

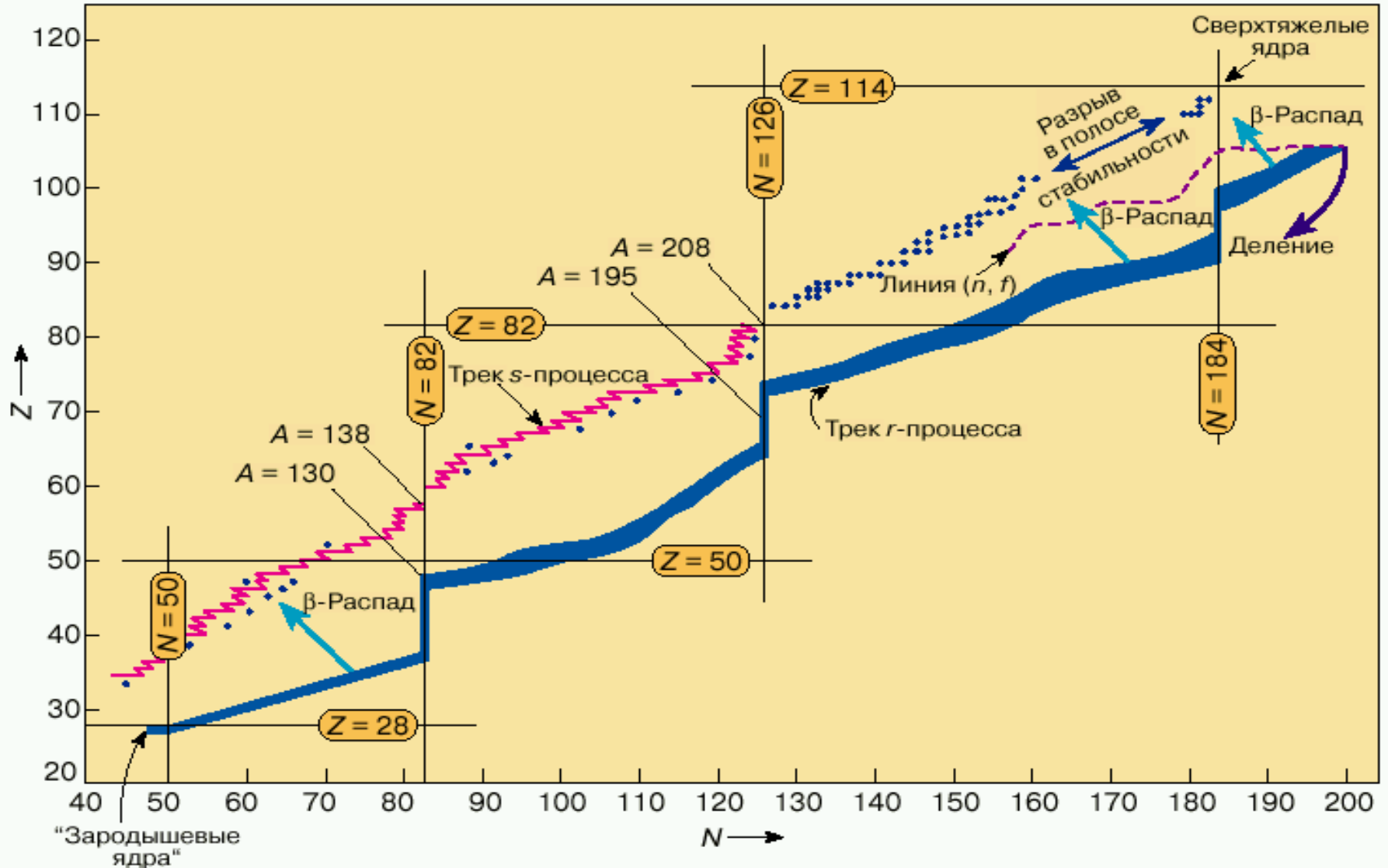
# TRANSFERMIUM NEUTRON-RICH NUCLEI PRODUCTION IN PULSED NEUTRON FLUXES OF NUCLEAR EXPLOSIONS

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# PROCESSES OF NUCLEOSYNTHESIS.



The tracks of elements synthesis in s (slow)- and r (rapid)- processes.

# NUCLEOSYNTHESIS OF THE HEAVY NUCLEI

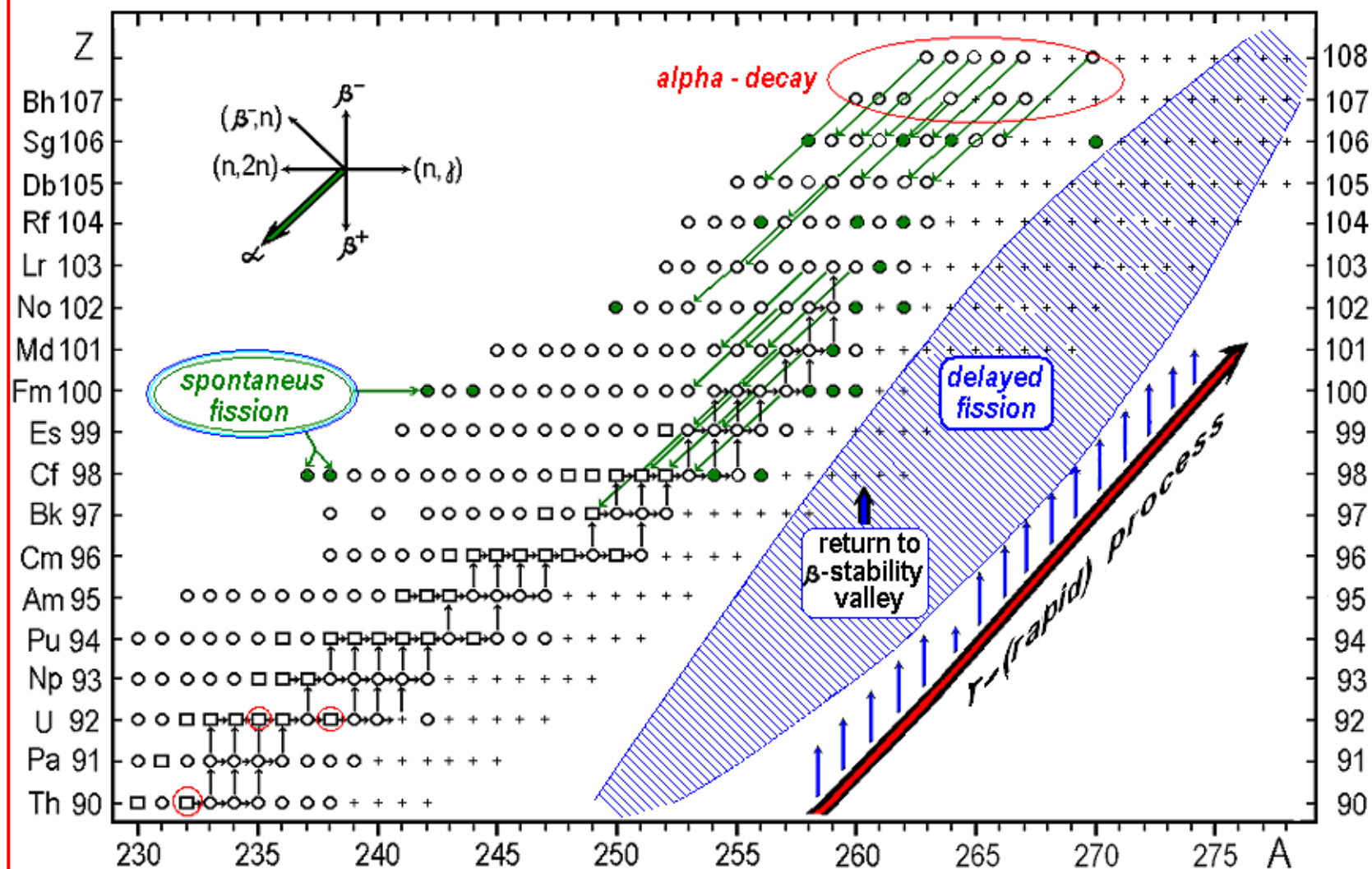


Схема образования актинидов в медленном ( $s$ -процесс) и быстром ( $r$ -процесс) нуклеосинтезе;  $\square$  – ядра с  $T_{1/2} \geq 1$  г ;  $\circ$  – ядра с  $T_{1/2} < 1$  г ;  $+$  - прогнозируемые нейтронно-избыточные ядра из базы данных NDS JAEA (Япония).



# I - METHOD: r -Process equations for the concentration calculations

Concentrations  $n(A,Z)$  are changing in time (may be more than 4000 equations):

$$\begin{aligned} \frac{dn(A, Z)/dt = & -\lambda_{\beta}(A, Z) \cdot n(A, Z) - \lambda_{n\gamma}(A, Z) \cdot n(A, Z) + \lambda_{\gamma n}(A+1, Z) \cdot n(A+1, Z) + \\ & + \lambda_{n\gamma}(A-1, Z) \cdot n(A-1, Z) - \lambda_{\gamma n}(A, Z) \cdot n(A, Z) + \\ & + \lambda_{\beta}(A, Z-1) \cdot n(A, Z-1) \quad P_{\beta}(A, Z-1) + \lambda_{\beta}(A+1, Z-1) \cdot n(A+1, Z-1) \quad P_{1n}(A+1, Z-1) + \\ & + \lambda_{\beta}(A+2, Z-1) \cdot n(A+2, Z-1) \quad P_{2n}(A+2, Z-1) + \lambda_{\beta}(A+3, Z-1) \cdot n(A+3, Z-1) \quad P_{3n}(A+3, Z-1) + \\ & + \Phi_{\nu}(A, Z) + F_f(A, Z), \end{aligned}$$

$\lambda_{n\gamma}$  and  $\lambda_{\gamma n}$  — rates of  $(n,\gamma)$  and  $(\gamma,n)$  -reactions,  $\lambda_{\beta} = \ln(2/T_{1/2})$  —  $\beta$ -decay rate,  $P_{\beta}$  - probability of  $(A, Z)$  nuclide creation after  $\beta$ -decay of  $(A, Z-1)$  nuclide. Branching coefficients of isobaric chains -  $P_{1n}, P_{2n}, P_{3n}$  corresponds to probabilities of one-, two- and three- neutrons emission in  $\beta$ - decay of the neutron-rich nuclei; the total probability of the delayed neutrons emission is the sum:

$$P_n = \sum_k P_{kn}$$

$F_f(A, Z)$  describes fission processes.

Neutrino capturing processes are not included ( $\Phi_{\nu}(A, Z) = 0$ )

**Inner time scale is strongly depends on the nuclear reactions rates.**

# NUCLEOSYNTHESIS WAVE MOVEMENT

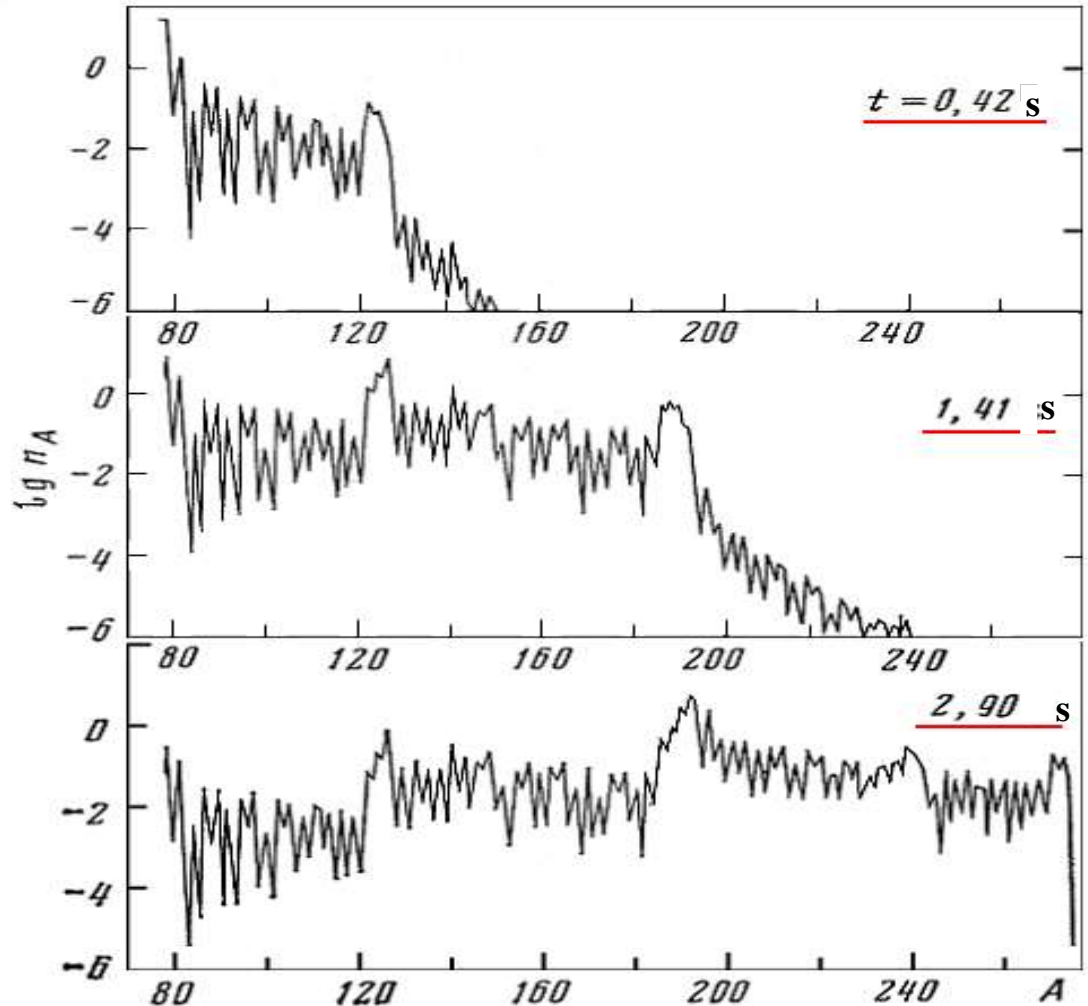
Concentrations:

$$n_A = \sum_Z n(A, Z)$$

for three time moments  
calculated for r-process  
conditions

$$n_n = 10^{24} \text{ cm}^{-3}, T_9 = 1. = 10^9 \text{ K}$$

Лютостанский Ю.С., Птицин  
Д.А., Синюкова О.Н.,  
Филлипов С.С., Чечеткин  
В.М. «Образование  
элементов с  $A > 80$  в  
нейтронном потоке при  
астрофизических условиях». *Ядерная Физика* 1985, т. 42, с.  
215.



