Secondary neutrons as the main source of the neutron rich fission residues production after the bombardment of a thick U target by 1 GeV protons: experimental evidences for Cs isotopes

O. Alyakrinskiy, M. Barbui, A.E. Barzakh, D.V. Fedorov,
V.S. Ivanov, G. Lhersonneau, K.A. Mezilev, P.L. Molkanov,
F.V. Moroz, S.Yu. Orlov, V.N. Panteleev,
L.B. Tecchio, Yu.M. Volkov

Petersburg Nuclear Physics Institute,
INFN Laboratori Nazionali di Legnaro, GANIL

1. Experimental data:
    a. Yield measurements.
        i. α- and γ- lines intensity measurement

Target: ^{238}U
(different thicknesses: from 3 to 150 g/cm^2;
different structures of target material;
different internal containers)

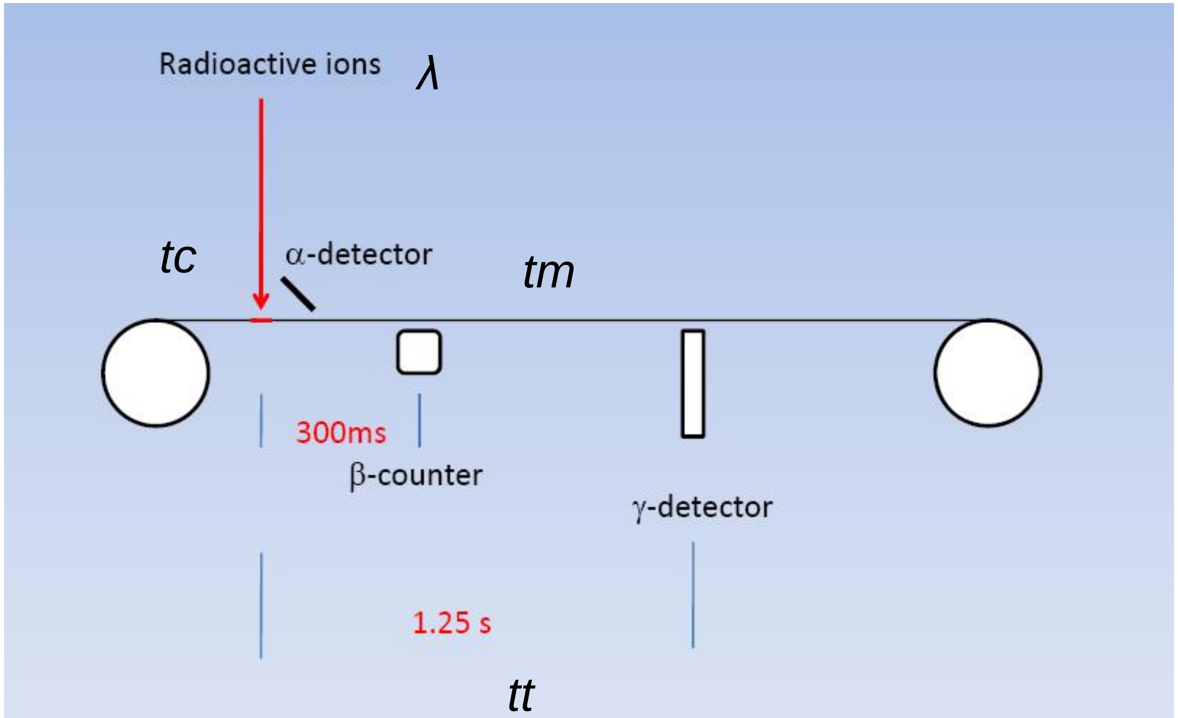
Reaction: p (1 GeV), ^{238}U

s.

) for release analysis.

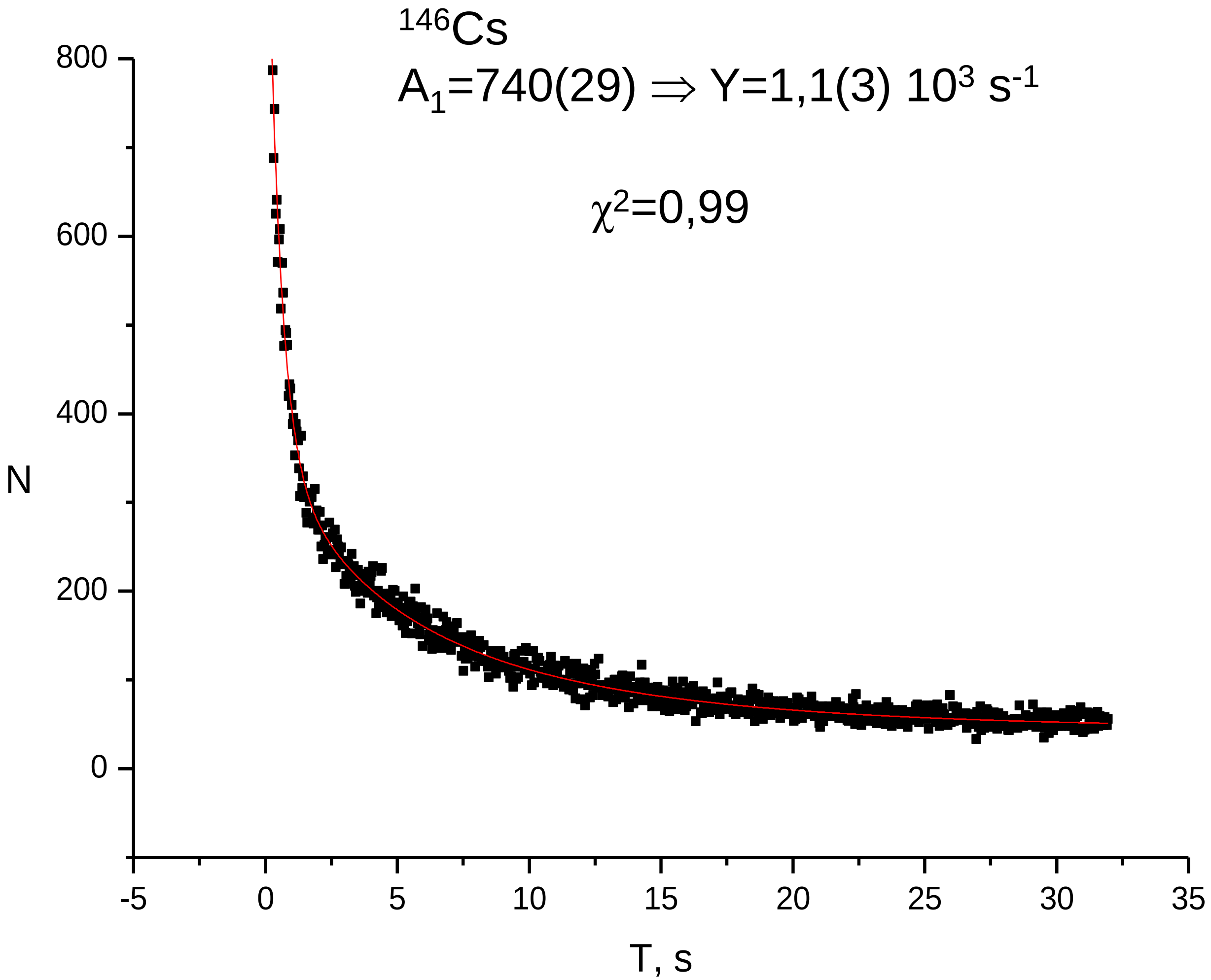
    a. Efficiency curve.
    b. Release curve.
3. Test of DEM applicability: Fr isotopes.
4. Secondary neutrons contribution to the yield of neutron rich isotopes
    a. Extraction of the neutron contribution from the comparison of neutron deficient and neutron rich Cs isotopes production efficiencies.
    b. Estimation of the neutron contribution: description of the experimental data

α- and γ- lines intensity measurement



$$Y_{\gamma(\alpha)} = \frac{S}{\exp(-\lambda\cdot tt)\cdot\left(1-\exp(-\lambda\cdot tc)\right)\cdot\left(1-\exp(-\lambda\cdot tm)\right)}\cdot\frac{1}{\varepsilon_{tr}\cdot b(E_{\gamma(\alpha)})\cdot\varepsilon_{\mathrm{det}}(E_{\gamma(\alpha)})}$$

