fluence=6.38 neutr./cm^2;
δ =32.9%.

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

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



1. RED line (ABM model)
Target:238U - 99.6%,
fluence – 3.50E+24 neutr/cm^2;
239Pu – 0.4%,
fluence=6.08 neutr./cm^2;
δ %=29.3%.

2. Bell G. I. *Phys. Rev.* B. 1965. V. 139. p. 1207; δ =33.5%.

3. $Y(A_i)/ Y(A=244) = \exp\{- b \cdot A_i + c\}$,
b=1.395, c=340.584;
δ %=60.2%.

Y. S. Lutostansky and V. I. Lyashuk, Nucleosynthesis of Heavy Elements in Thermo-nuclear Explosions "Mike", "Par" and "Barbell". Proc. The 3rd Int. Conf. on Part. Phys. and Astrophys., KnE Energy & Physics, pp. 57–64 (2018).

https://knepublishing.com/index.php/KnE-Energy/article/view/1723/3993

CONCLUSION

It was developed the static and dynamic (adiabatic) models for production of transuranium isotopes under the condition of explosive nucleosynthesis. The models allow to use the target with complex isotopes composition (Adiabatic Binary Model).

The model include the data on delay decays of created neutron rich transuranic isotopes (which are taken into account by the "losing factor").

It were realized calculation of transuranium isotope production for several experiments (MIKE, ANACOSTIA, PAR, BARBEL, VULCAN) with (U+Pu)-target (i.e., we based on the idea of target component mixing during the explosive nucleosynthesis).

In simulation it was demonstrated that the model allows to calculate the yield within the factor of ~ "two" and "work" in the "right direction" at $A \geq 250$, where appears the odd-even effect.

Thank you a lot for attention !

Большое спасибо

за внимание !