# β-Delayed processes in very neutron-rich nuclei

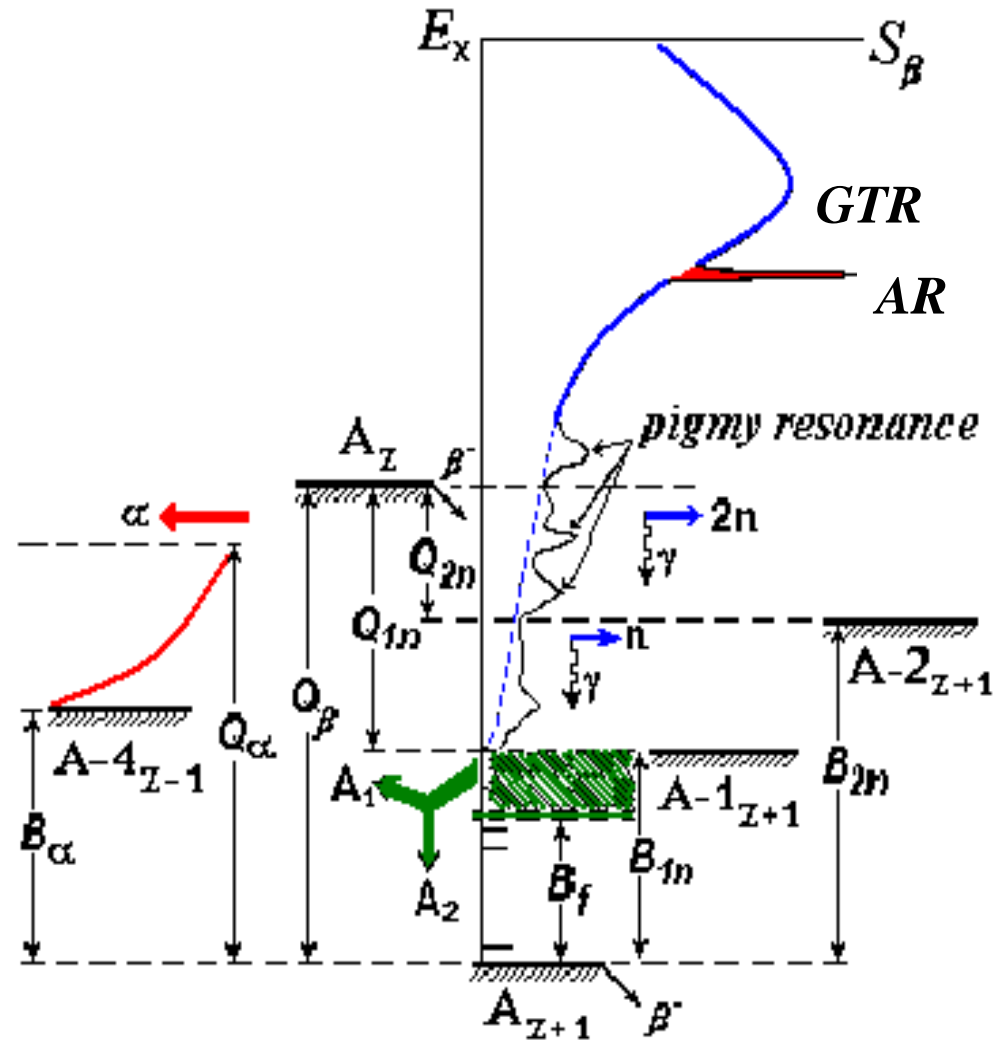
Delayed neutrons emission -  $(\beta, n)$

Multi-neutron  $\beta$  - delayed emission -  $(\beta, kn)$

$\beta$  - delayed  $\alpha$  - emission -  $(\beta, \alpha)$

$\beta$  - delayed protons emission -  $(\beta, p)$

$\beta$  - delayed fission -  $(\beta, f)$



# Beta – Delayed Multy-Neutron Emission

**Probability for 2n - emission:**

$$P_{2n} = \frac{\int_0^{Q_\beta} \sum_{ijlm} f(Z+1, Q_\beta - E) S_\beta^i(E) W_2^{ijl}(E_n, E_m) dE}{\int_0^{Q_\beta} \sum_i f(Z+1, Q_\beta - E) S_\beta^i(E) dE}$$

**Probability for kn - emission:**

$$P_{kn} = \int_{B_{kn}}^{Q_\beta} \int_0^{Q_{kn}} I_\beta(U) W_n(U, E) dU dE$$

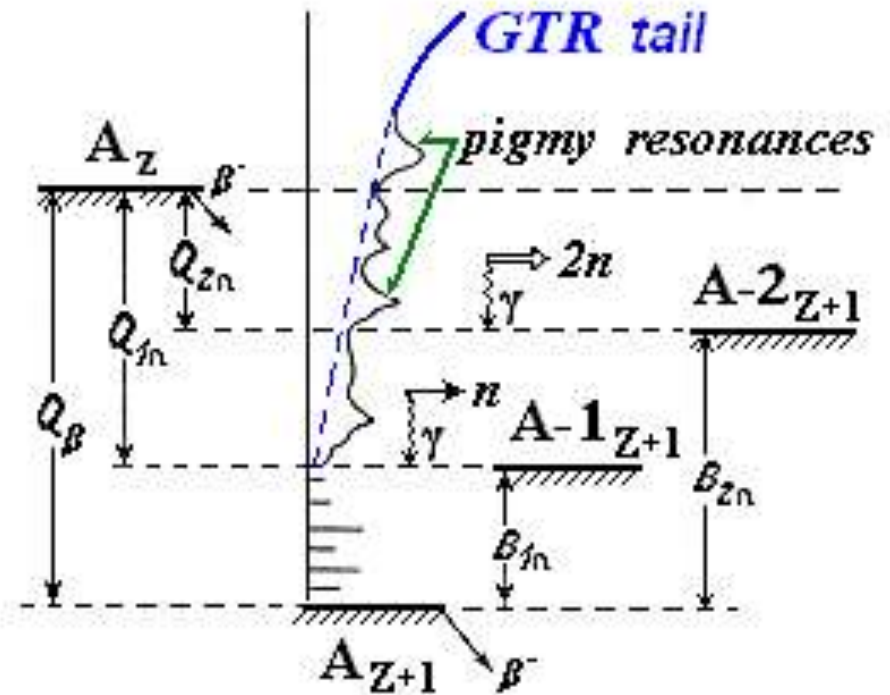
$U, I_\beta(U)$  – energies and intensities in the daughter nucleus,

$W_n(U, E)$  – probability of neutron emission:

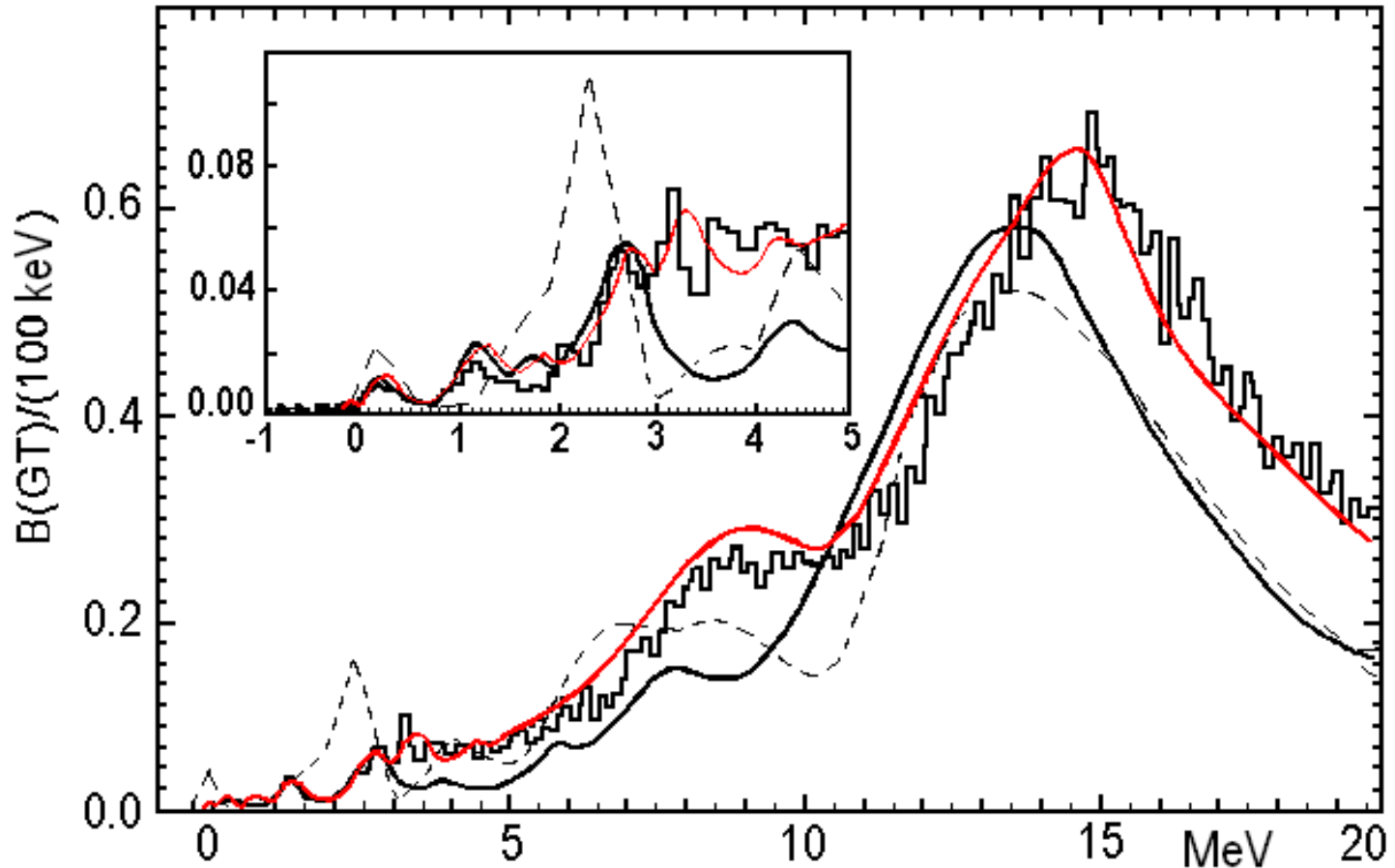
$$W_n(U, E) = \frac{T_n(E) q_f(U - E - B_n)}{\int_0^{U - B_n} T_n(E') q_f(U - E' - B_n) dE' + 2\pi q_i(U) \Gamma_\gamma(U)}$$

$q_i$  and  $q_f$  – level densities of compound and final nucleus,

$T_n(E)$  — transitivity factor



# BETA-STRENGTH FUNCTION FOR $^{127}\text{Xe}$



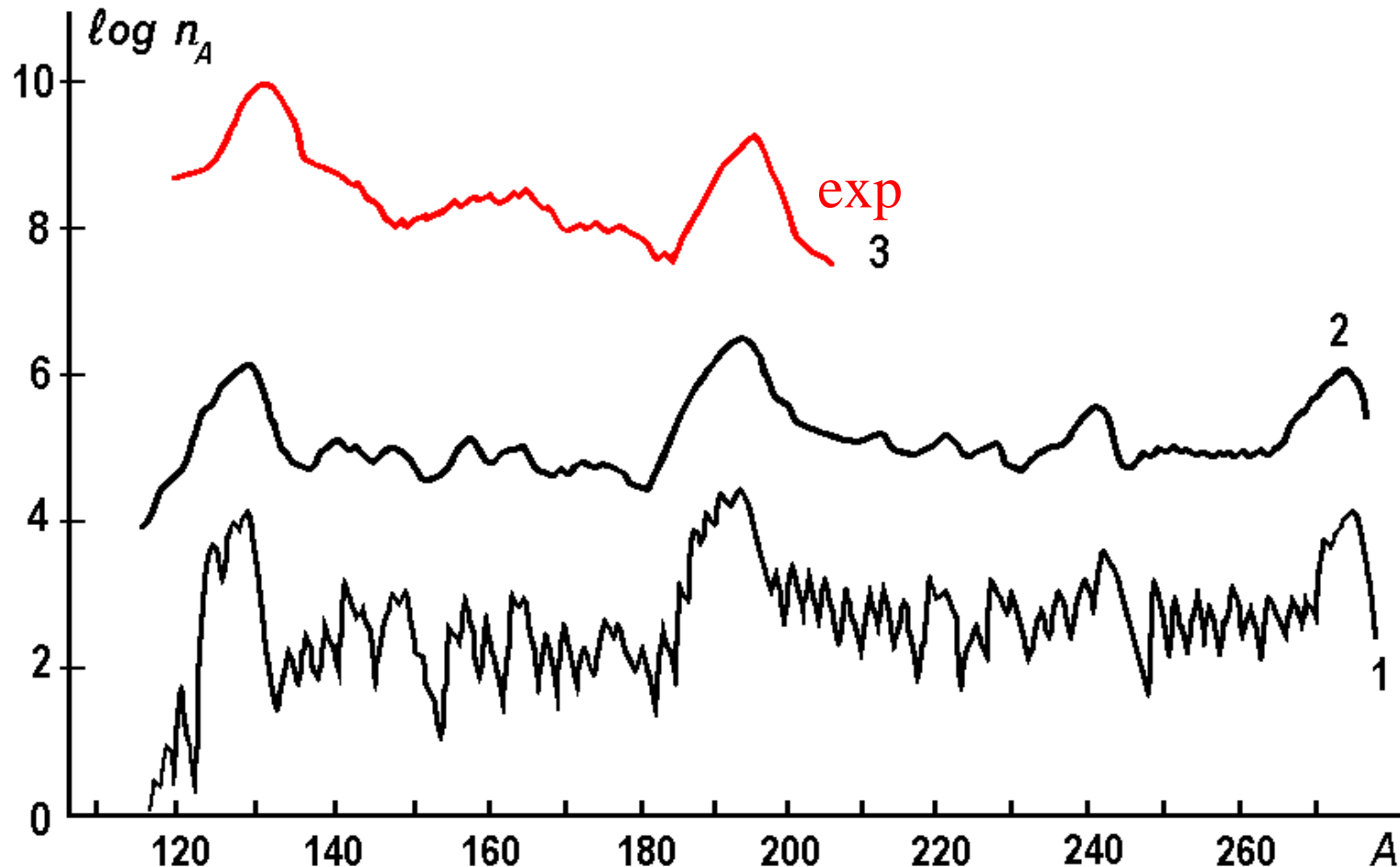
1 - Breaking line – experimental data (1999): M. Palarczyk, *et. al.* Phys. Rev. 1999. V. 59. P. 500;

2 – Solid red line TFFS calculations with  $e_q = 0.9$  ;

3 - Solid black line – calculations with  $e_q = 0.8$  : Yu.S. Lutostansky, N.B. Shulgina. Phys. Rev. Lett. 1991. V.67. P. 430;



# BETA-DELAYED NEUTRONS IN NUCLEOSYNTHESIS



Calculated abundancies: 1– with out (d,n)-effect; 2 – with (d,n)-effect; in the relative units ( $T=10^9$  K,  $n_n=10^{24}$  cm $^{-3}$ ).