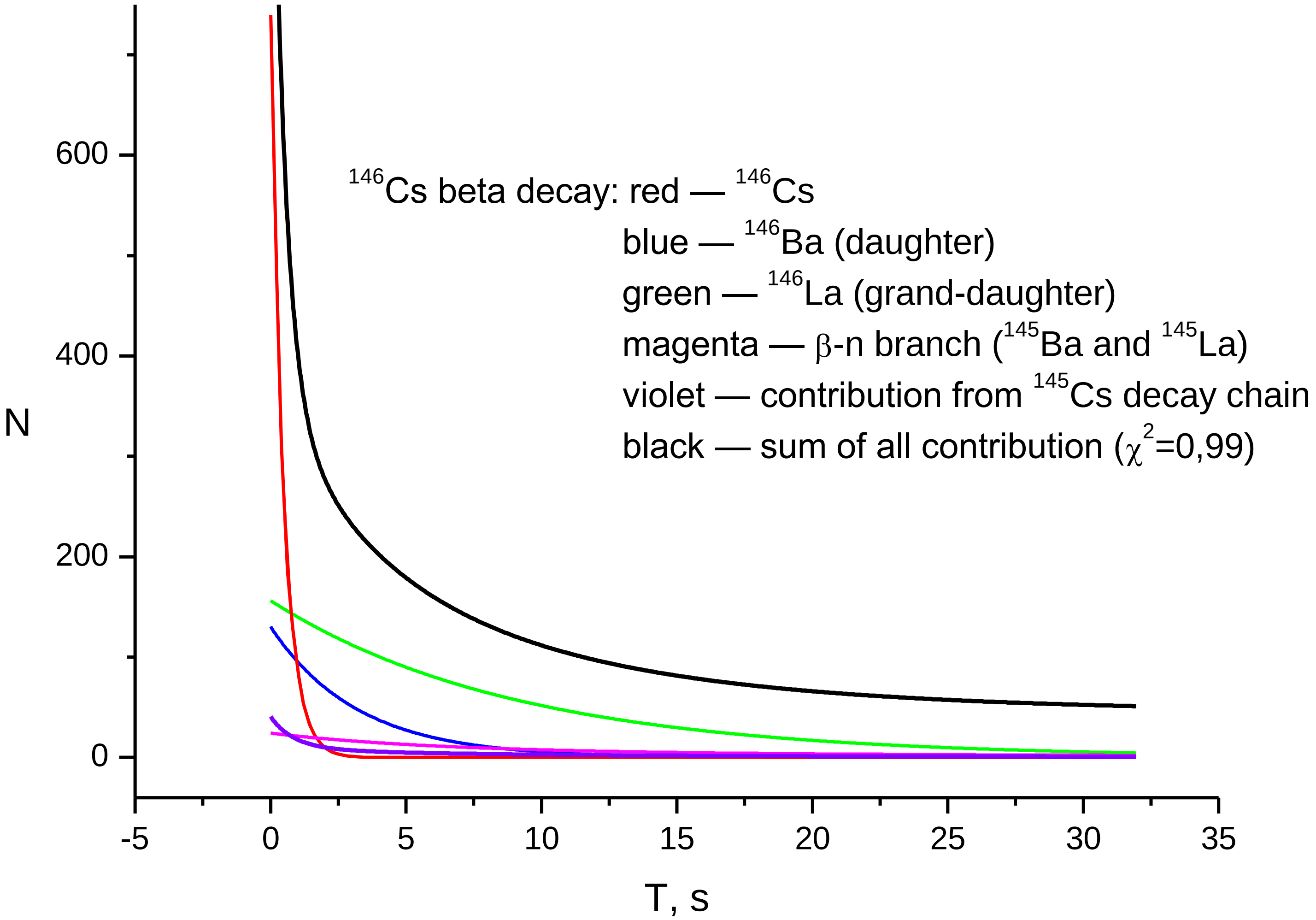
β decay curve analysis



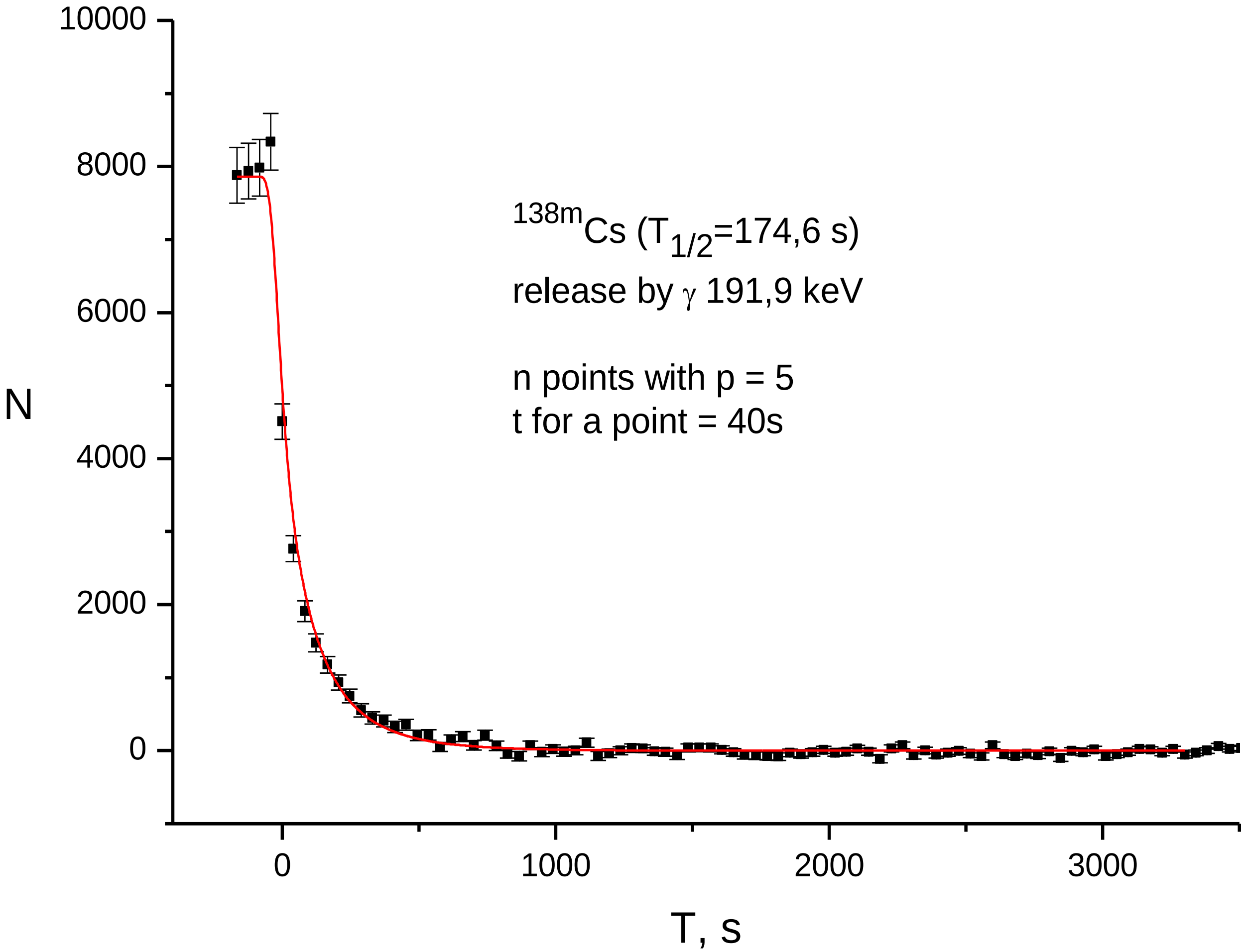
^{146}Cs

$A_1 = 740(29) \Rightarrow Y = 1{,}1(3)\ 10^3\ s^{-1}$

$\chi^2 = 0{,}99$


^{146}Cs beta decay: red — ^{146}Cs
blue — ^{146}Ba (daughter)
green — ^{146}La (grand-daughter)
magenta — β-n branch (^{145}Ba and ^{145}La)
violet — contribution from ^{145}Cs decay chain
black — sum of all contribution (χ^2=0,99)
N
0
200
400
600
-5
0
5
10
15
20
25
30
35
T, s

Release curve



^{138m}Cs ($T_{1/2}$=174,6 s)

release by γ 191,9 keV

n points with p = 5

t for a point = 40s

Diffusion-effusion model

Release curve analysis

Experimental release curve in DEM (pure diffusion):

$$R(t_k) = n \cdot \frac{6}{\pi^2} \cdot \exp(-\lambda \cdot tc) \exp(-\lambda \cdot tt) \cdot (1 - \exp(-\lambda \cdot tm)) \cdot \exp(-\lambda \cdot t_k)$$

$$\cdot \sum_{k=0}^{\infty} \frac{\exp(-k^2 \cdot \lambda_D \cdot t_k) \cdot (1 - \exp(-k^2 \cdot \lambda_D \cdot tc))}{k^2 \cdot (k^2 \cdot \lambda_D + \lambda)} \cdot (1 - \exp(-(k^2 \cdot \lambda_D + \lambda) \cdot tp))$$

$$\lambda_D = \frac{\ln(2)}{t_d}$$

Efficiency analysis:

$$\varepsilon = \frac{Y_{\text{exp}}(A)}{Y_{\text{in target}}(A)}$$

$$Y_{\text{in target}}(A) = \sigma_A \cdot th \cdot I_p$$

$$Y(A) = Y_{\text{in target}}(A) \cdot \varepsilon_{diff}(T_{1/2}) \cdot \varepsilon_{eff}(T_{1/2}) \cdot \varepsilon_{ioniz} \equiv Y_{\text{in target}}(A) \cdot \varepsilon_R(T_{1/2})$$

$$\varepsilon_{diff} = 3 \cdot \alpha^{1/2} \left[\coth(\alpha^{-1/2}) - \alpha^{1/2} \right] \qquad \alpha = \frac{T_{1/2}}{\pi^2 \cdot t_d}$$

t_d — diffusion time

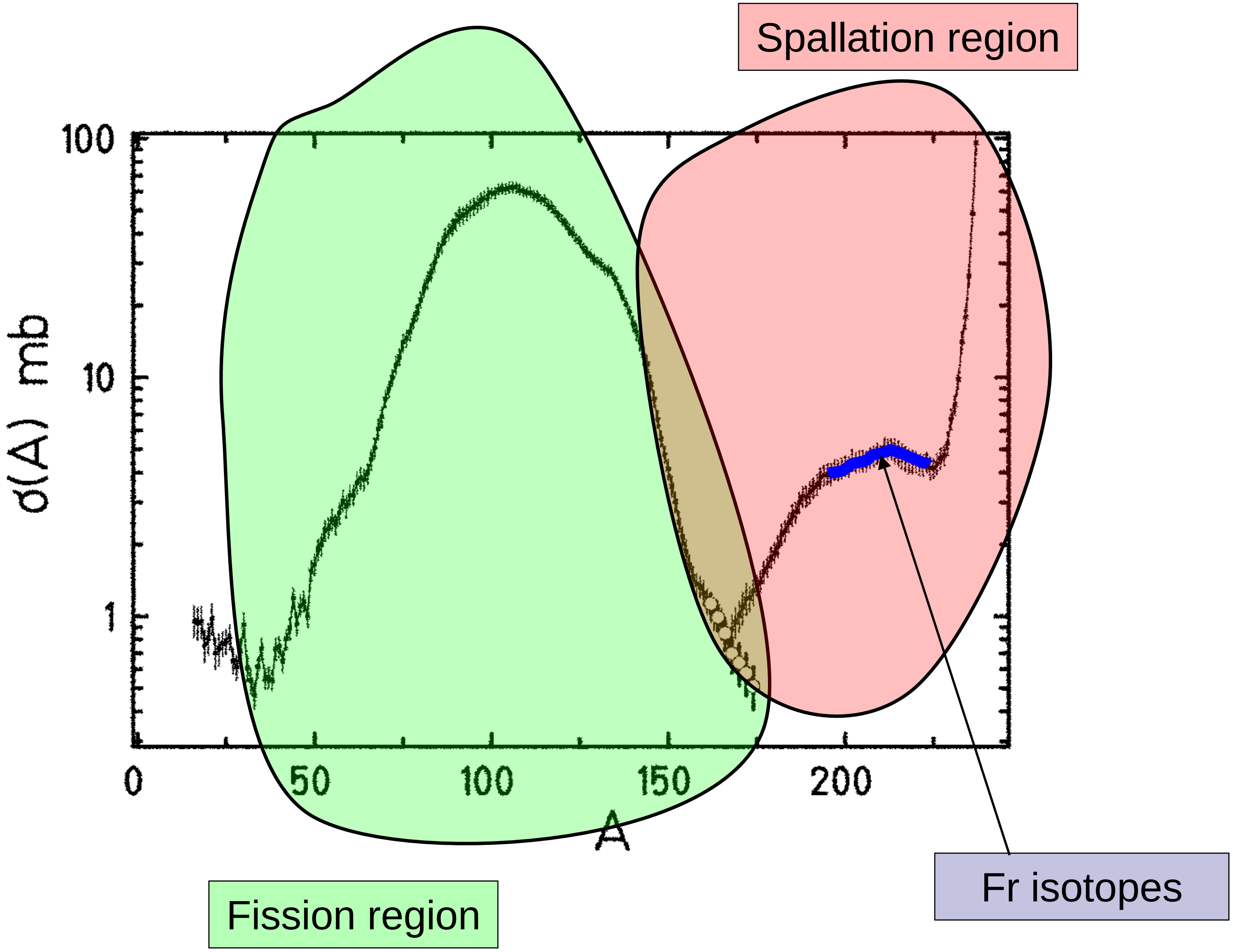
$$\varepsilon_{eff} = \frac{T_{1/2}}{T_{1/2} + t_e}$$

t_e — effusion time

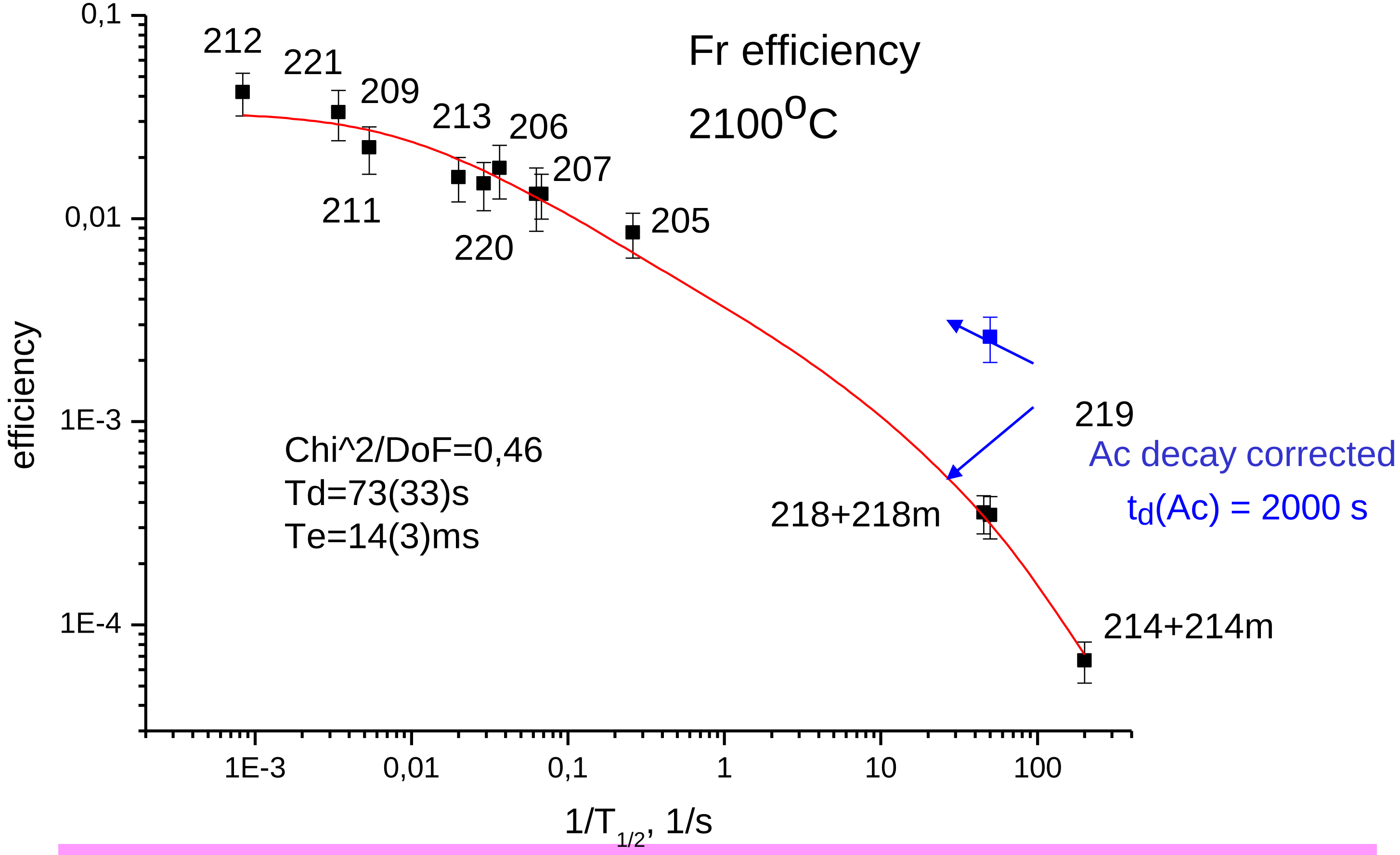
Experimental efficiency:

$$\varepsilon_R(T_{1/2}(A)) = \varepsilon_{ioniz} \cdot \varepsilon_{diff}(T_{1/2}, t_d) \cdot \varepsilon_{eff}(T_{1/2}, t_e)$$

Measured A-distribution in p (1 GeV) + ^{238}U reaction (Darmstadt)

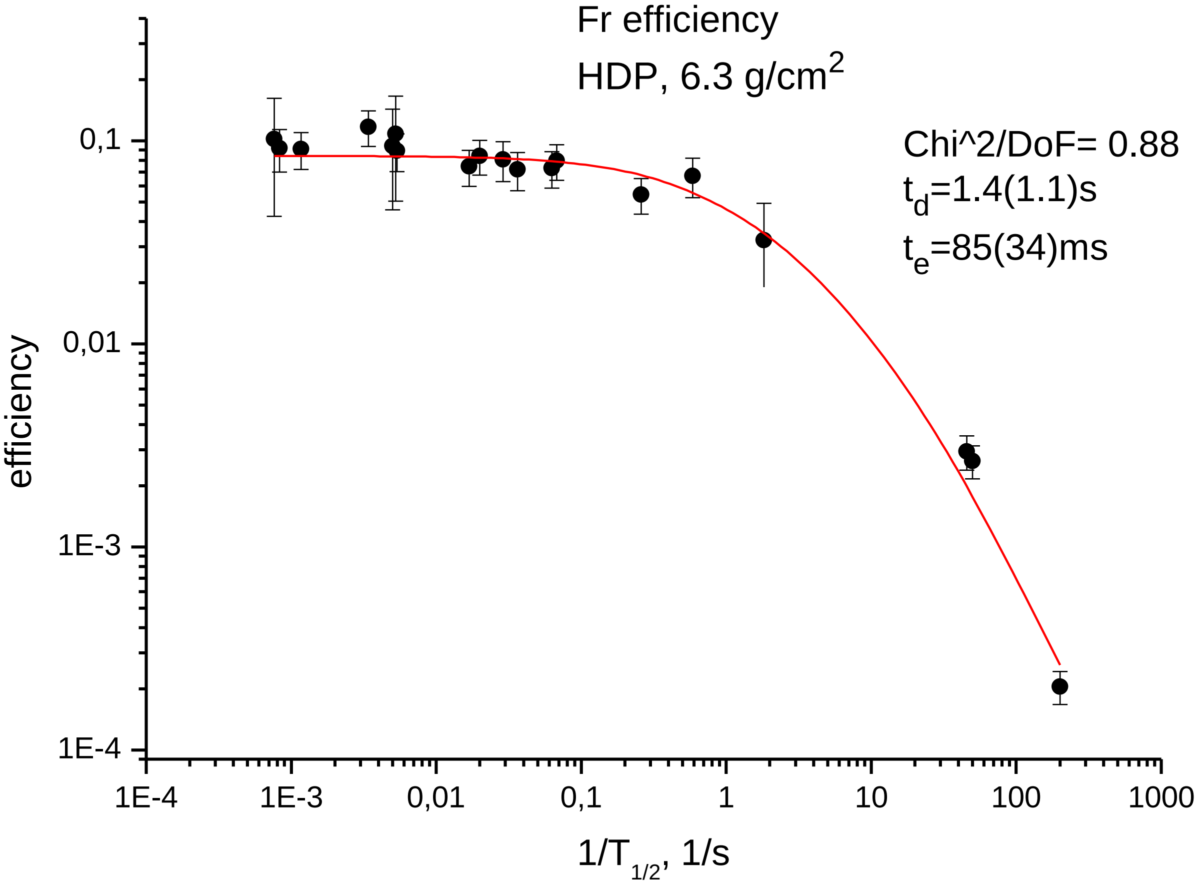


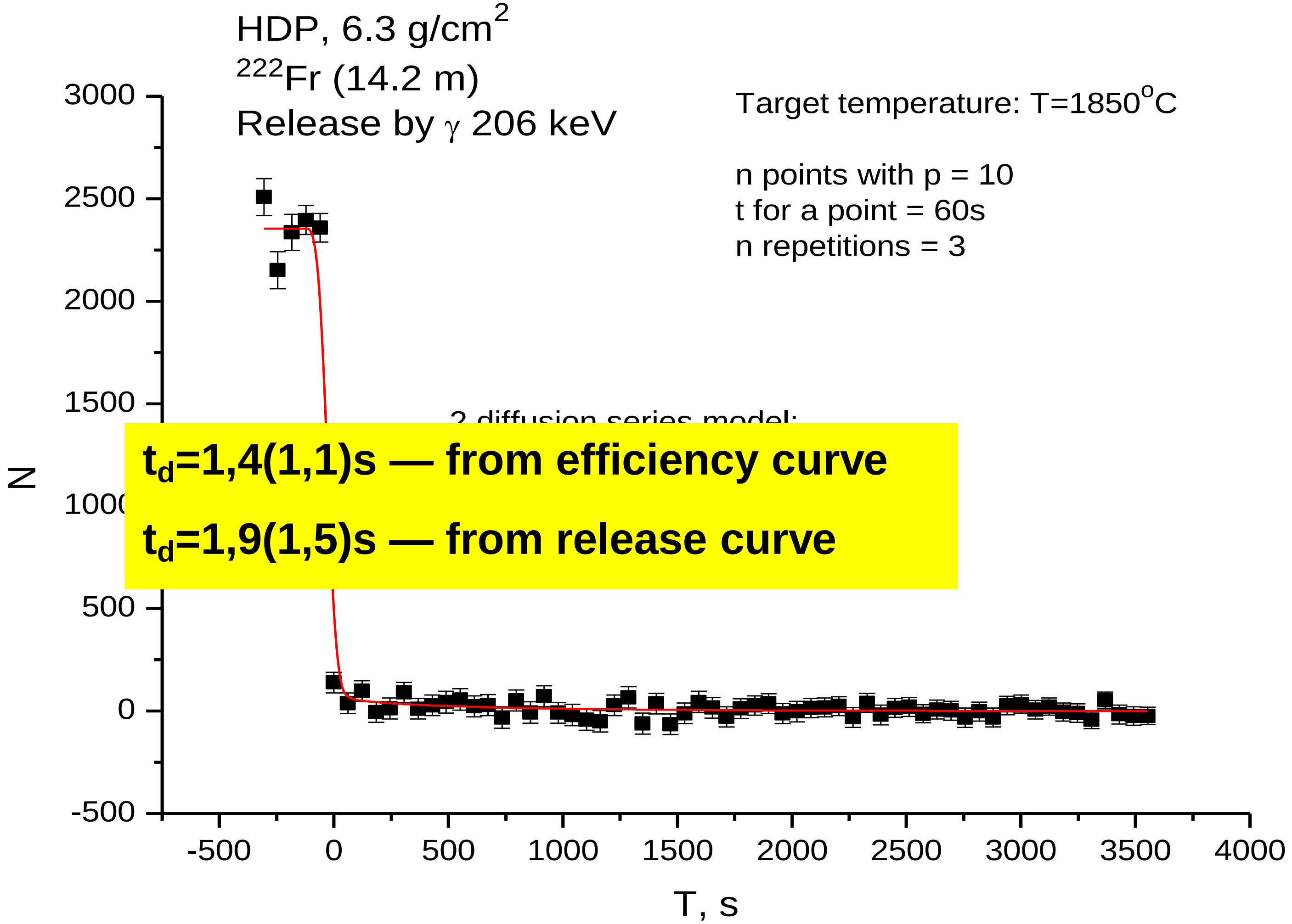
Test of DEM applicability



Correction needed:

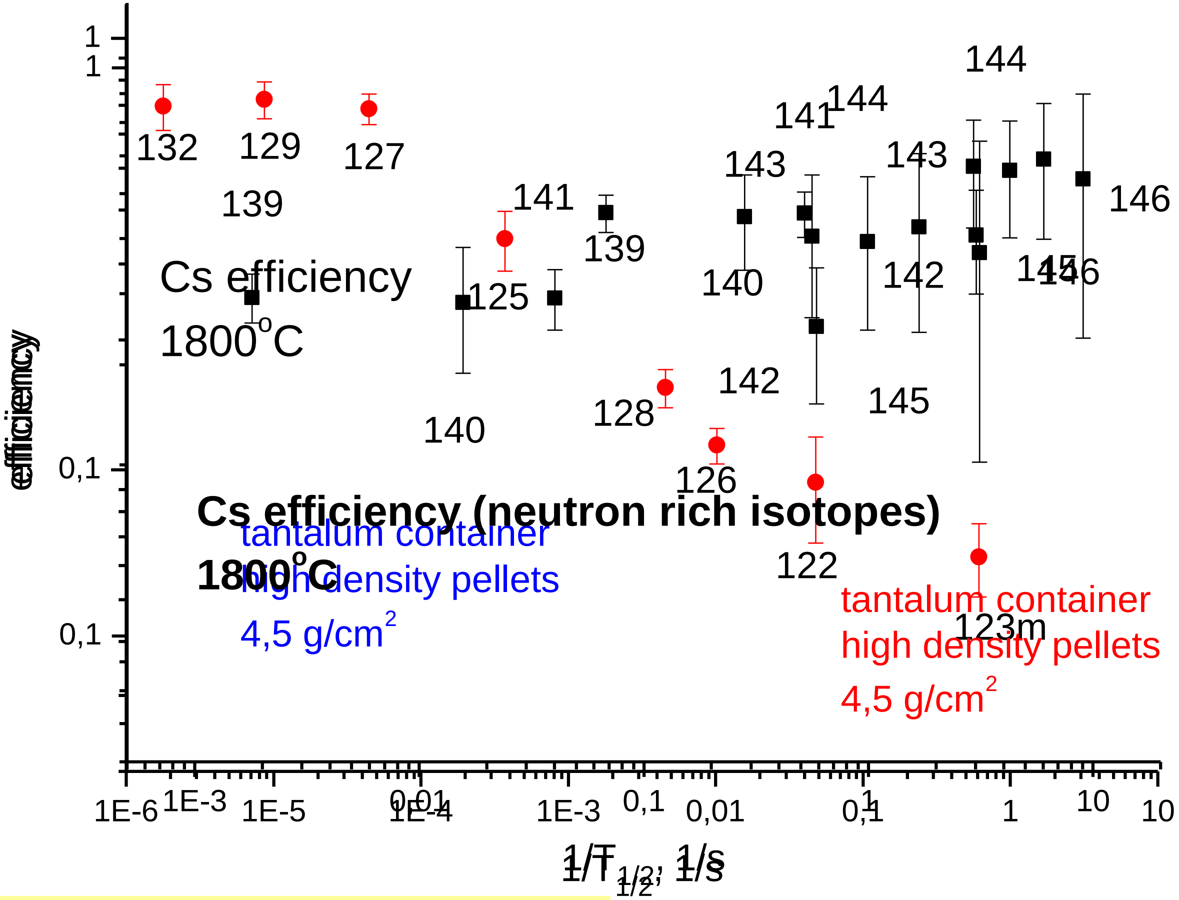
1. mother nuclei in target decay should be taken into account
2. ground state and isomer yields should not be simply added: some selfconsistency procedure should be applied in the case of different life times.