Расчет: Лютостанский Ю.С., Панов И.В., Синюкова О.Н., Филлипов С.С., Чечеткин В.М.  
Роль запаздывающих нейтронов при образовании элементов в r –процессе. ЯФ. 1986. т.44. с.66.

# DYNAMIC MODEL OF NUCLEOSYNTHESIS

Yields of Nuclei,  
calculated in static  
(curve 2) and  
dynamic pulse r –  
processes  
(curves 3 and 4).

Numbers denote:

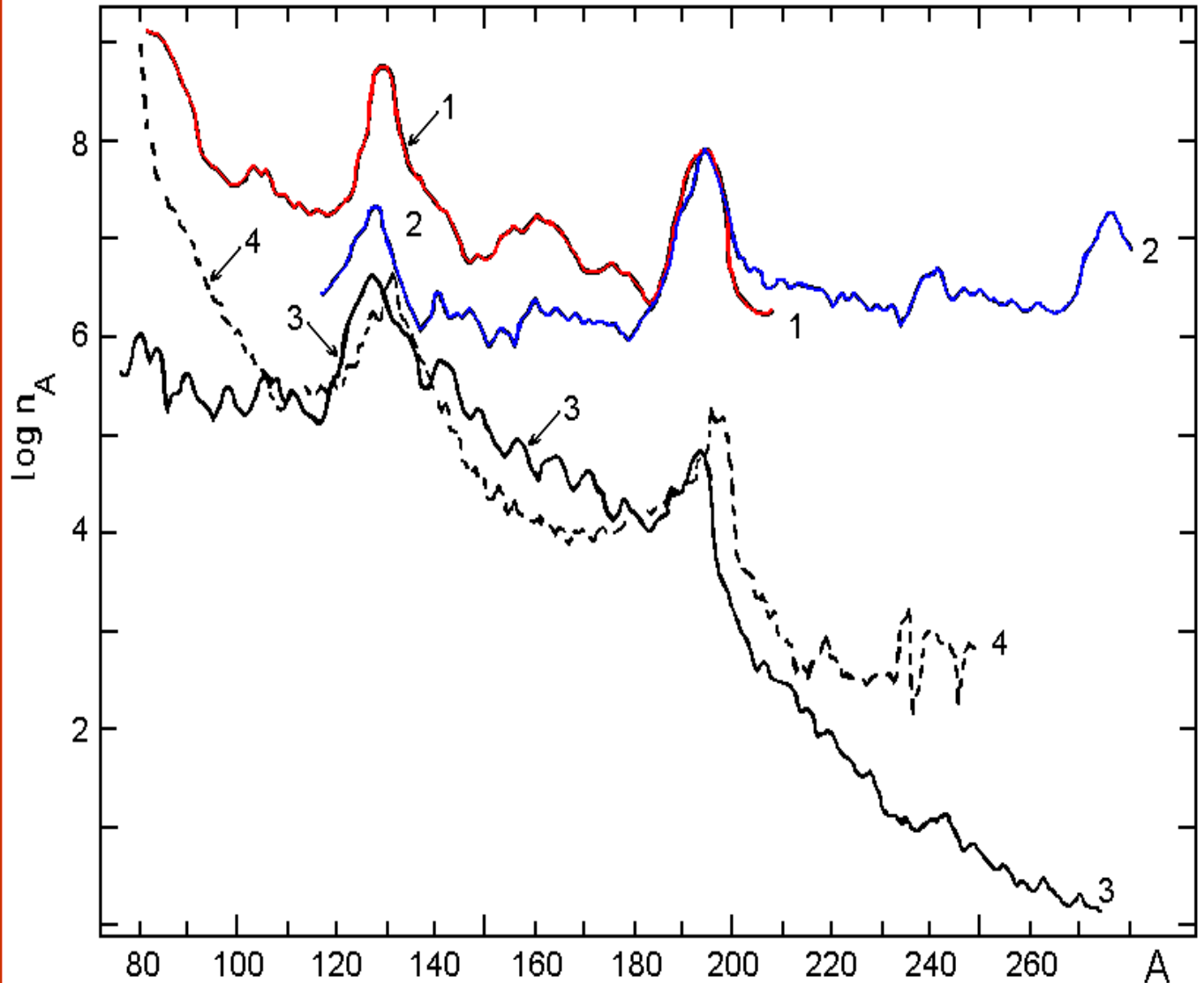
**1**- exp abundance  
data.

Curves 2 and 3  
- Calculations with  
different parameters.

Curve 2:  $n_n=10^{24} \text{ cm}^{-3}$ ,  
 $T_9=10^9 \text{ K}$ ,  $\tau_H=\infty$ ;

Curve 3:  
 $\rho_0 = 2 \cdot 10^5 \text{ g} \cdot \text{cm}^{-3}$ ,  
 $T_9=2 \times 10^9 \text{ K}$ ,  $\tau_H=1 \text{ s}$ .

Curve 4 (dotted line) –  
calculations with  
solar abundance  
initial nuclei  
concentration of  
s – elements.

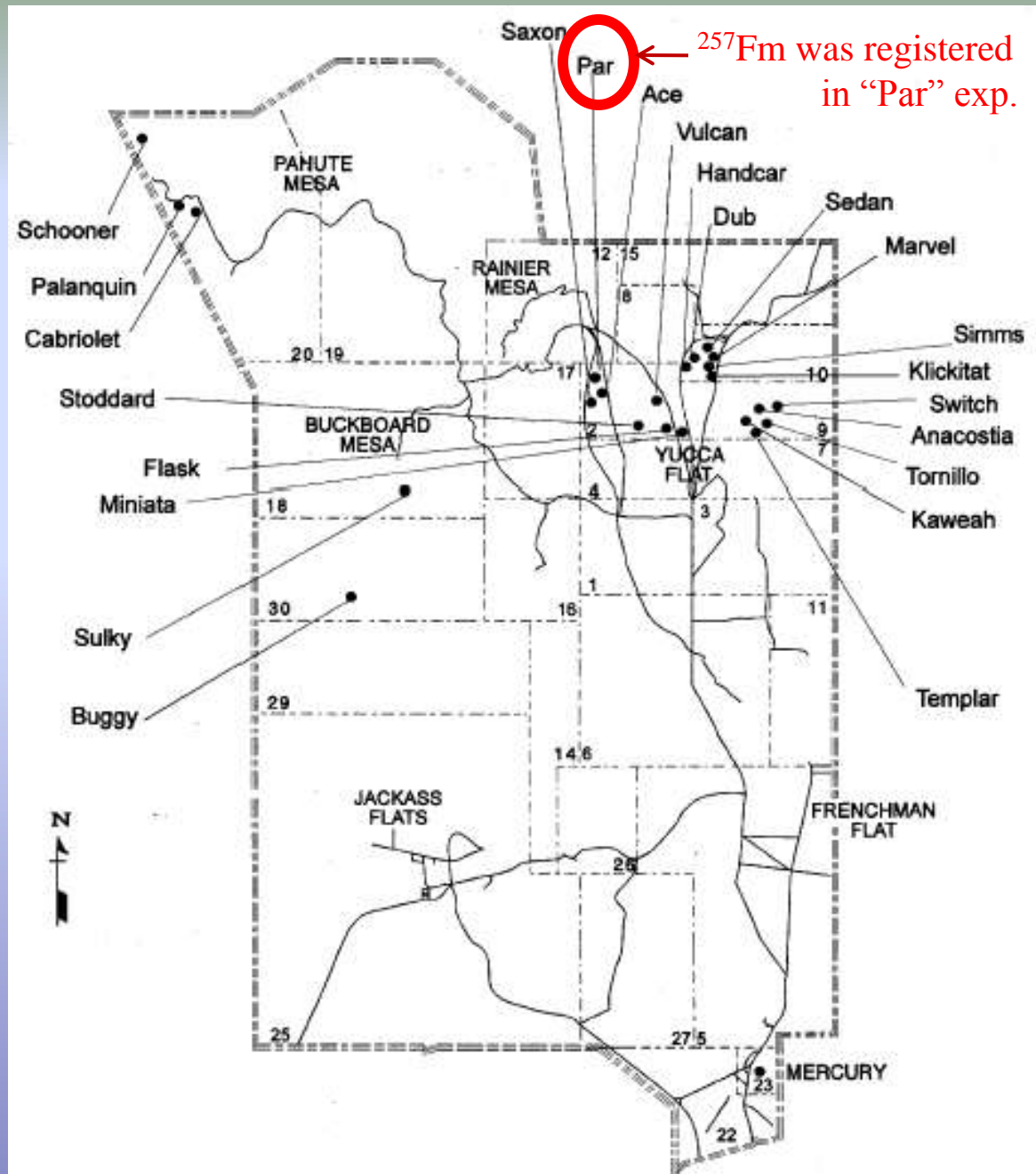


The production of transuranium isotopes was accomplished for the first time in the “Mike” thermonuclear experiment. Under the “**Plowshare**” project (United States), a series of experiments were performed (“Anacostia,” “Par,” “Barbel,” “Tweed,” “Cyclamen,” “Kankakee,” “Vulcan,” “Hutch,” and so on) to study heavy isotope production.

Программа “Плаушер” (США)  
исследование образования  
трансурановых  
элементов в ядерных/термоядерных  
взрывах

Nevada U.S.A.

In the “Cyclamen” and “Hutch” experiments integral fluxes were  $1.2 \cdot 10^{25}$  neutron  $\text{cm}^{-2}$  and  $4.5 \cdot 10^{25}$  neutron  $\text{cm}^{-2}$ , respectively.

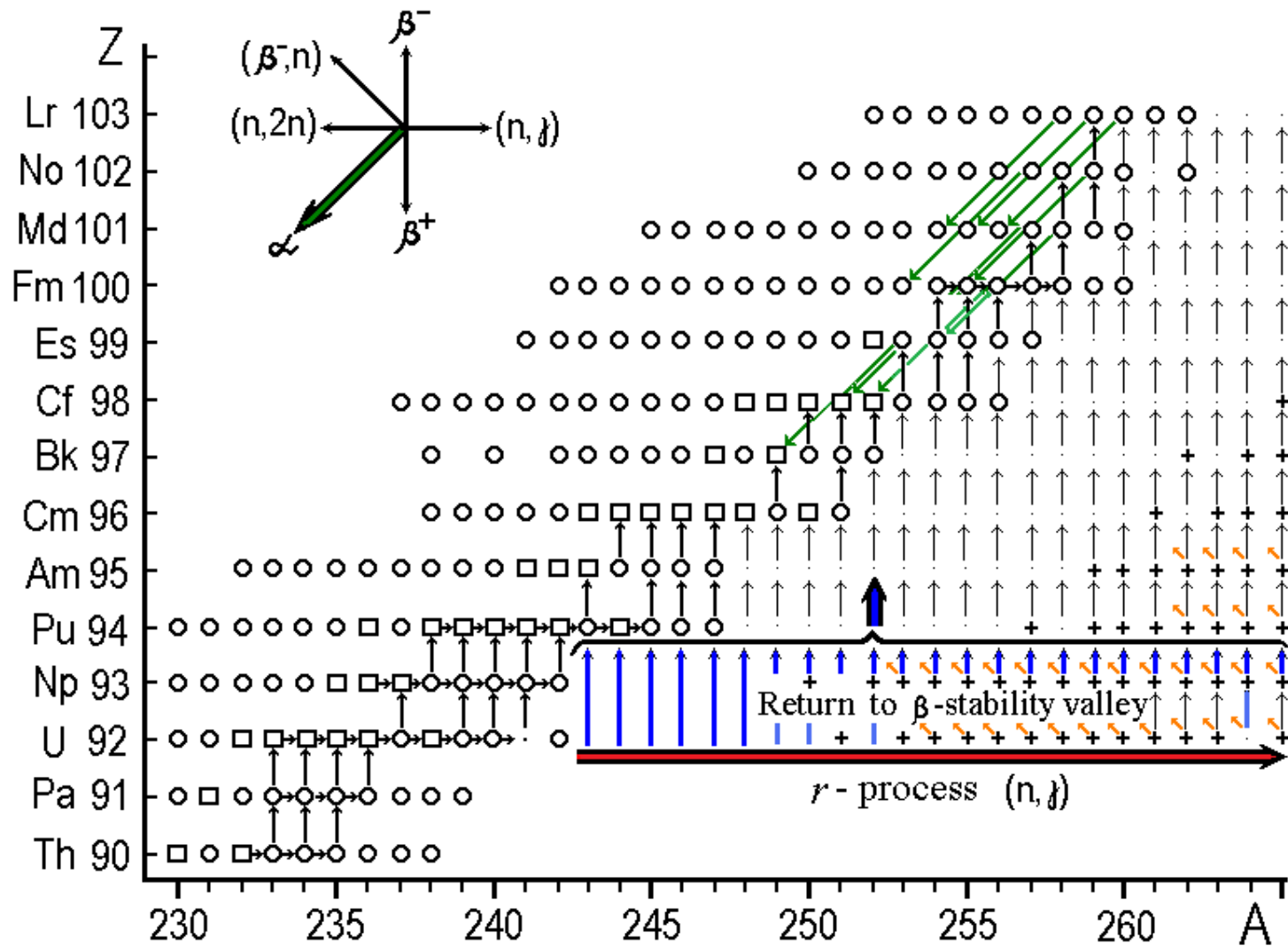


## Significant Nuclear Tests in the U.S.A. Heavy Element Program

Event	Date	Power	Fluence (n/cm <sup>2</sup> )	Target
<u>Mike</u>	31.10.1952	<u>10.4 Mt</u>	<u>(1.2-1.8) · 10<sup>24</sup></u>	<sup>238</sup> U
Anacostia	27.11.1962	< 20 kt	3 · 10 <sup>24</sup>	<sup>238</sup> U
<u>Par</u>	09.10.1964	<u>38 kt</u>	<u>~ 6.6 · 10<sup>24</sup></u>	<sup>238</sup> U
Barbel	16.10.1964	< 20 kt	~ 6.6 · 10 <sup>24</sup>	<sup>238</sup> U
Tweed	21.05.1965	< 20 kt	~ 7.2 · 10 <sup>24</sup>	<sup>237</sup> Np, <sup>242</sup> Pu
Cyclamen	05.05.1966	12 kt	~ 10.8 · 10 <sup>24</sup>	<sup>238</sup> U, <sup>243</sup> Am
Kankakee	15.06.1966	(20 – 200) kt	7.2 · 10 <sup>25</sup>	<sup>238</sup> U
Vulcan	25.06.1966	25 kt	~ 7 · 10 <sup>25</sup>	<sup>238</sup> U
Hutch	16.07.1969	(20 – 200) kt	(2.1–2.7) · 10 <sup>25</sup>	<sup>238</sup> U, <sup>232</sup> Th

There is no obvious relationship between the power of the explosion and the neutron flux

# r(rapid) – process in the explosive nucleosynthesis



Multiple neutron capturing process on Uranium material (U – target),  $t < 10^{-6}$  s.