Fr efficiency
HDP, 6.3 g/cm^2
Chi^2/DoF= 0.88
t_d=1.4(1.1)s
t_e=85(34)ms
efficiency
0,1
0,01
1E-3
1E-4
1E-4
1E-3
0,01
0,1
1
10
100
1000
1/$T_{1/2}$, 1/s



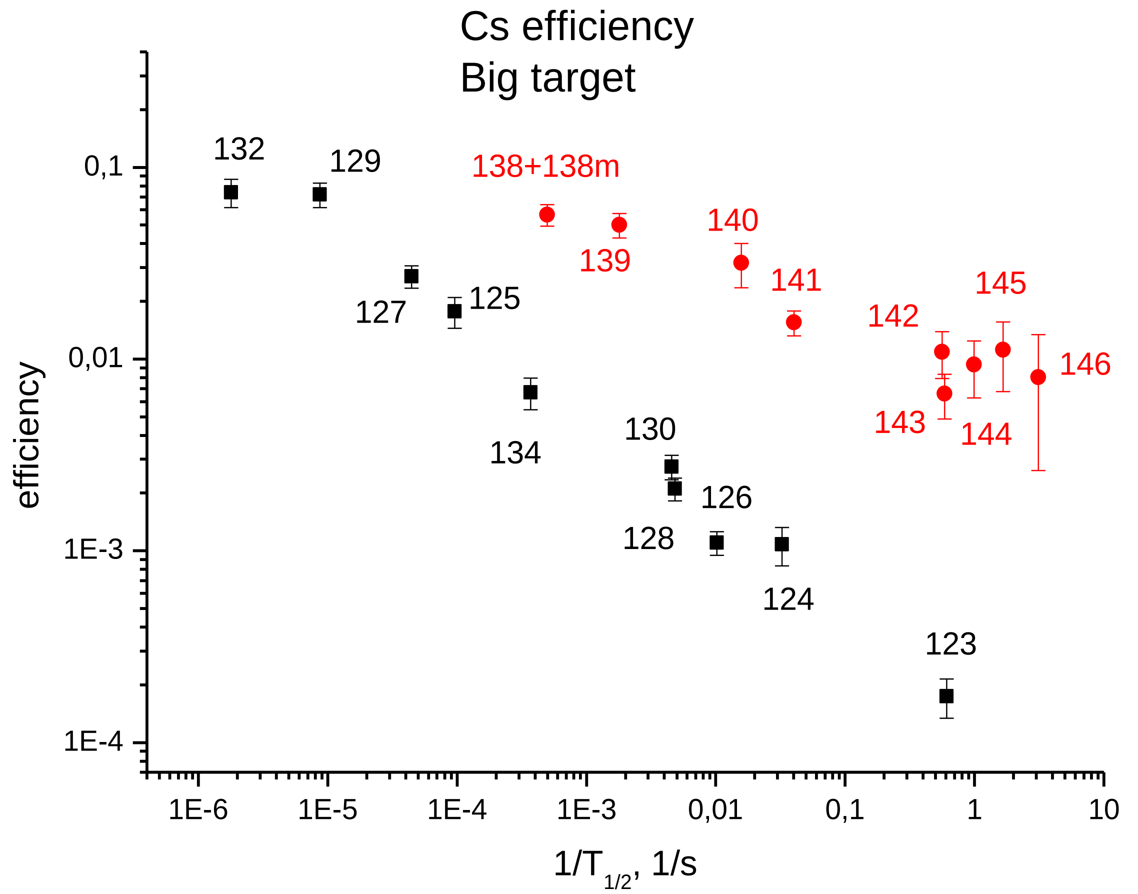
t_d=1,4(1,1)s — from efficiency curve

t_d=1,9(1,5)s — from release curve

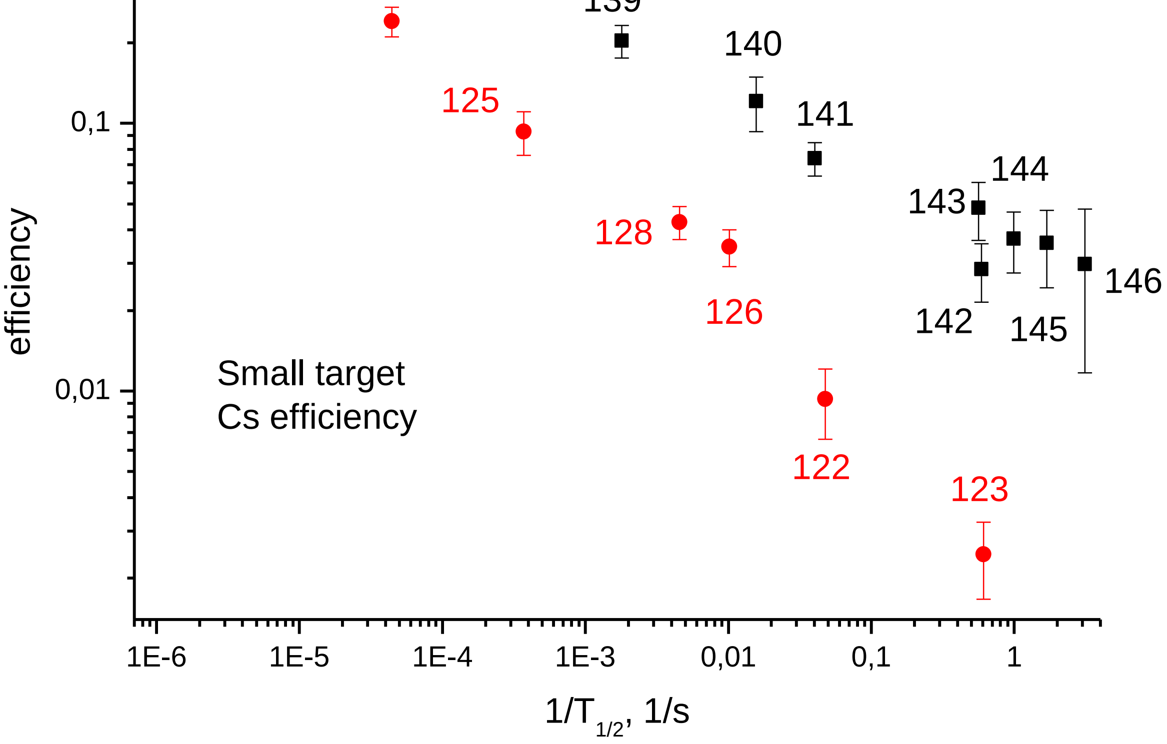


Cs efficiency curve for neutron rich isotopes calculated with pure proton cross sections can’t be explained by DEM.

Moreover: “proton” efficiencies for neutron rich and neutron deficient isotopes can’t be conciliated even after taking into account mother nuclei in-target decay

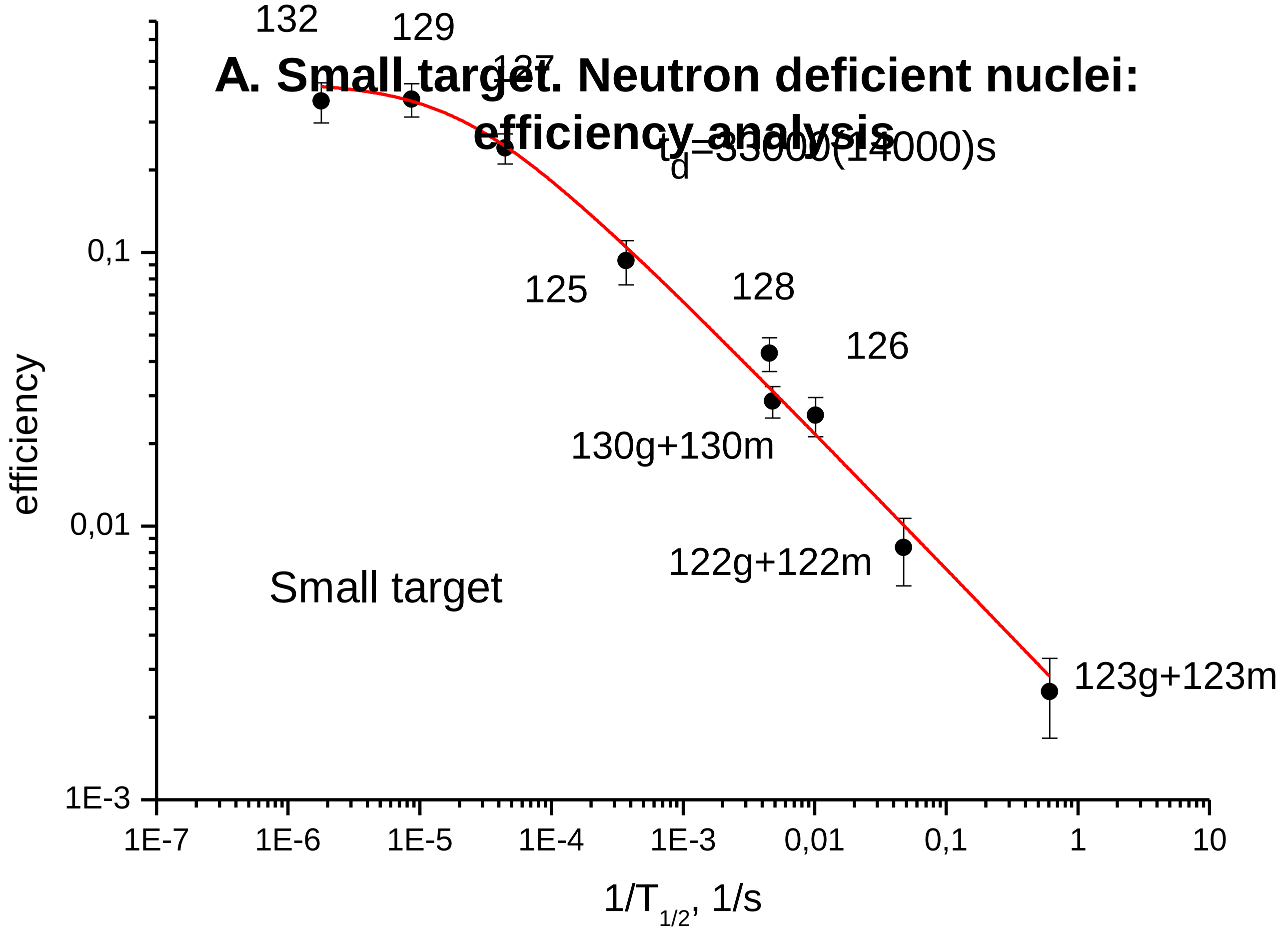


Big Uranium target
(mass of the target M=730 g,
diameter D=24,5 mm, length in the proton beam direction L=129 mm,
uranium density ρ=11,4 g/cm^3, target U thickness th= 146 g/cm^2).
Target temperature was 1900^{o}C.

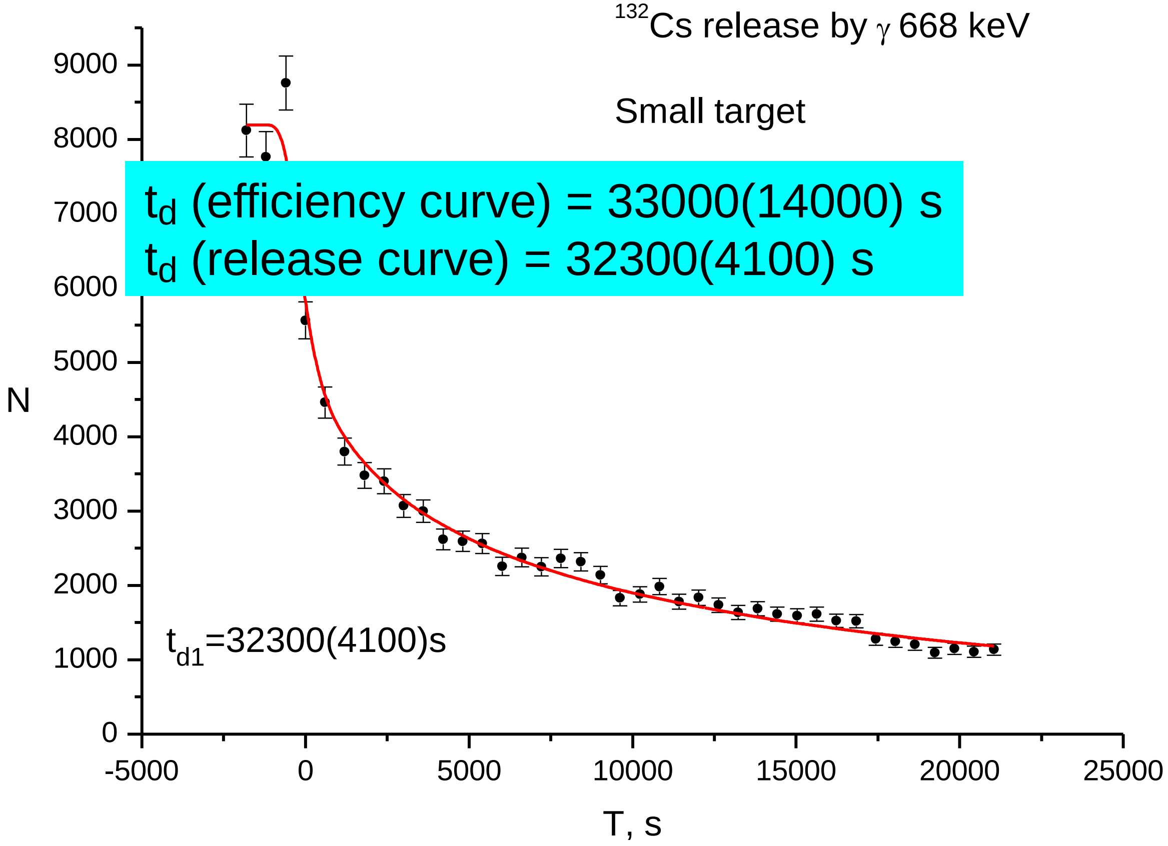


Small Uranium target (diameter *D*=11 mm,
length in the proton beam direction *L*=5,9 mm,
uranium density ρ=11 g/cm^3, target U thickness *th*= 6,3 g/cm^2)
Target temperature was 2000^{o}C

A. Small target. Neutron deficient nuclei: efficiency analysis



B. Small target. Neutron deficient nuclei: release curves analysis

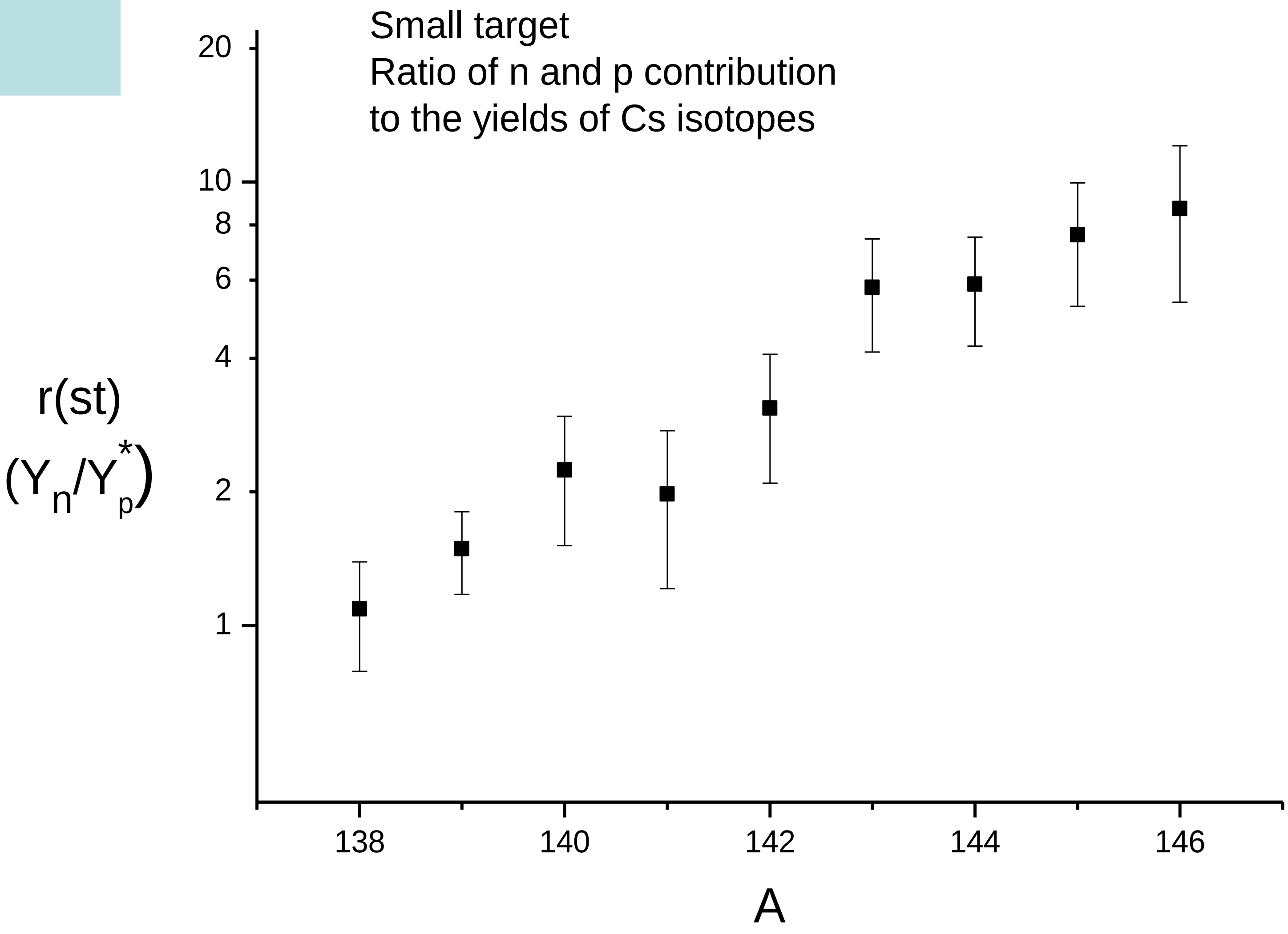


t_d (efficiency curve) = 33000(14000) s
t_d (release curve) = 32300(4100) s

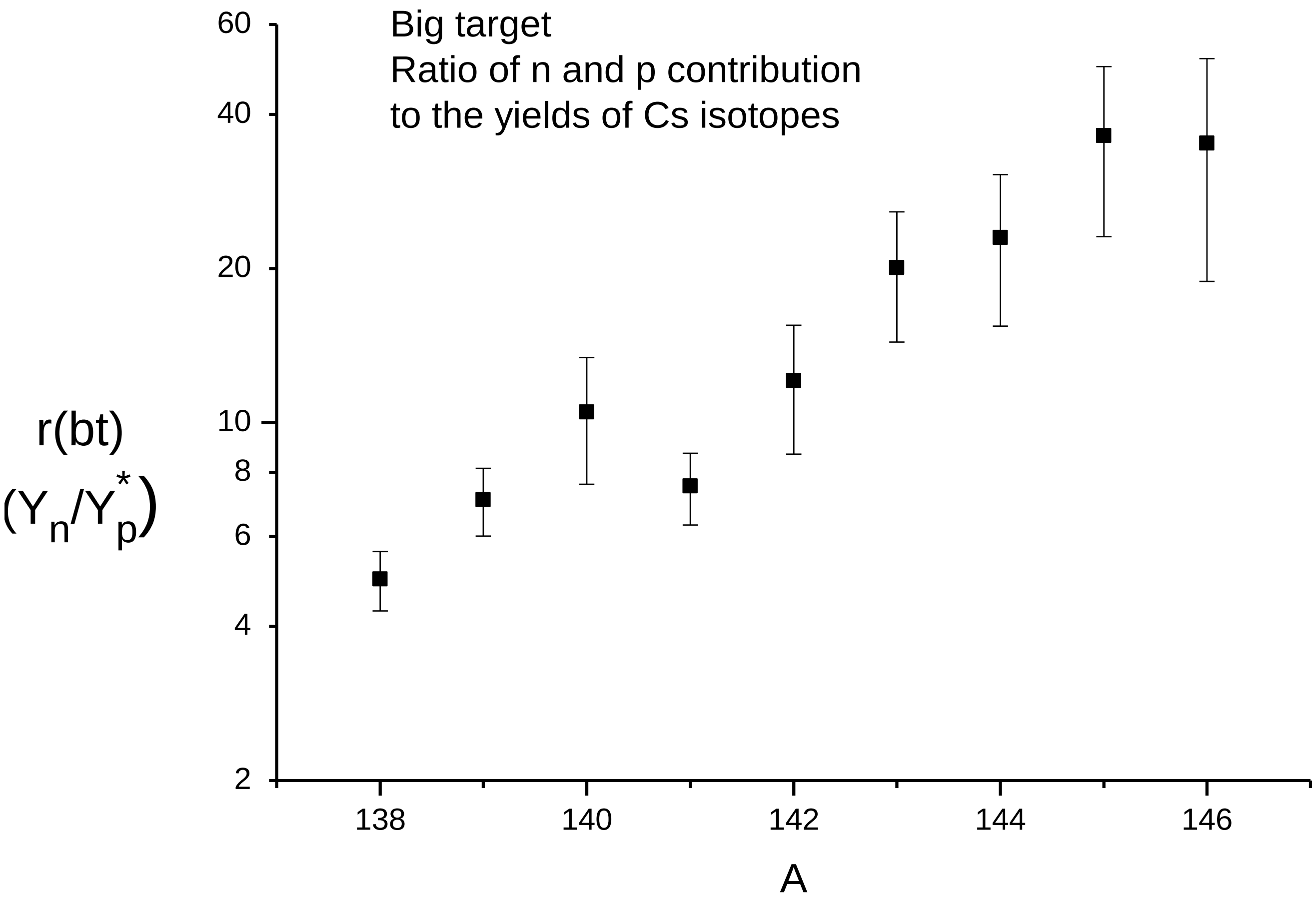
$$Y_p^* = \sigma_A(p) \cdot th \cdot I_p \cdot \varepsilon_{diff}(T_{1/2}) \cdot \varepsilon_{ioniz}$$