## II - METHOD: r-Process equations for the concentration calculations

Concentrations  $n(A,Z)$  are changing in time (may be more than 4000 equations):

$$\begin{aligned}
 \frac{dn(A, Z)/dt = & -\lambda_{\beta}(A, Z) \cdot n(A, Z) - \lambda_{n\gamma}(A, Z) \cdot n(A, Z) + \lambda_{\gamma n}(A+1, Z) \cdot n(A+1, Z) + \\
 & + \lambda_{n\gamma}(A-1, Z) \cdot n(A-1, Z) - \lambda_{\gamma n}(A, Z) \cdot n(A, Z) + \\
 & + \lambda_{\beta}(A, Z-1) \cdot n(A, Z-1) \cdot P_{\beta}(A, Z-1) + + \lambda_{\beta}(A+1, Z-1) \cdot n(A+1, Z-1) \cdot P_{1n}(A+1, Z-1) + \\
 & + \lambda_{\beta}(A+2, Z-1) \cdot n(A+2, Z-1) \cdot P_{2n}(A+2, Z-1) + \lambda_{\beta}(A+3, Z-1) \cdot n(A+3, Z-1) \cdot P_{3n}(A+3, Z-1) + \\
 & + \cancel{\Phi_{\nu}(A, Z)} + F_f(A, Z),
 \end{aligned}$$

$\lambda_{n\gamma}$  and  $\lambda_{\gamma n}$  — rates of  $(n, \gamma)$  and  $(\gamma, n)$  -reactions,  $\lambda_{\beta} = \ln(2/T_{1/2})$  —  $\beta$ -decay rate,  $P_{\beta}$  - probability of  $(A, Z)$  nuclide creation after  $\beta$ -decay of  $(A, Z-1)$  nuclide. Branching coefficients of isobaric chains -  $P_{1n}$ ,  $P_{2n}$ ,  $P_{3n}$  corresponds to probabilities of one-, two- and three- neutrons emission in  $\beta$ - decay of the neutron-rich nuclei; the total probability of the delayed neutrons emission is the sum:

$$P_n = \sum_k P_{kn}$$

$F_f(A, Z)$  describes fission processes.

Neutrino capturing processes are not included ( $\Phi_{\nu}(A, Z) = 0$ )

### **III - METHOD: neutron capturing and $\beta$ -decay process are separated in (ADIABATIC MODEL) time and calculated separately.**

All time scale of the explosion is divided on small nanoseconds time intervals where all parameters considered to be constant and the expansion process is - adiabatic. For subsequent steps, the initial conditions for all isotopes are nonzero and are equal to the accumulation of the given nuclides by the moment of the preceding time step's completion. Concentrations  $N(A,Z)$  in every time interval are changing in time:

$$\left\{ \begin{array}{l} \frac{\partial N_z^n}{\partial t} = -(\lambda_{n,\gamma} N)_z^n \\ \frac{\partial N_z^{n+1}}{\partial t} = (\lambda_{n,\gamma} N)_z^n - (\lambda_{n,\gamma} N)_z^{n+1} \\ \cdot \\ \cdot \\ \cdot \\ \frac{\partial N_z^{n+i}}{\partial t} = (\lambda_{n,\gamma} N)_z^{n-1+i} - (\lambda_{n,\gamma} N)_z^{n+i} \end{array} \right. \quad (3)$$

The neutron capture rates were calculated in the statistical model [1]. The neutron rich isotopes half-lives, probabilities of  $\beta$ -delayed one and two neutrons emission, probabilities of  $\beta$ -delayed fission were calculated taking into account the  $\beta$ -strength function, which was obtained from the theory of finite-Fermi systems [2].

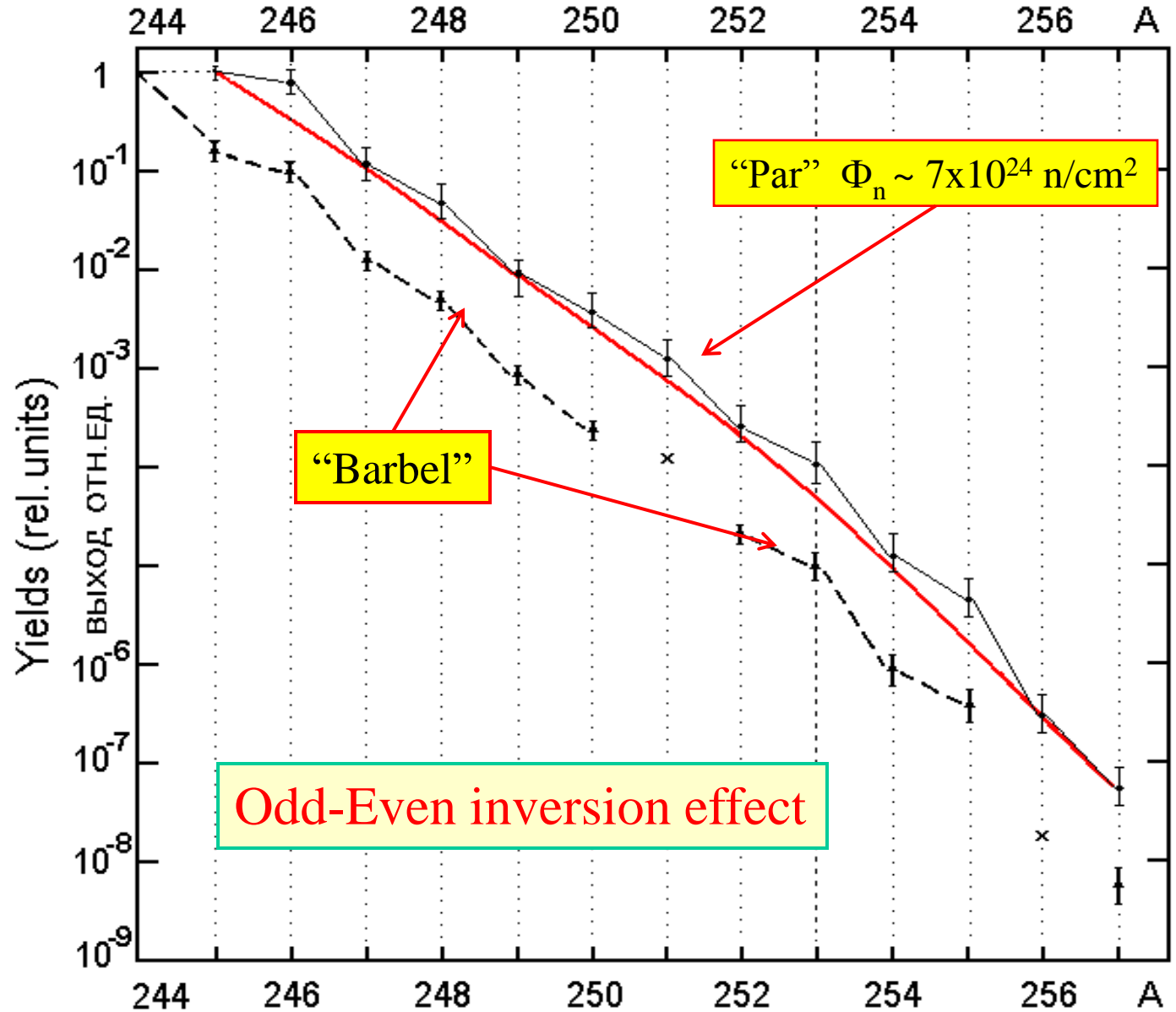
1. Panov I. V., Kolbe E., Pfeiffer B., Rauscher T., Kratz K.-L., Thielemann F.-K. Nucl. Phys. A. 2005. V.747, p.633.
2. Gaponov Yu. V., Lutostansky Yu. S. Yad. Fiz. 2010, V.73, №8, p. 665

# Yields (concentrations in relative units) for “Par” and “Barbel” experiments

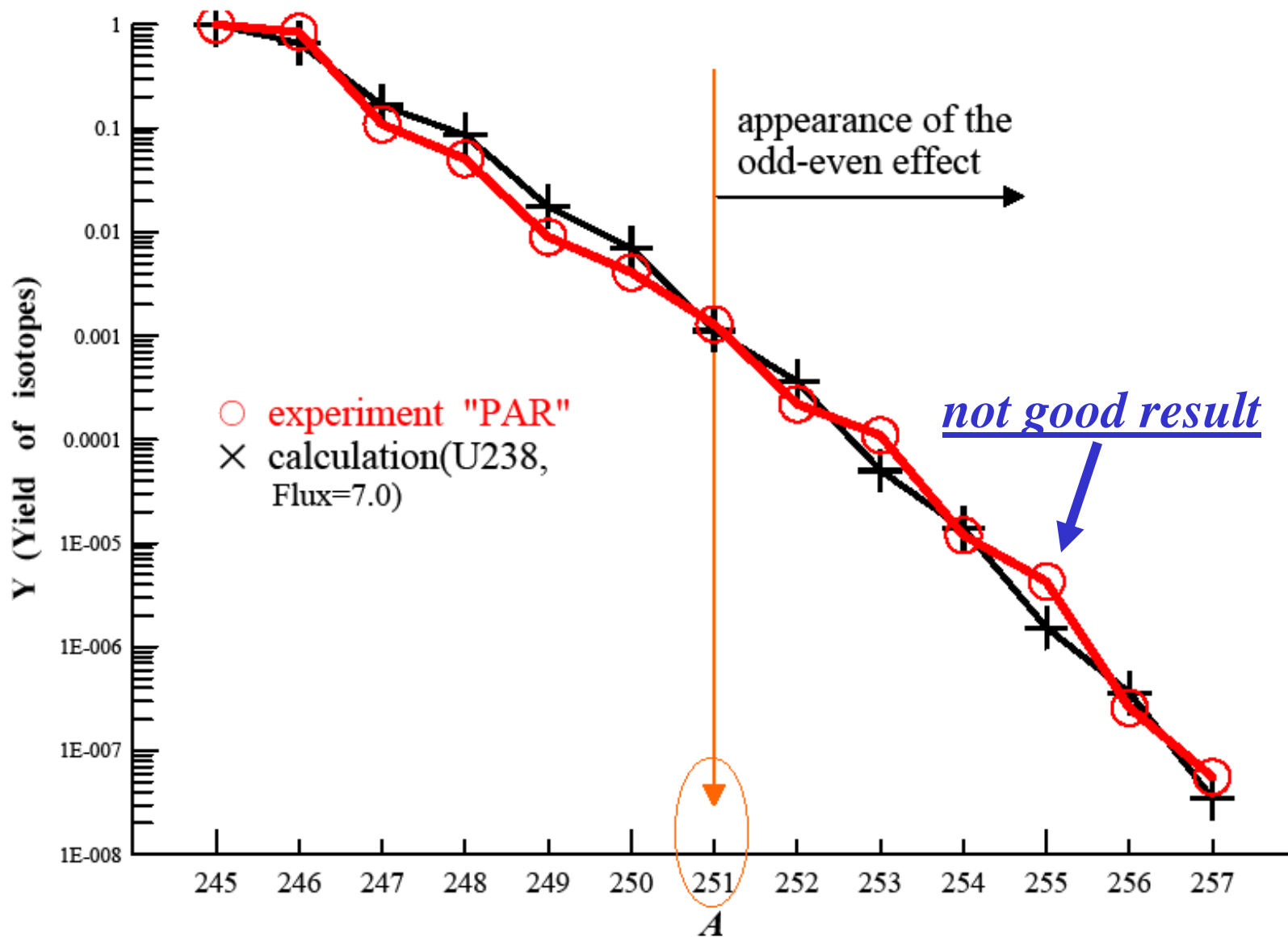
Yields of transuranium nuclei measured in the thermonuclear explosions “Par” and “Barbel” (LANL data).

— Red line – approximation

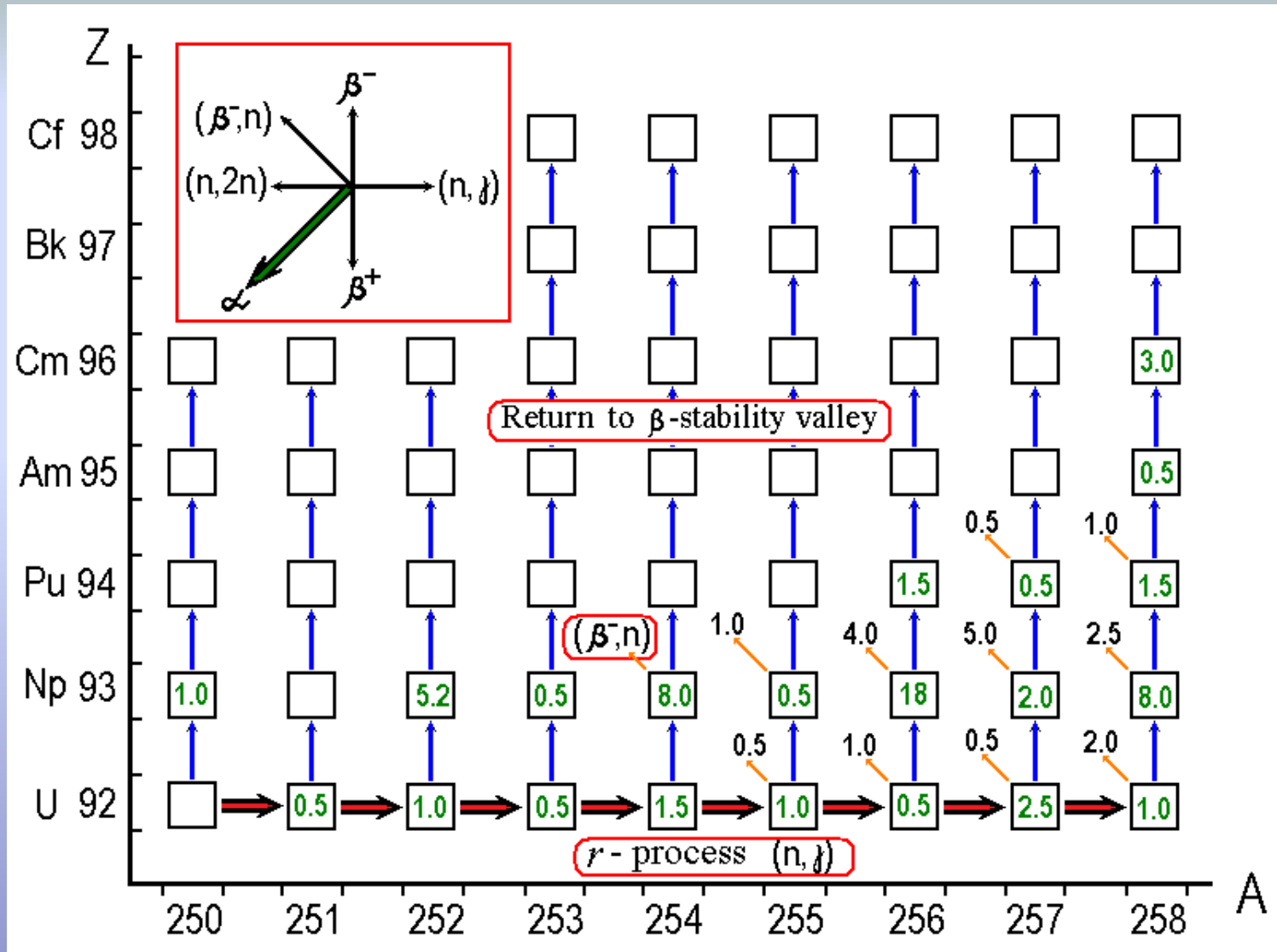
$$Y = \exp(-1.442A + 354.56)$$



# Yields (concentrations in relative units) for "Par" experiment with U - target (calculations not including "Losing effects")



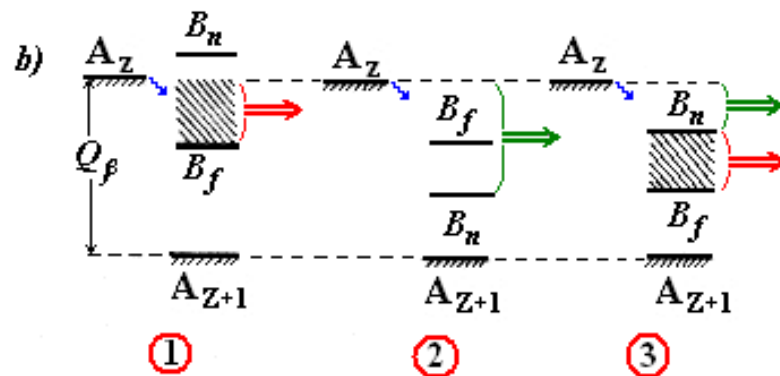
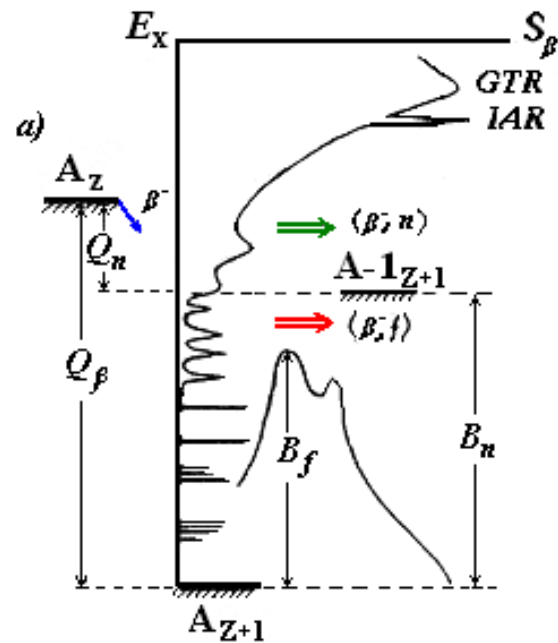
# r(rapid) – process in the explosive nucleosynthesis



The initial concentration changed because of  $\beta$ -delayed fission and neutron emission during cooling processes (return to the value of  $\beta$ -stability).



# BETA - DELAYED FISSION



# Beta – Delayed Fission Calculations

**Probabilities -  $P_{\beta f}$ :**

$$P_{\beta f} = \frac{\int_0^{Q_\beta} \sum_i f(Z, Q_\beta - E) S_\beta^i(E) \frac{\Gamma_f}{\Gamma_{\text{tot}}} dE}{\int_0^{Q_\beta} \sum_i f(Z, Q_\beta - E) S_\beta^i(E) dE}$$

**Beta Strength function:**

$$S_\beta(E) = \frac{C_N}{2\pi} \sum_i M_i^2(E_i) \frac{\Gamma(E)}{(E_i - E)^2 + \frac{\Gamma^2(E)}{4}}$$

**#  $\Gamma(E)$  widths approximation:  $\Gamma(E) = \alpha \cdot E^2 + \beta \cdot E^3 + \dots$**

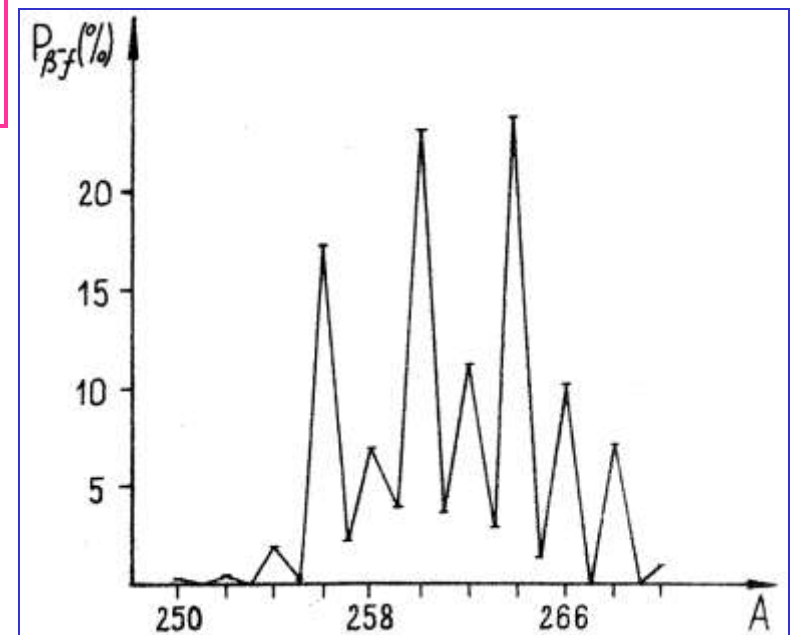
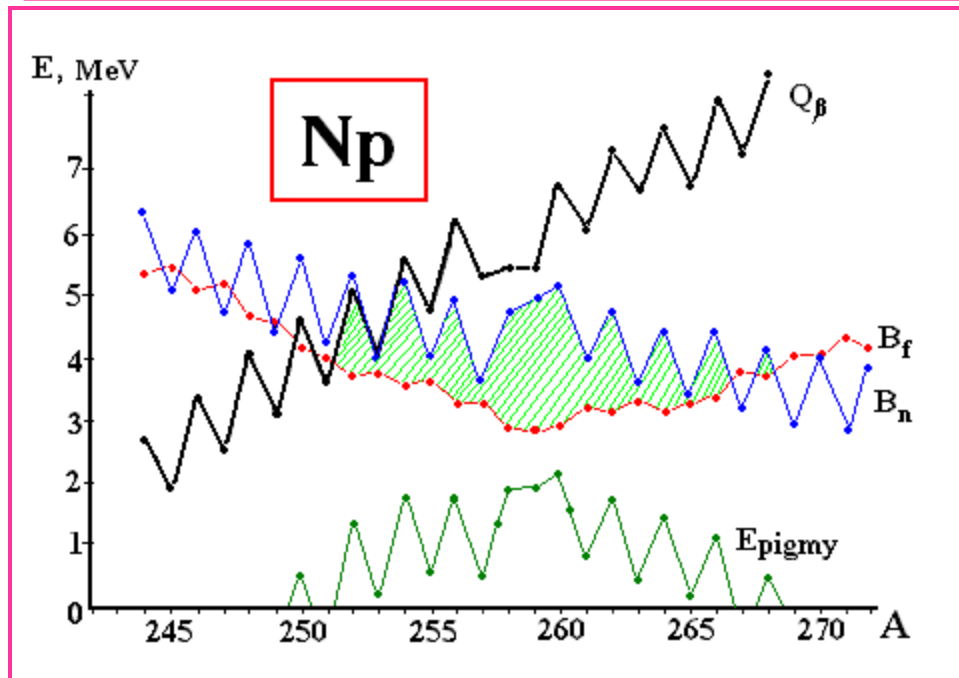
**where  $\alpha \approx 1/\varepsilon_F$  and  $\beta \ll \alpha$ , so we used only the first term.**

**# As  $\Gamma_f \ll \Gamma_n$  so neutron emission dominates when this energetically possible.**

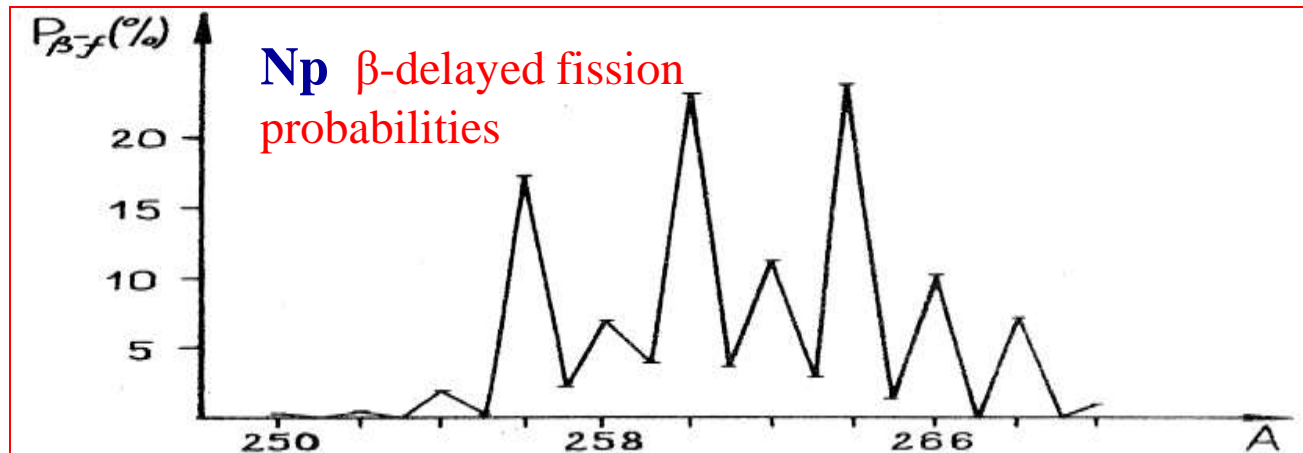
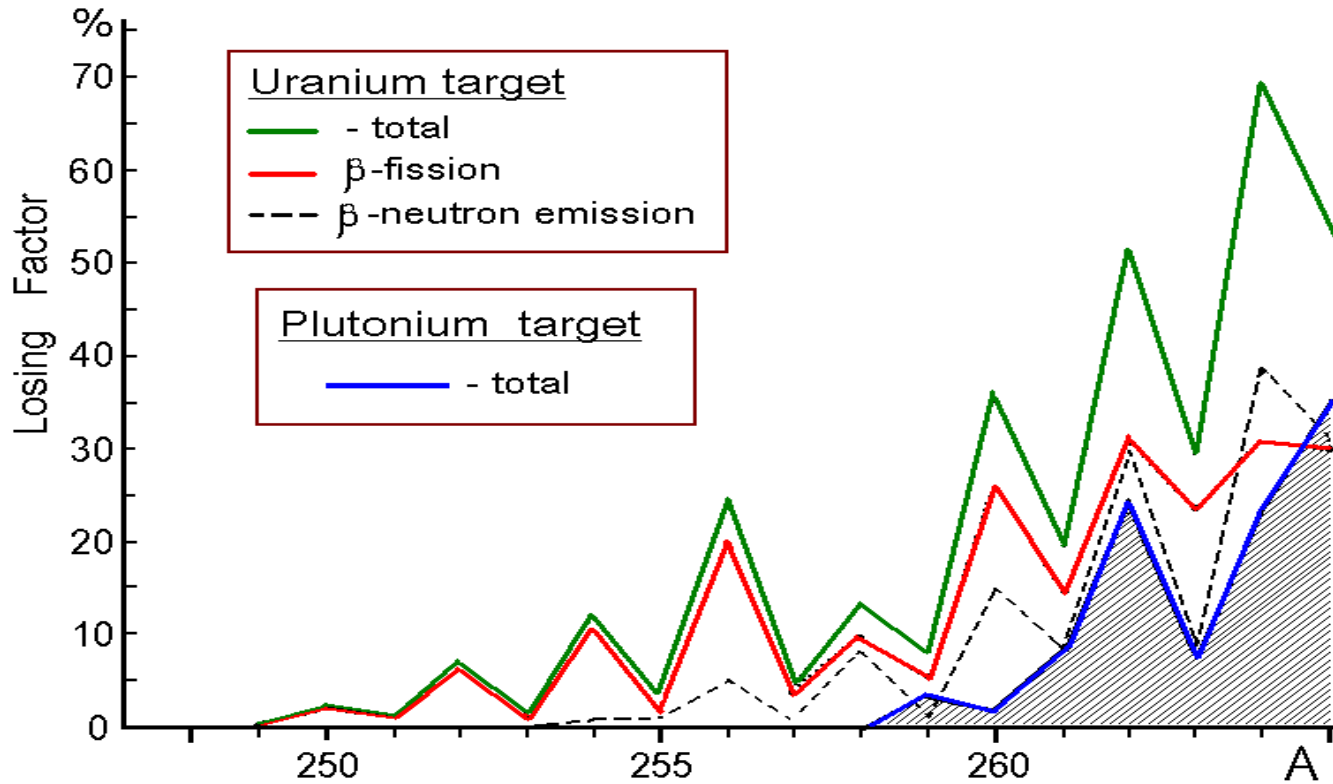
**# Sub-barrier fission probabilities in the daughter nucleus are small to gamma decay of excited states (barrier was taken in standard parabolic form).**