$$Y_n = Y_A^{\exp} - Y_p^*;$$

$$r = \frac{Y_n}{Y_p^*}$$



Small target
Ratio of n and p contribution
to the yields of Cs isotopes



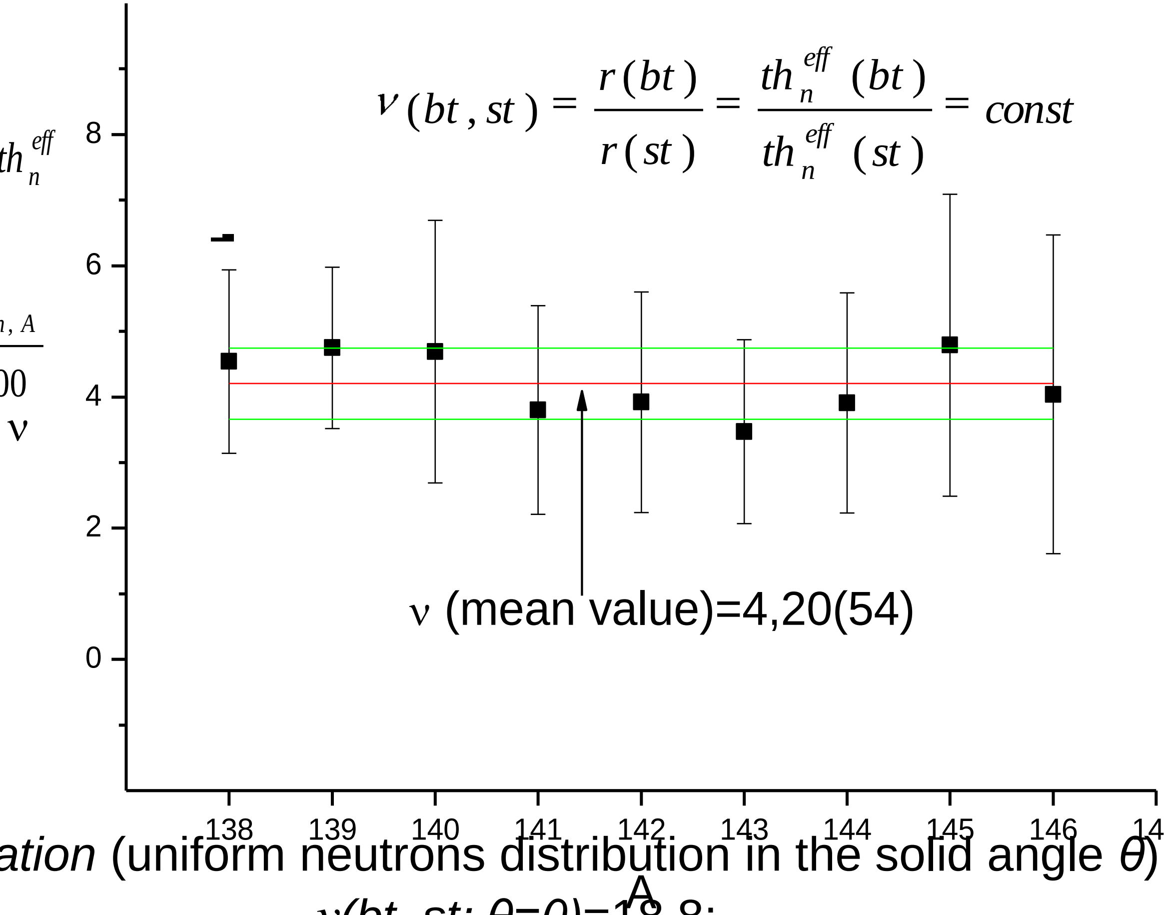
No simple scaling: thickness increases 20 times, but relative neutron contribution increases less than 5 times.

$$I_n = I_p \cdot \sigma_{p,t} \cdot th \cdot M_n$$

$$n_{fiss,n} = \sigma_{n,t} \cdot th_n$$

$$N_{fiss,n} = I_n \cdot \sigma_{n,t} \cdot th_n^{eff}$$

$$Y_n(A) = N_{fiss,n} \cdot \frac{u_{n,A}}{100}$$



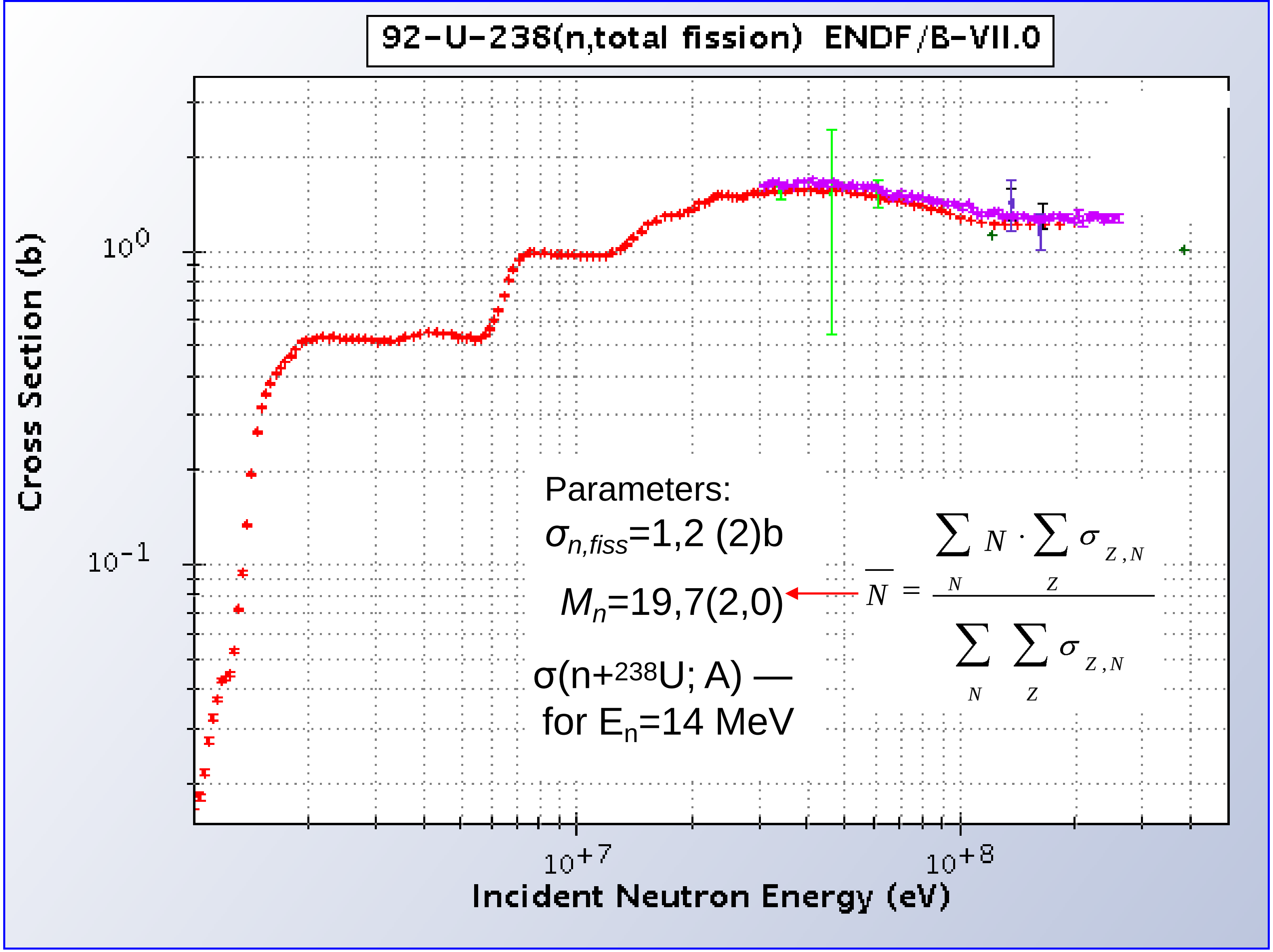
$$\nu(bt,st) = \frac{r(bt)}{r(st)} = \frac{th_n^{eff}(bt)}{th_n^{eff}(st)} = const$$

ν (mean value)=4,20(54)

Calculation (uniform neutrons distribution in the solid angle *θ*):

ν(bt, st; θ=0)=18,8;

ν(bt, st; θ=4π)=3,7.



Parameters:

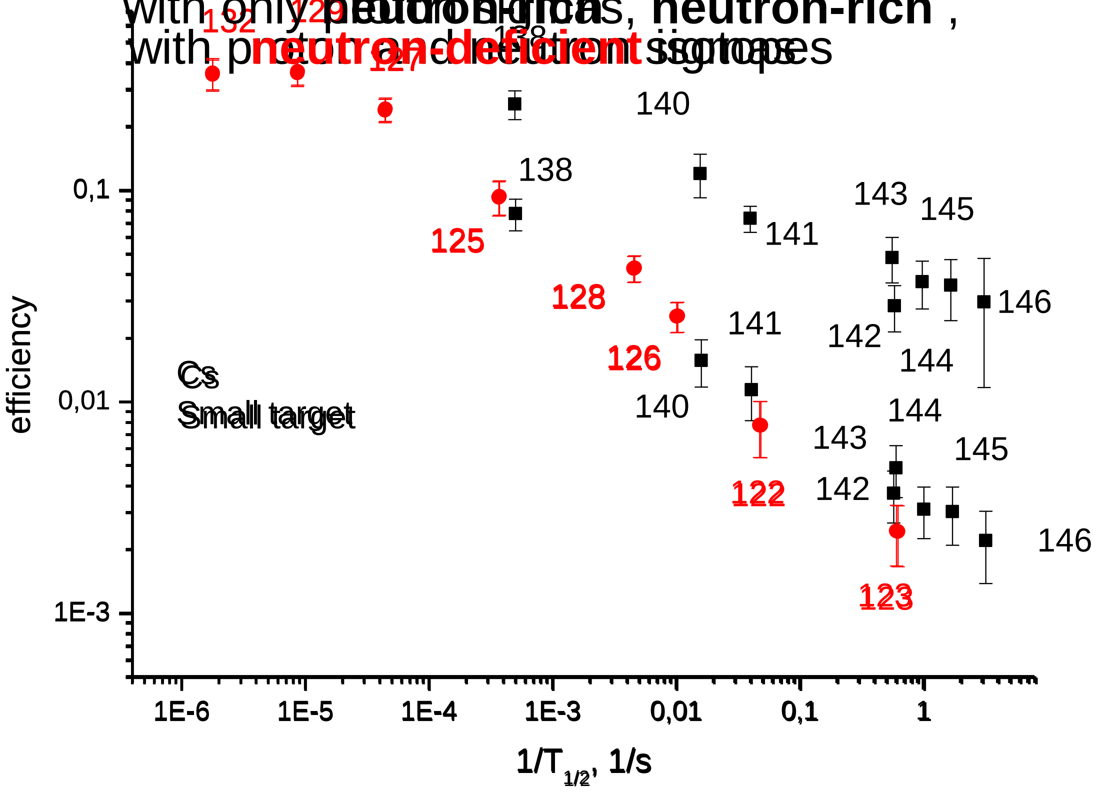
$\sigma_{n,fiss}$=1,2 (2)b

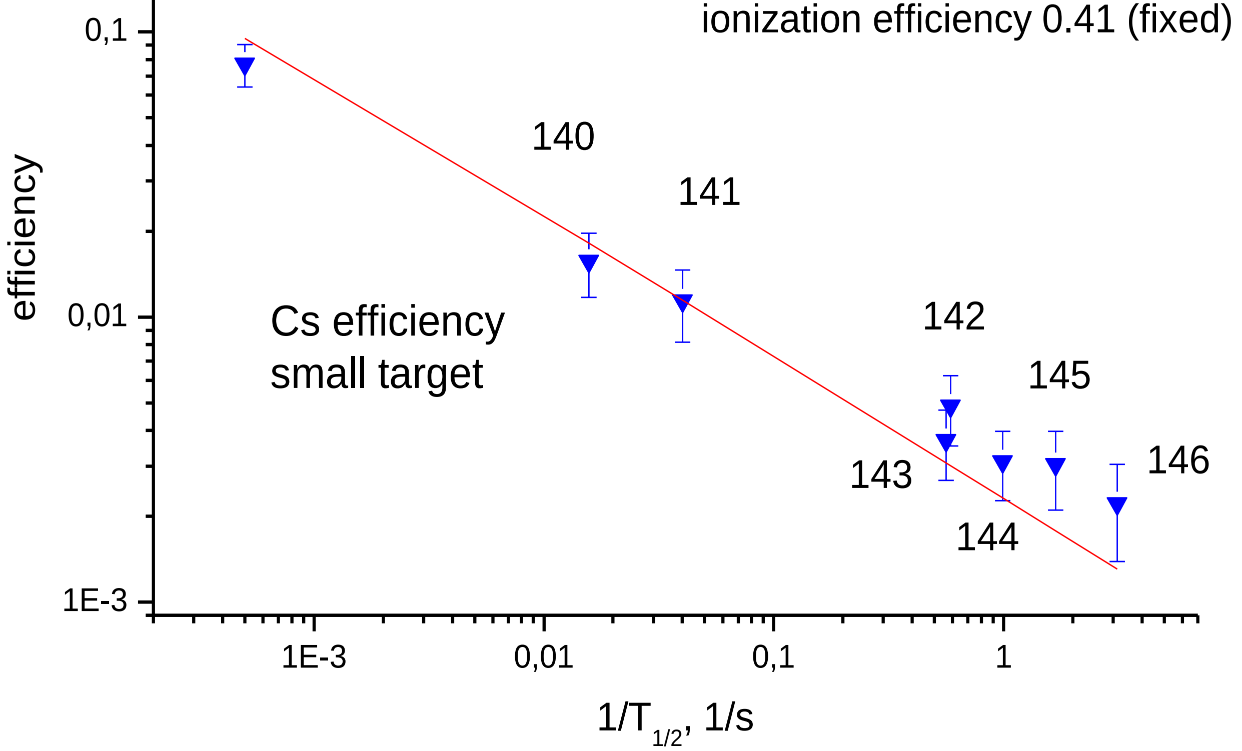
M_n=19,7(2,0) $\leftarrow$

$$\overline{N} = \frac{\sum_N N \cdot \sum_Z \sigma_{Z,N}}{\sum_N \sum_Z \sigma_{Z,N}}$$