**# Main dependence of  $P_{\beta f}$  is from barrier energy  $B_f$  but not from barrier thickness or form.**

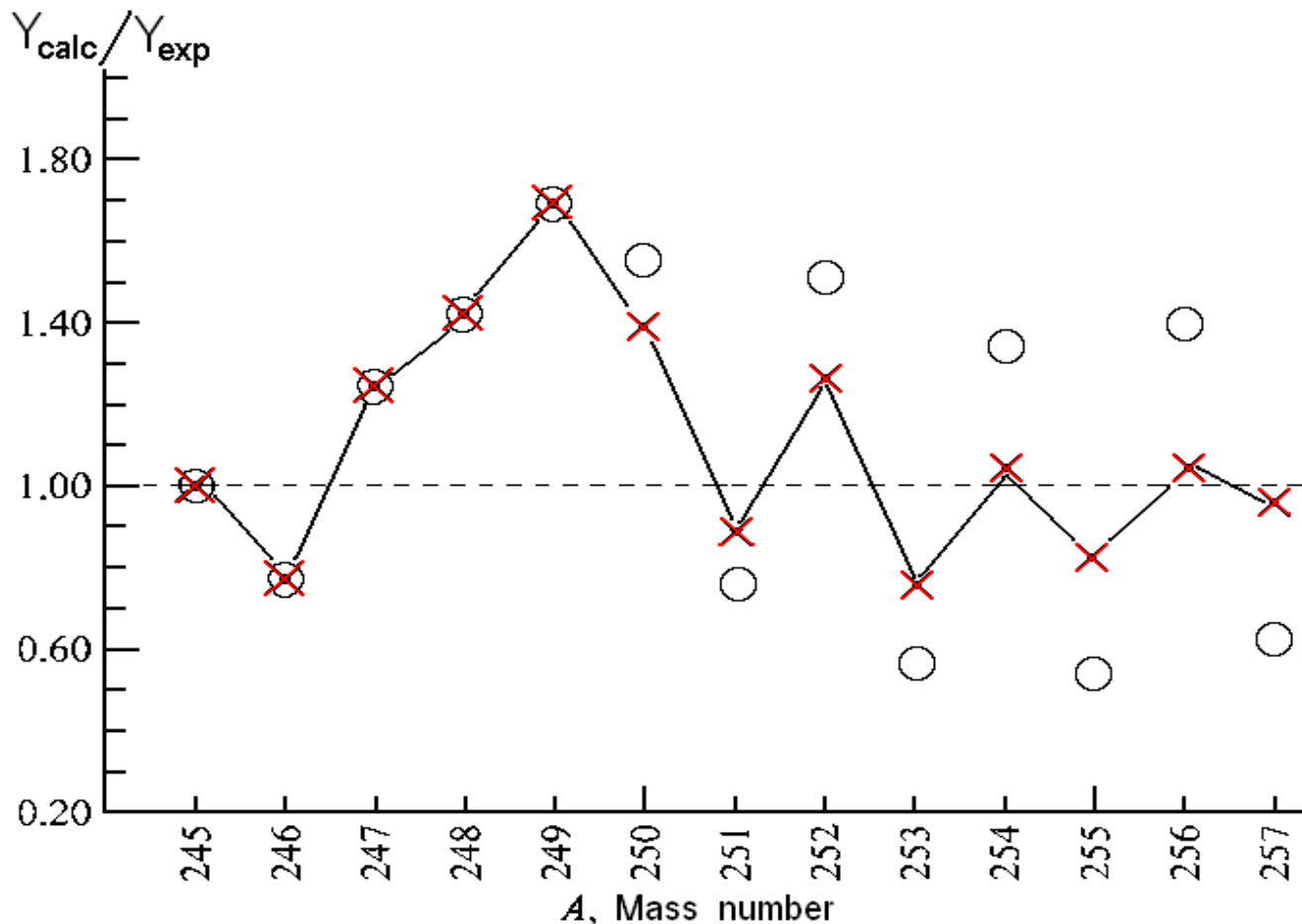
# Neptunium Beta – Delayed Fission Calculations



# Factor of the concentration losing



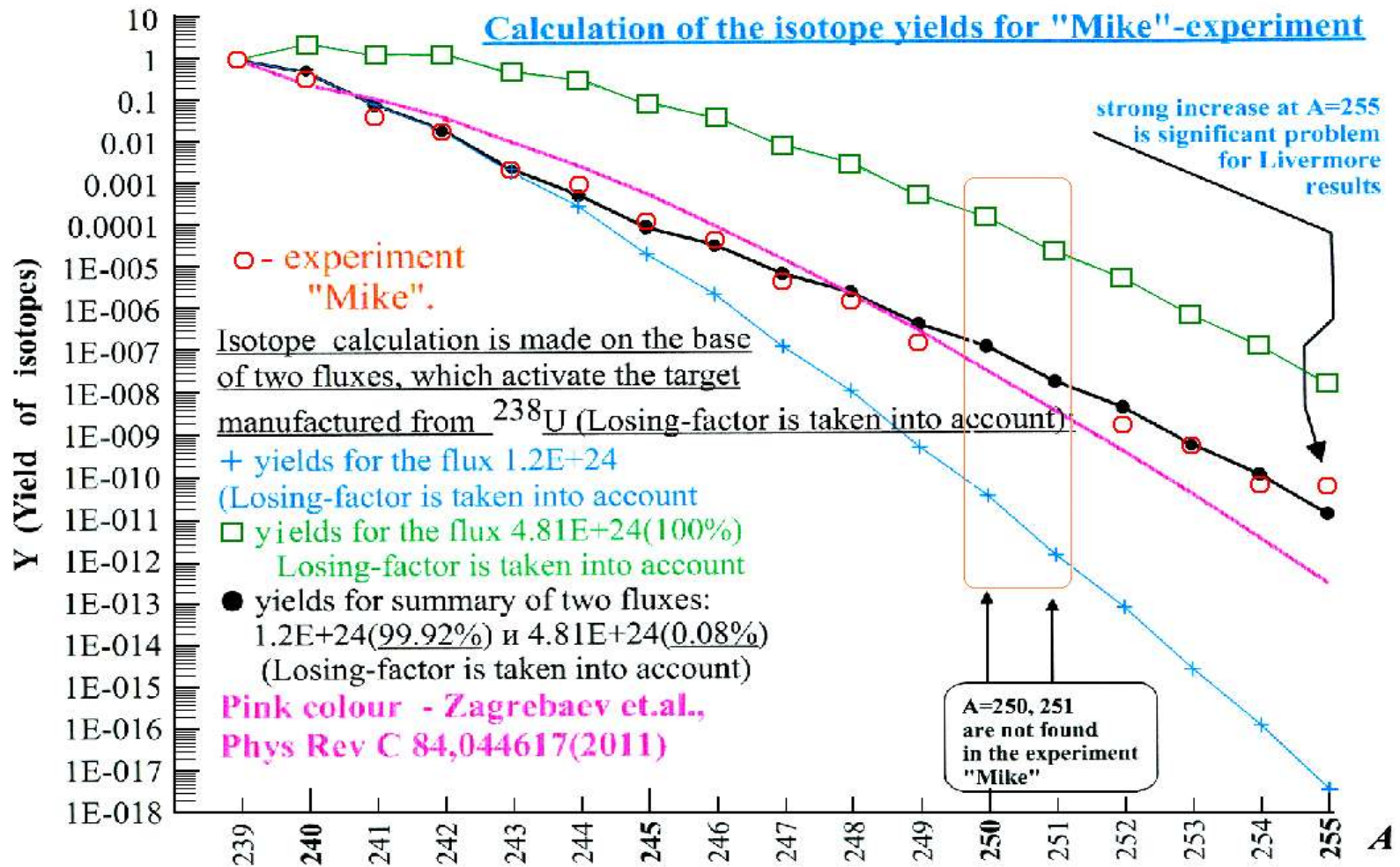
# Calculations for "Par" experiment (Calc. Yields rel. to exp. data)



Отношение расчетных значений к экспериментальным выходам  
O – расчетные результаты для адиабатической модели с  $^{238}\text{U}$  как единственным изотопом в мишени; × - расчетные результаты, с учетом запаздывающего деления (соединены линией).

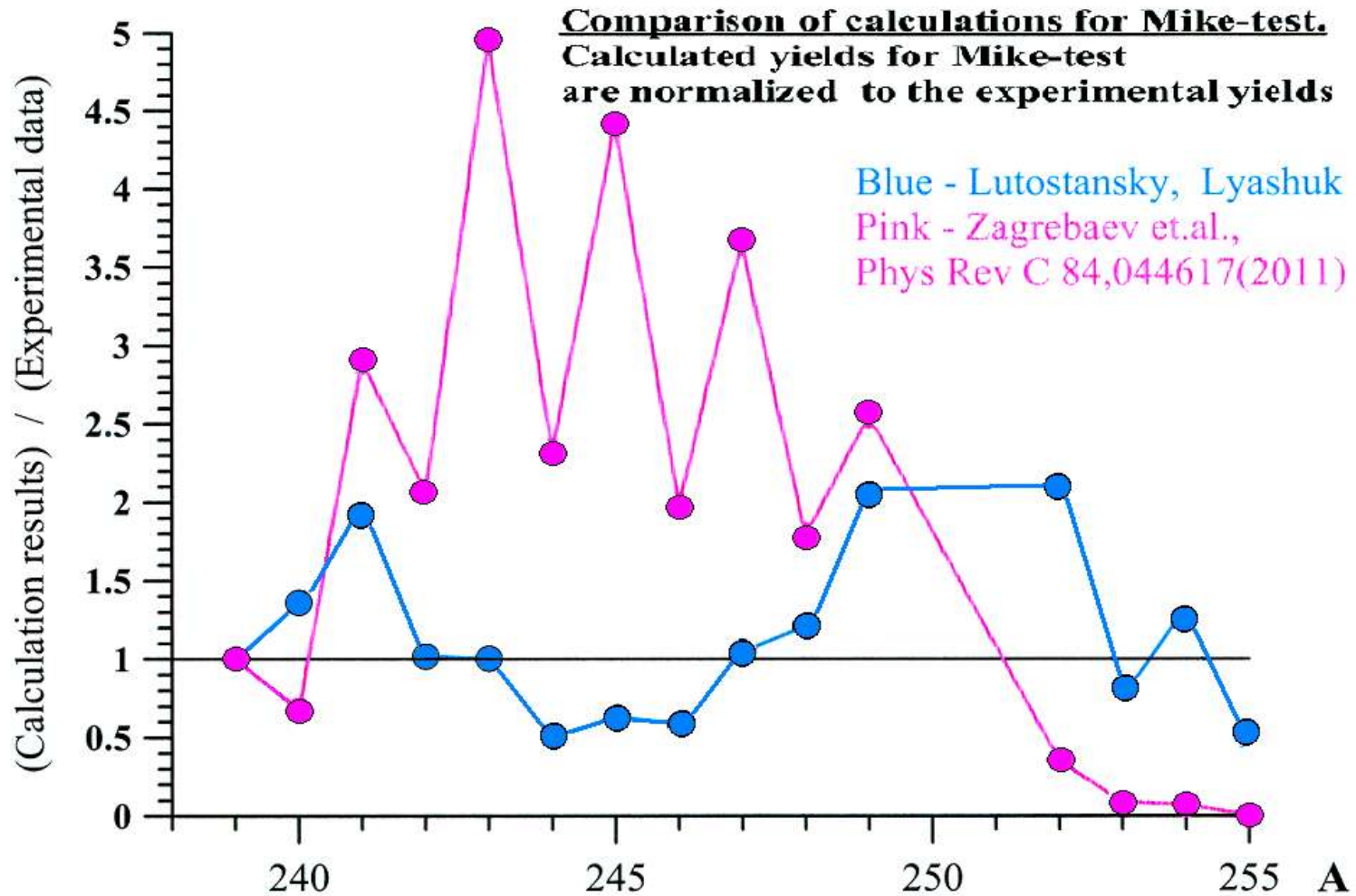


# “Mike” experiment - 1

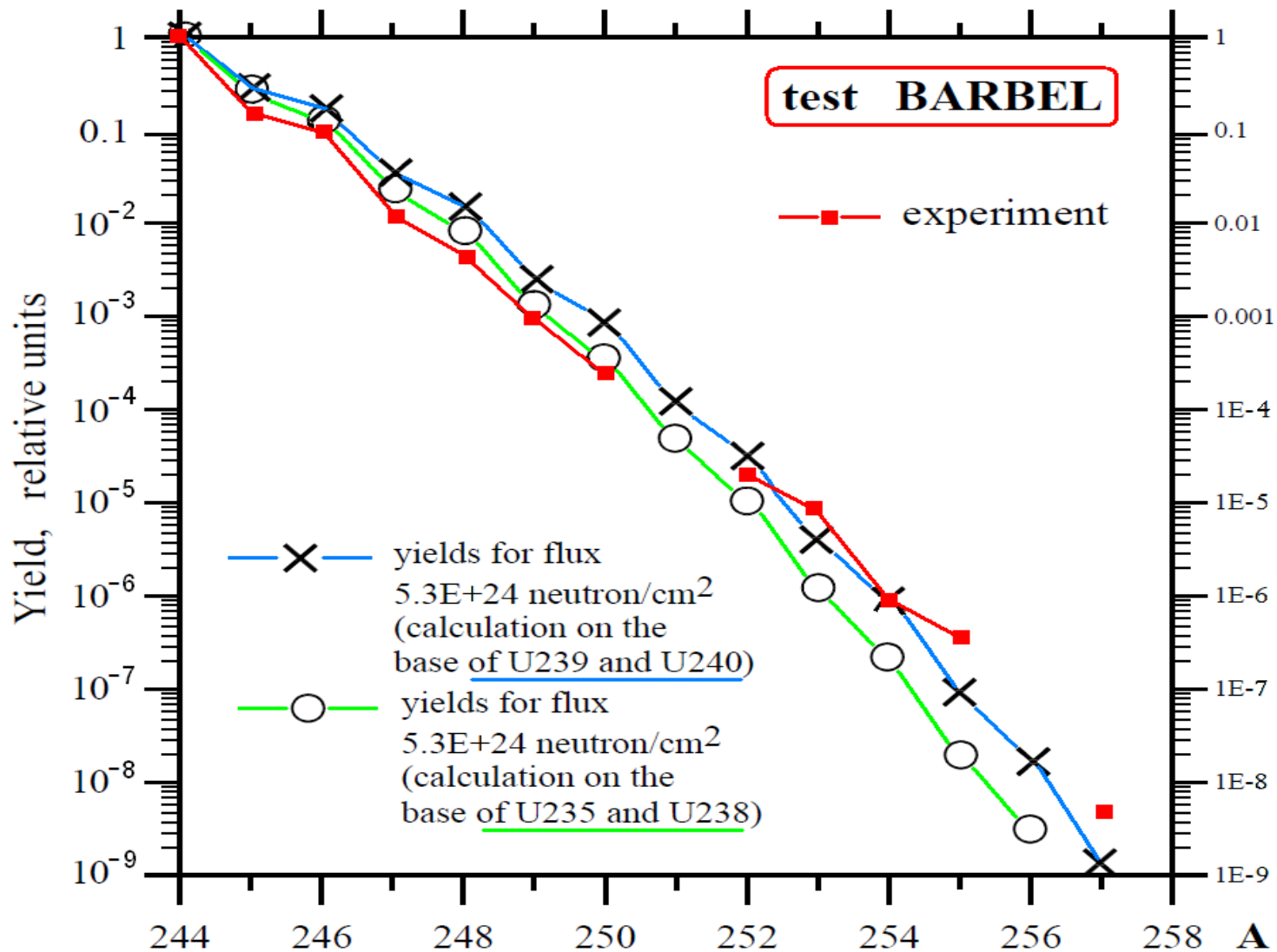


V. I. Zagrebaev, A. V. Karpov, I. N. Mishustin, and Walter Greiner  
 “Production of heavy and superheavy neutron-rich nuclei in neutron capture processes”  
 PHYSICAL REVIEW C 84, 044617 (2011)

## “Mike” experiment - 2

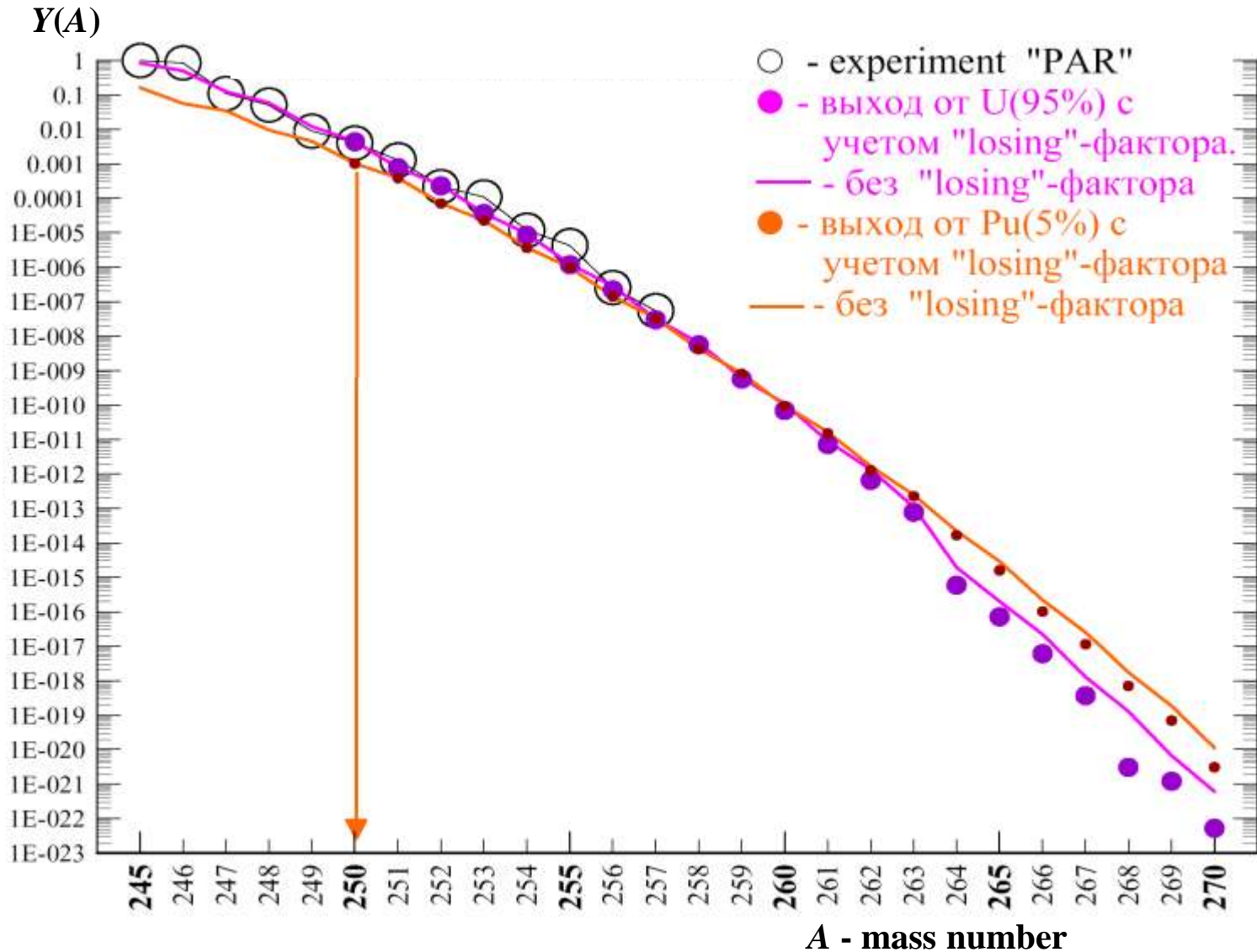


V. I. Zagrebaev, A. V. Karpov, I. N. Mishustin, and Walter Greiner  
“Production of heavy and superheavy neutron-rich nuclei in neutron capture processes”  
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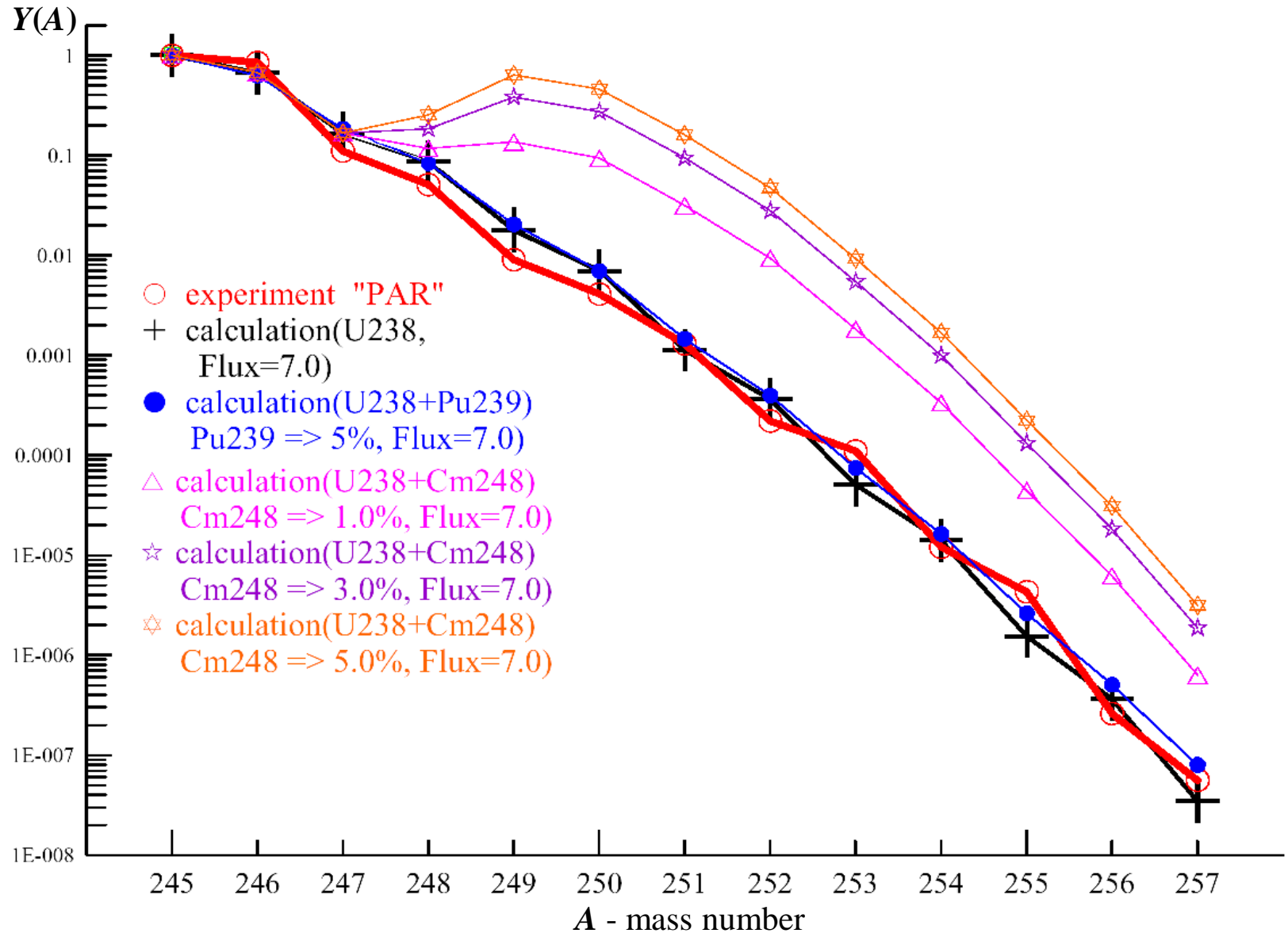


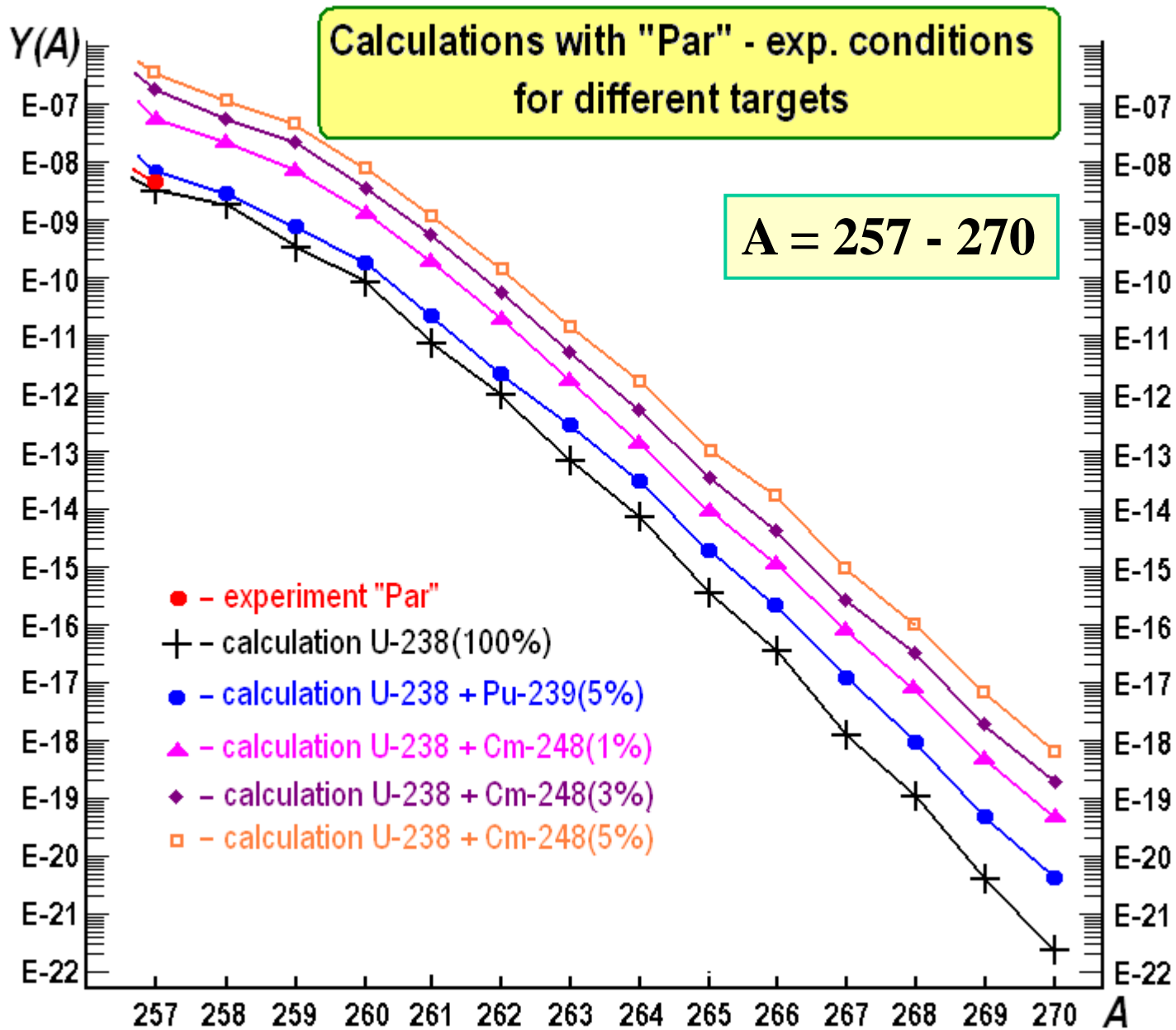
**Strong dependence from target isotopes**