$\sigma(n+^{238}U; A)$ —
for E_n=14 MeV

with only proton signals, **neutron-rich** ,
with proton and neutron signals
with **neutron-deficient** isotopes





T_d=33000(14000)s — from neutron deficient Cs isotopes (efficiencies)

T_d=32300(4100)s — from ^{132}Cs release curve

T_d=28600(6200)s — from neutron rich Cs isotopes (efficiencies)

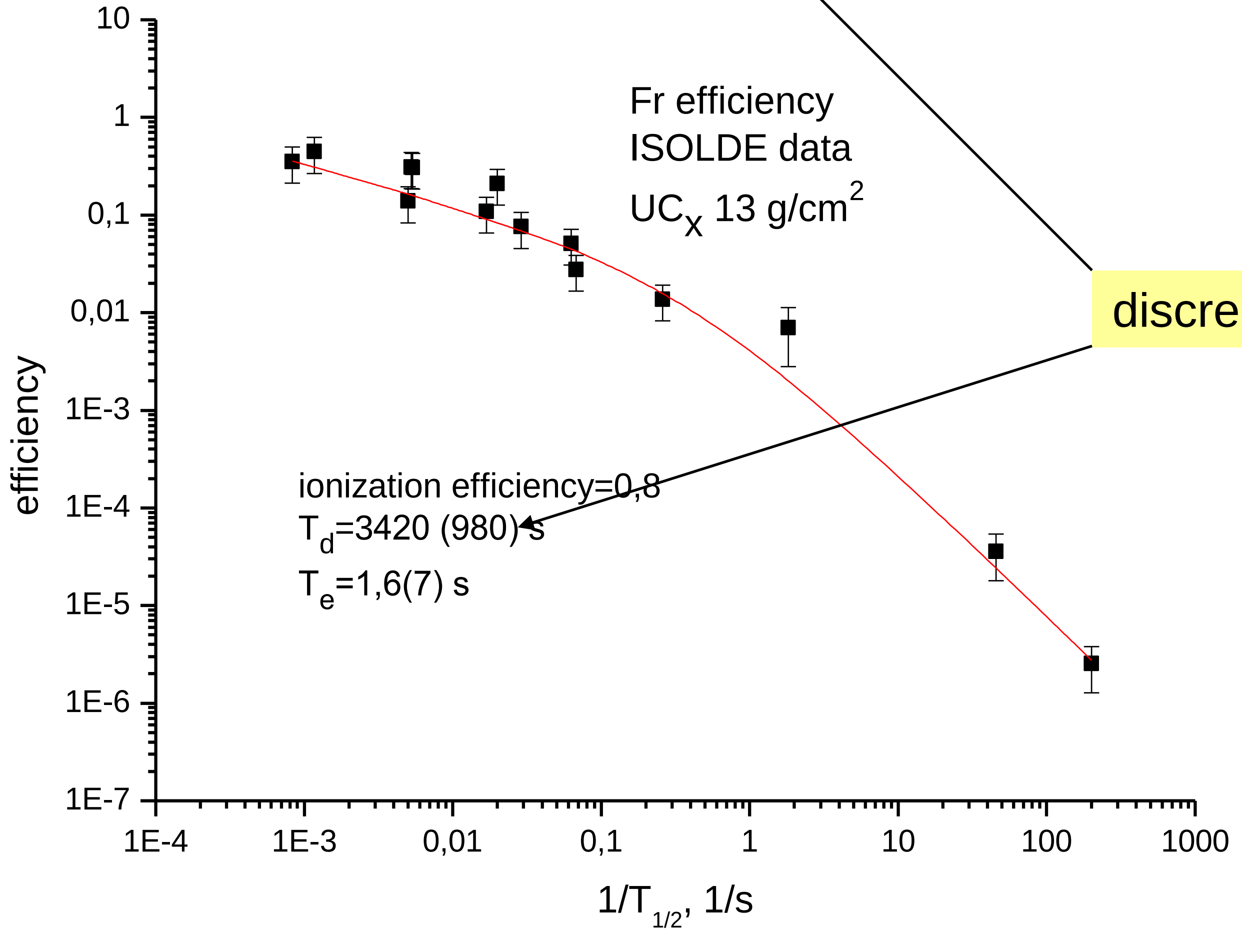
Conclusions

1. The applicability of DEM for both efficiency and release description has been shown by the analysis of the Fr isotopes yields and release.
2. Experimental yields of the mass separated Cs and Rb isotopes produced by 1 GeV proton beam in thick U targets have been analyzed in the framework of DEM. As the by-product of this analysis absolute branchings for twelve Cs, Rb and Fr nuclei as well as some isomeric ratios have been determined.
3. Comparison of the neutron rich and neutron deficient Cs isotopes production efficiencies allows to divide contributions from the direct reaction ($p+^{238}U$) and secondary reaction (secondary $n+^{238}U$) in the neutron rich Cs isotopes yields. Comparison of “the neutrons shares” in the yields favors the isotropic secondary neutrons angular distribution.
4. Simple calculations of the “neutron contribution” with the known secondary neutron multiplicity, the isotope production cross-sections in the reaction ($n_{14MeV}+^{238}U$) and the assumption of the isotropic angular distribution of the secondary neutrons describe these data fairly well.

ISOLDE

Release curve parameters (2-exponent model, ISOLDE):

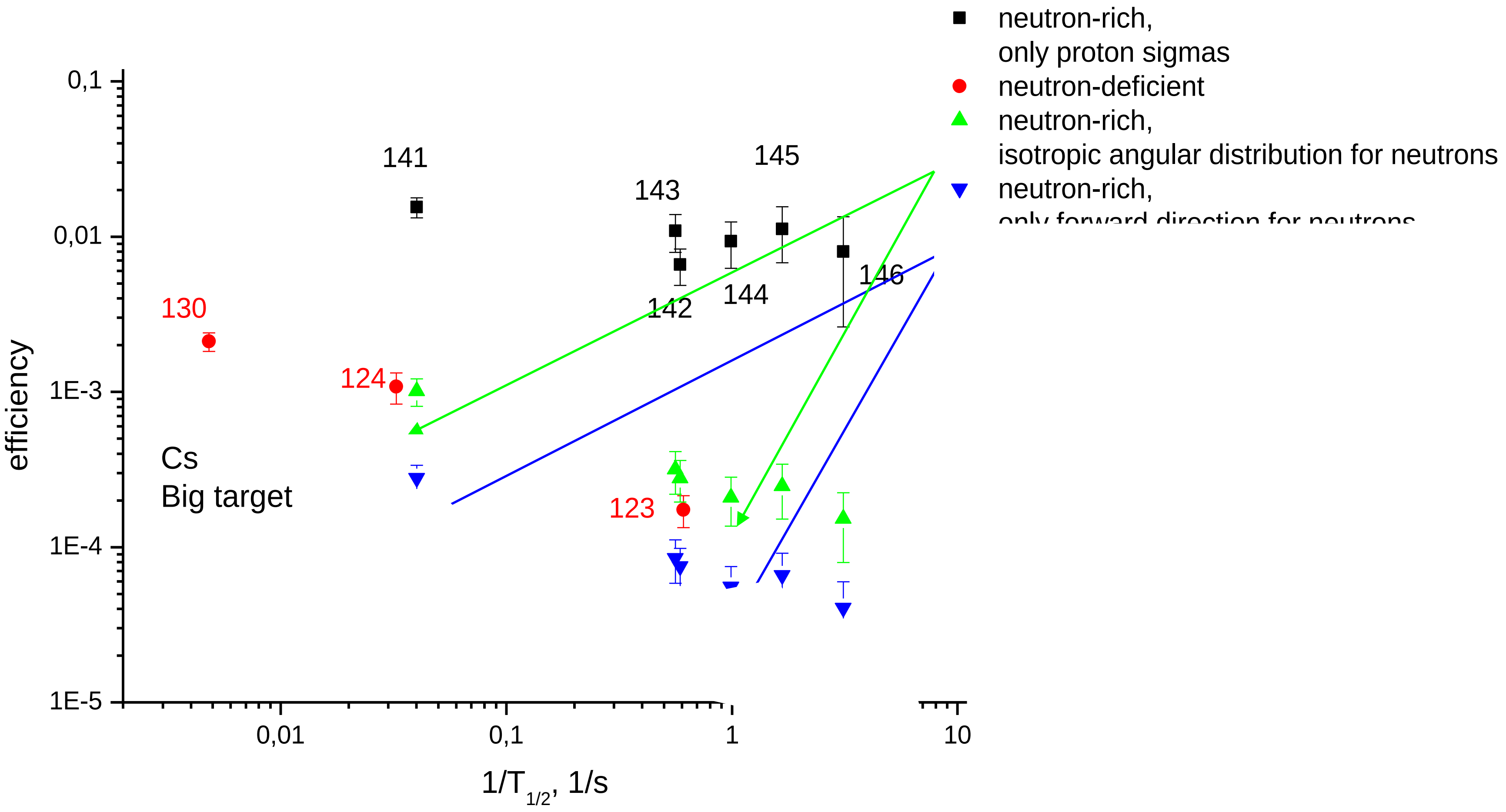
$\tau_f = 0.6$ s, $\tau_s = 6$ s, $\alpha = 0.9$

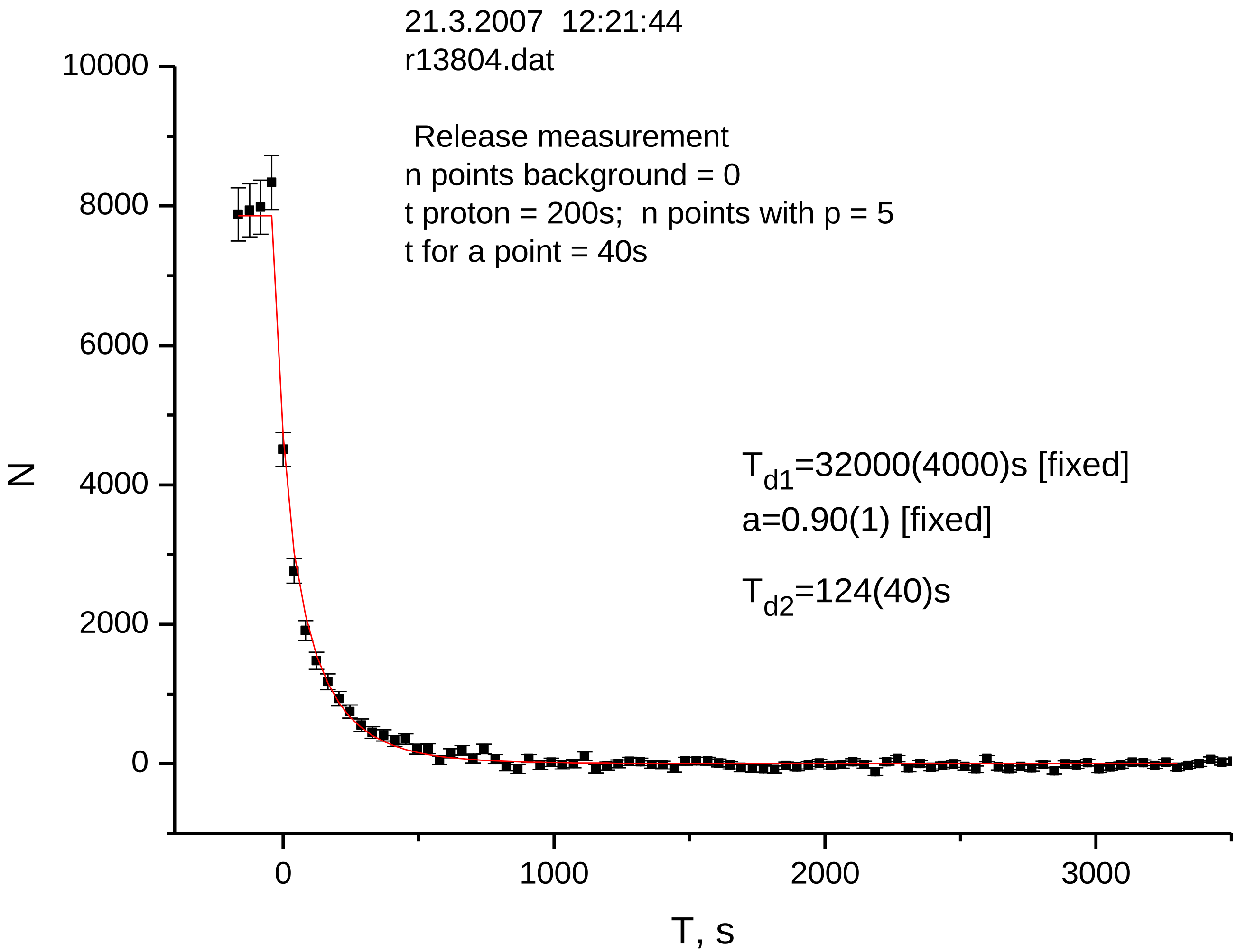


discrepancy

The reasons of discrepancy: a. release curve measurement for isotope with $T_{1/2} \ll t_d$,

b. using 2-exponent model rather than DEM for release curve description


neutron-rich,
only proton sigmas
neutron-deficient
neutron-rich,
isotropic angular distribution for neutrons
neutron-rich,
only forward direction for neutrons
efficiency
0,1
0,01
1E-3
1E-4
1E-5
0,01
0,1
1
10
1/T_{1/2}, 1/s
141
143
145
142
144
146
130
124
123
Cs
Big target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r13804.dat