# Calculations for "PAR"- experiment to $A = 270$

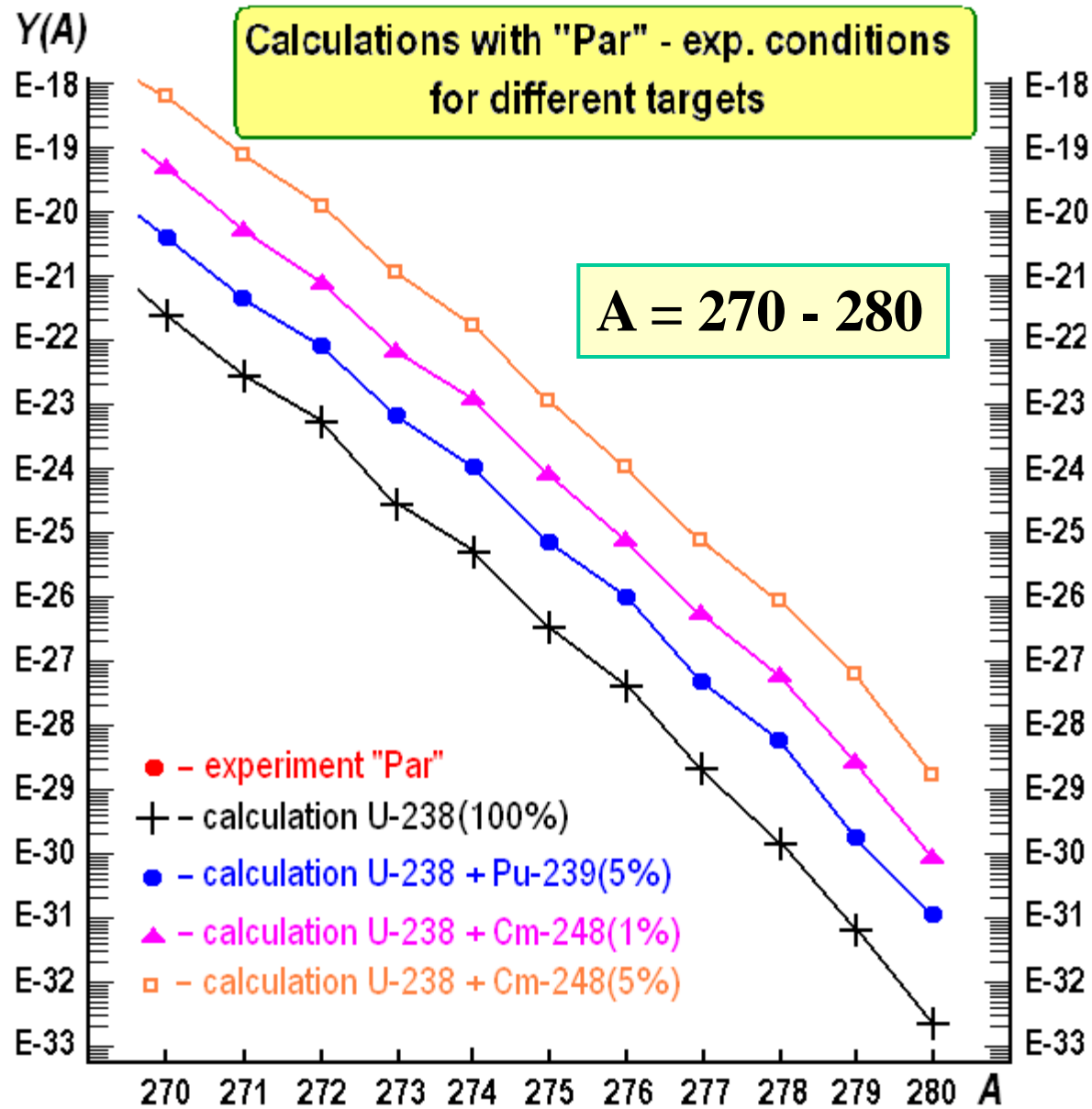


# Calculations for "PAR"- experiment with $^{238}\text{U} + ^{248}\text{Cm}$ target









# NUCLEOSYNTHESIS AFTER NEUTRON PULSE – 1

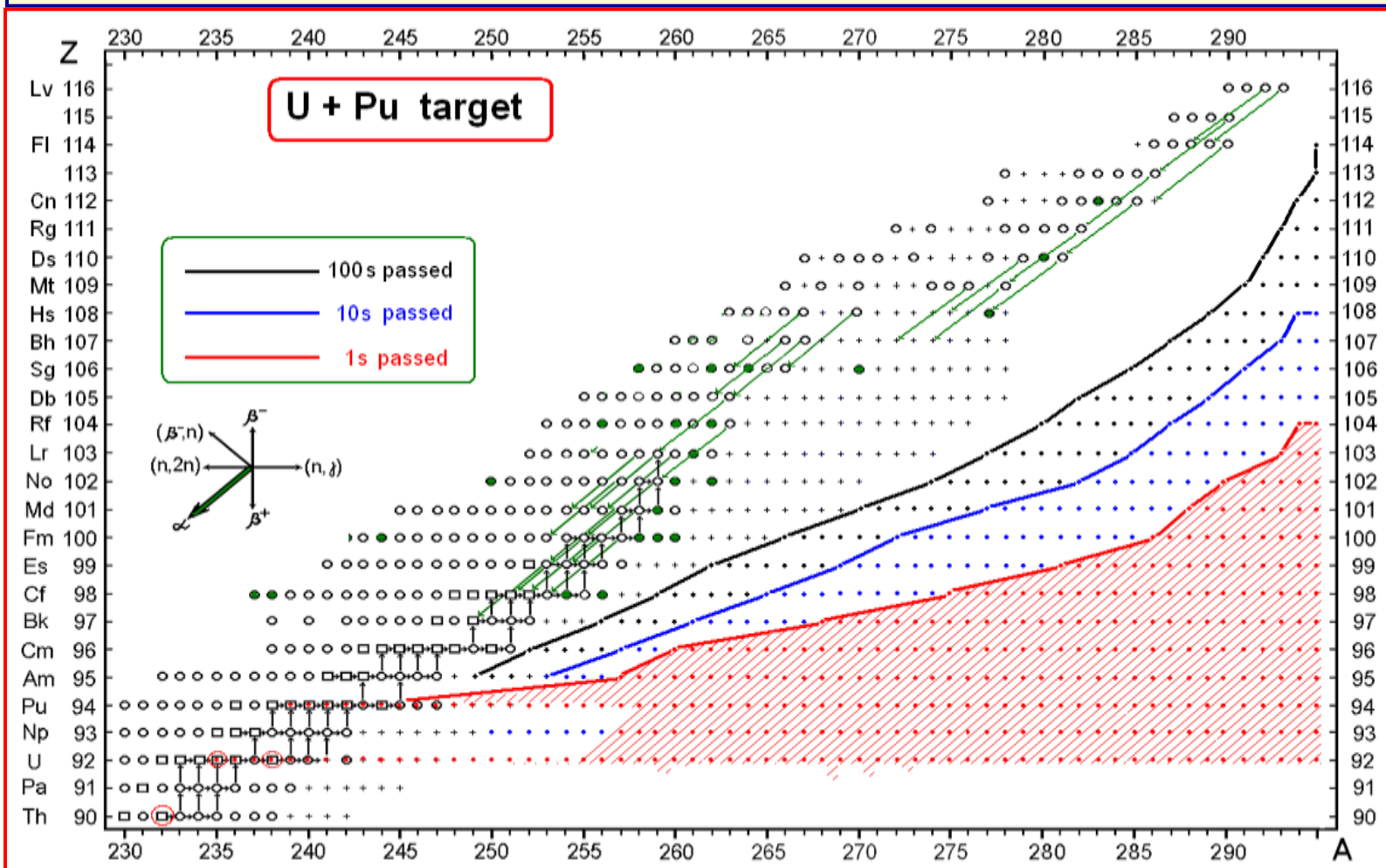
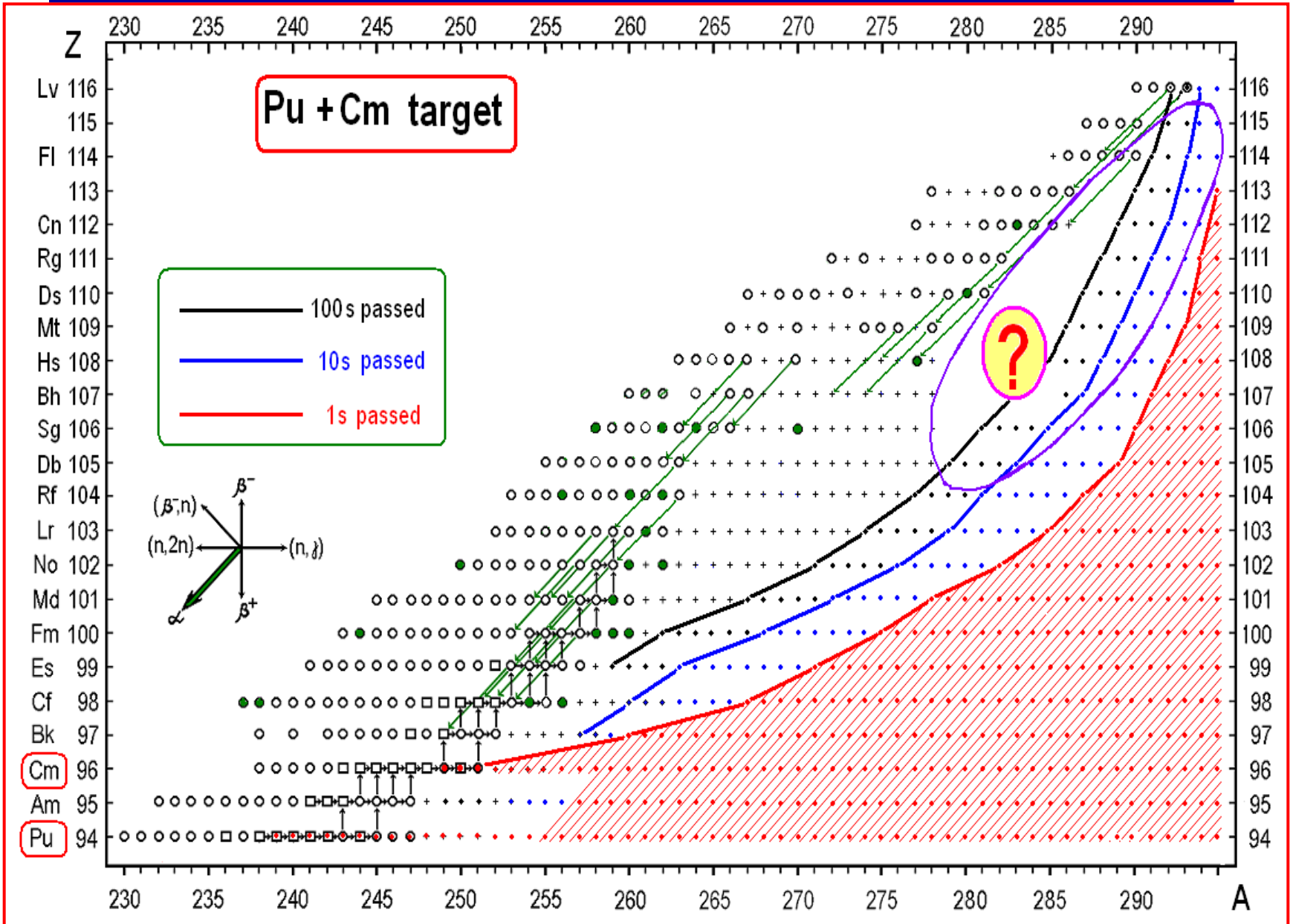


Схема образования актинидов в быстром (*r*-процесс) нуклеосинтезе (ядерный взрыв);  $\square$  – ядра с  $T_{1/2} \geq 1$  г ;  $\circ$  – ядра с  $T_{1/2} < 1$  г ; + - прогнозируемые нейтронно-избыточные ядра



# NUCLEOSYNTHESIS AFTER NEUTRON PULSE – 2



# Conclusion

- The binary model of transuranium elements production in pulse neutron fluxes is developed. The calculations of the transuranium isotopes yields up to  $A = 295$  in pulsed neutron fluxes of high intensity in the adiabatic binary model were performed with start isotopes:  $^{238}\text{U}$ ,  $^{239}\text{Pu}$ ,  $^{248}\text{Cm}$  and  $^{251}\text{Cf}$ .
- Comparison of yields calculations up to  $A = 257$  for binary targets with experiment data were carried out to “*Par*”, “*Barbel*” and “*Mike*” thermonuclear explosions.
- Experimental data on the yields of transuranium isotopes in nuclear explosions revealed anomalous odd-even inversion effect, that is not small in the mass number  $A > 250$ , and may be explained by the delayed fission process, calculations with which leads to better agreement with experiment. The agreement of the calculated isotopes yields with the experimental data is up to 50%.
- The “eating away” effect – the losing effect, which gives the relative decreasing of the concentration of nuclei with fix  $A$  and which is due to the emission of delayed neutrons and delayed fission - the processes leading to changes in the concentrations of  $\beta$ -decay of short-lived intermediates, formed nuclei with large neutron excess. It is shown that the  $L(A)$ -effect associated with the observed even-odd inversion in yields of the transuranium nuclides.
- It is shown that nuclei with  $A \approx 270$  can be obtained in the “*Par*” experiments with the yields  $\sim 10^{-22}$  using a uranium target, and – with the yields  $\sim 10^{-18} \div 10^{-20}$  using binary U+Pu and U+Cm targets.
- Heavier nuclei with  $A \approx 280$  can be obtained with a yields  $\sim 10^{-29} \div 10^{-31}$  using binary U + Pu and U + Cm target. Such low concentrations can not be detected by modern experimental methods. Moreover, these nuclides decay rapidly.
- So the calculations were carried out up to the values of  $A = 295$ . It was obtained that the isotopic relations of some transuranium elements, for example - curium, depends on the value of the pulse neutron flux. This may be an indicator of the pulse component in cases of extreme accidents at the nuclear power stations.

## Заключение

- Проведены расчеты выходов трансурановых изотопов до  $A = 295$  в импульсных нейтронных потоках высокой интенсивности в адиабатической бинарной модели со стартовыми изотопами:  $^{238}\text{U}$ ,  $^{239}\text{Pu}$ ,  $^{248}\text{Cm}$  и  $^{251}\text{Cf}$ .
- Сравнение расчетов выходов до  $A = 257$  для U - и (U + Pu)-мишеней проведено по данным экспериментов “Par”, “Barbel” и “Mike”.
- Экспериментальные данные по выходам трансуранов в ядерных взрывах выявили аномальный нечетно-четный эффект, проявляющийся при массовом числе  $A > 250$ , который объясняется введением в модель запаздывающего деления, что ведет к улучшению согласия с экспериментом. Получено согласие в выходах изотопов в пределах до 50%.
- Изучен эффект “выедания” (*losing*-эффект), который дает относительное снижение концентраций для ядер данного  $A$  и который объясняется эмиссией запаздывающих нейтронов и запаздывающим делением – процессами ведущими к изменению концентраций при  $\beta$ -распаде образовавшихся промежуточных короткоживущих ядер с большим избытком нейтронов. Показано, что этот  $L$ -эффект связан с наблюдаемой четно-нечетной инверсией в выходах трансурановых нуклидов.
- Показано, что ядра с  $A \approx 270$  могут быть получены в экспериментах типа “Par” с выходами  $\sim 10^{-22}$  при использовании урановой мишени и – с выходами  $\sim 10^{-18}$   $10^{-20}$  при использовании бинарной U + Pu или U + Cm мишени.
- Более тяжелые ядра с  $A \approx 280$  могут быть получены с выходами  $\sim 10^{-29}$   $10^{-31}$  при использовании бинарной U + Pu или U + Cm мишени. Такие малые концентрации не могут быть определены современными методами. Более того, такие нуклиды быстро распадаются.



**THE END**

**СПАСИБО = THANK YOU**

# Экспериментальное исследование образования трансураниевых элементов в интенсивных нейтронных потоках

Впервые трансураниевые элементы были обнаружены в продуктах термоядерного взрыва “**Mike**” в 1952 г с мишенью  $^{238}\text{U}$ .

Ядерные и термоядерные взрывы обеспечивают большой поток нейтронов ( $10^{24} - 10^{25}$  нейтронов/см<sup>2</sup>) при кратковременной экспозиции ( $\sim <10^{-6}$  с – для реакций захвата) и тем самым являются уникальным инструментом для исследований в ядерной физике.

Для сравнения:

максимальный поток, достигнутый на реакторе **HFIR** –  $5.5 \cdot 10^{15}$  нейтронов/с;

в ловушке **ПИК** (проектное значение) –  $4 \cdot 10^{15}$  нейтр./(см<sup>2</sup>с),

за 1 год работы -  $1.2 \cdot 10^{23}$  нейтр./см<sup>2</sup> ;

в ловушке **СМ-2** –  $5 \cdot 10^{15}$  нейтр./(см<sup>2</sup>с);

**ИГР** (импульсный графитовый) - максимальный интегральный поток –  $1 \cdot 10^{18}$  нейтр./см<sup>2</sup>

**БИГР** (импульсный) -  $1.2 \cdot 10^{16}$  нейтр./см<sup>2</sup> в центральном канале;

**Гидра** (импульсный растворный) –  $8 \cdot 10^{14}$  нейтр./см<sup>2</sup> ;

**ЯГУАР** (импульсный растворный) –  $2.5 \cdot 10^{18}$  нейтр./(см<sup>2</sup>с) в импульсе в выводящем канале.