Release measurement
n points background = 0
t proton = 200s; n points with p = 5
t for a point = 40s

T_{d1}=32000(4000)s [fixed]
a=0.90(1) [fixed]

T_{d2}=124(40)s

$$\begin{tikzcd}
(Z,N) \arrow[r, Rightarrow, "\beta^-"] \arrow[dr, Rightarrow, "\beta^- n"] & (Z+1,N-1) \arrow[r, Rightarrow, "\beta^-"] & (Z+2,N-2) \\
 & (Z+1,N-2) \arrow[r, Rightarrow, "\beta^-"] & (Z+2,N-3)
\end{tikzcd}$$

$$\begin{aligned} N(t) = A_i \cdot \Bigg[& C_1 \cdot Tp_1 \cdot M_1 \cdot \left(\frac{1}{\lambda_1} + \frac{\lambda_2}{\lambda_1 \cdot (\lambda_2 - \lambda_1)} + \frac{\lambda_2 \cdot \lambda_3}{\lambda_1 \cdot (\lambda_2 - \lambda_1) \cdot (\lambda_3 - \lambda_1)} \right) \cdot \exp(-\lambda_1 t) - \\ & C_2 \cdot Tp_2 \cdot M_2 \cdot \left(\frac{\lambda_1}{\lambda_2 \cdot (\lambda_2 - \lambda_1)} + \frac{\lambda_1 \cdot \lambda_3}{\lambda_2 \cdot (\lambda_2 - \lambda_1) \cdot (\lambda_3 - \lambda_2)} \right) \cdot \exp(-\lambda_2 t) + \\ & C_3 \cdot Tp_3 \cdot M_3 \cdot \left(\frac{\lambda_1 \cdot \lambda_2}{\lambda_3 \cdot (\lambda_3 - \lambda_1) \cdot (\lambda_3 - \lambda_2)} \right) \cdot \exp(-\lambda_3 t) \Bigg] \end{aligned}$$

where: $C_i = 1 - \exp(-\lambda_i \cdot tc);\ Tp_i = \exp(-\lambda_i \cdot tt);\ M_i = 1 - \exp(-\lambda_i \cdot tm)$

Contribution of each decay channel to the decay curve:

1 — index for the main isotope; 2 — for the daughter isotope; 3 — for the grand-daughter isotope

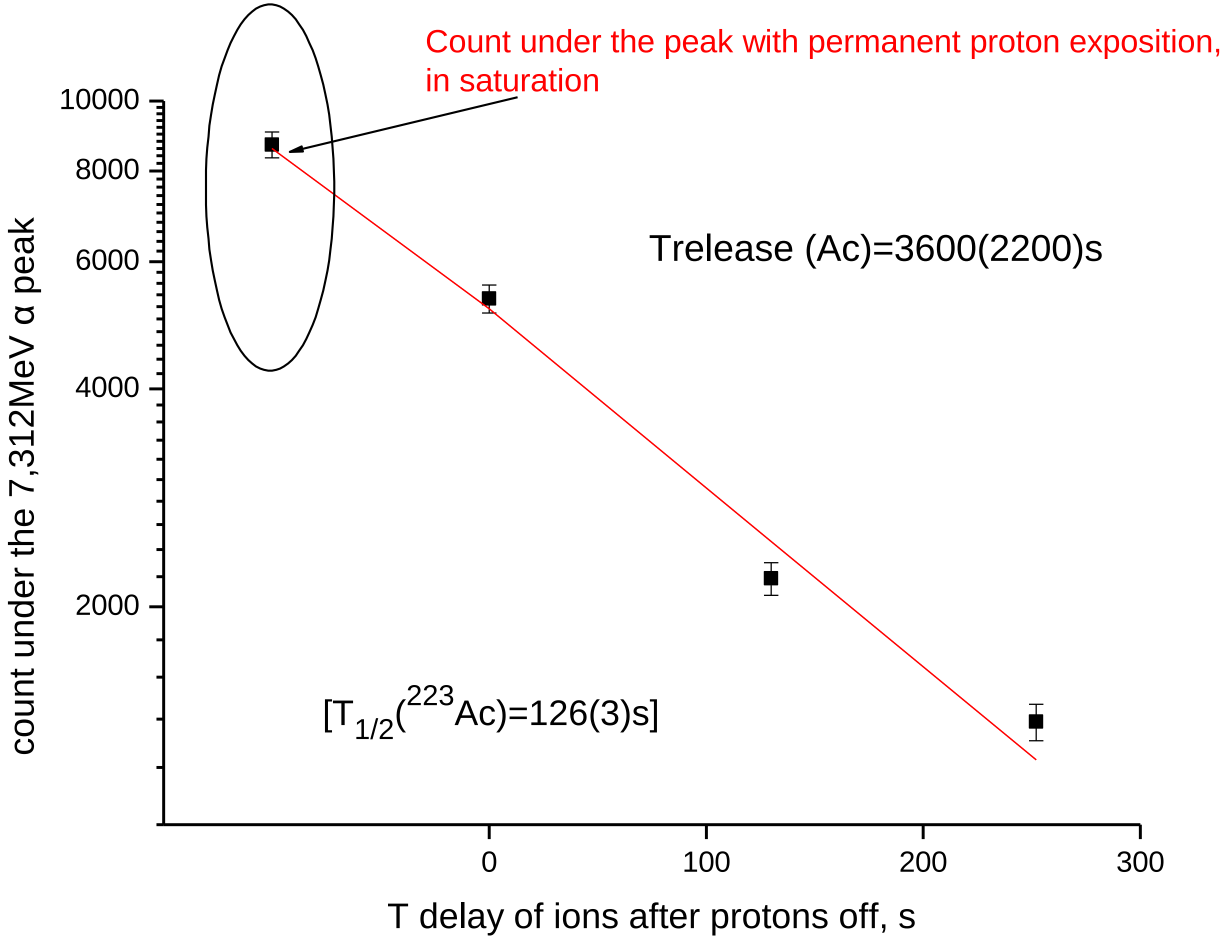
λ_i — decay constant for nuclide *i*

tm — measurement time (for a point on the curve)

tc — collection time

tt — time of the tape driving (waiting time included)

Experimental determination of release time for Ac isotopes



Count under the peak with permanent proton exposition, in saturation

Trelease (Ac)=3600(2200)s

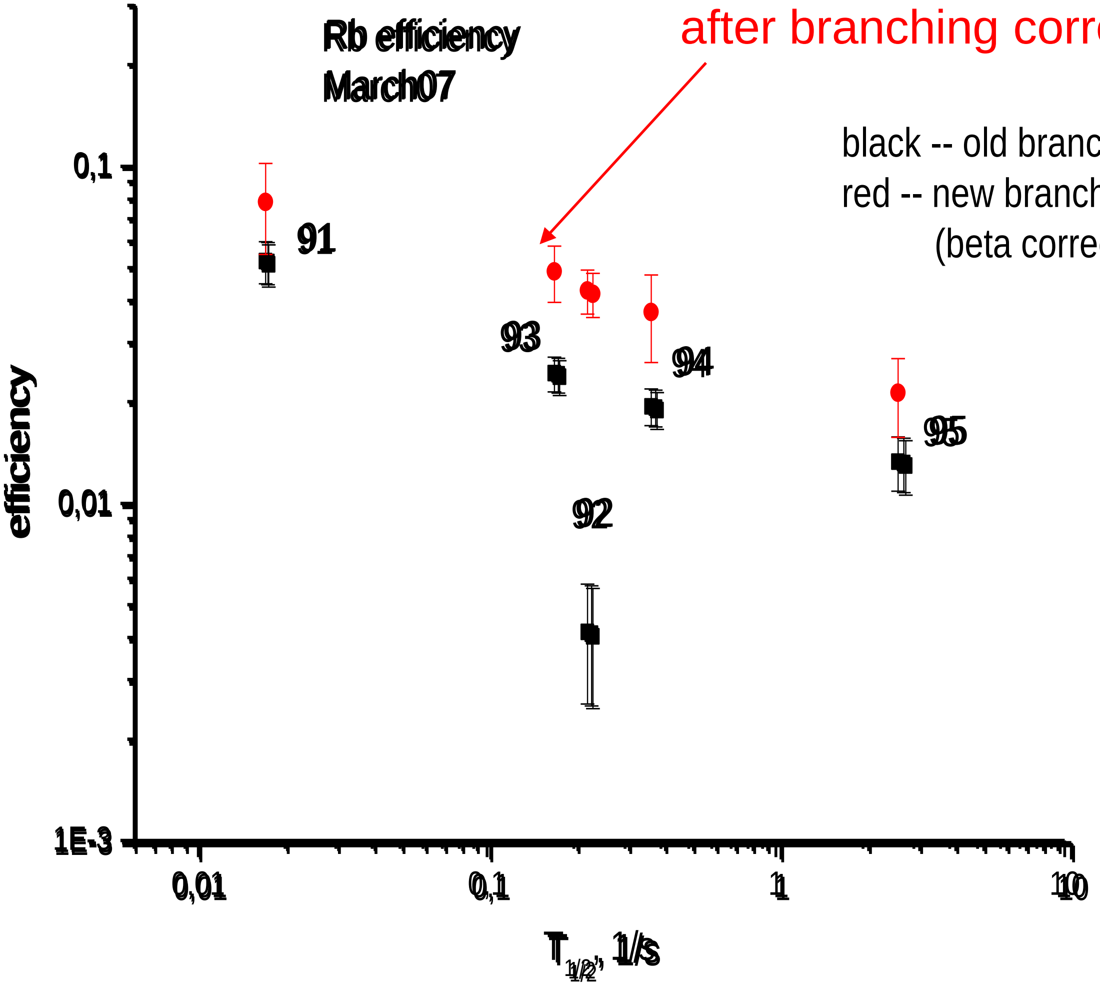
[$T_{1/2}$(^{223}Ac)=126(3)s]

^{223}Ac ($T_{1/2}$=2,1min, σ(p+^{238}U, ^{223}Ac)=2,54mb) $\xrightarrow{\alpha}$
^{219}Fr ($T_{1/2}$=20ms, σ (p+^{238}U, ^{219}Fr) =0,44mb),

A(Rb)	E gamma	br %, NDS	br %, present work
91	93,63	33,8(2,0)	20,2(3,2)
92	814,98	33(12)	3,2(0,2)
93	432,61	20,2(1,0)	9,5(1,7)
94	836,90	87,1(0,4)	44,2(6,4)
95	352,02	49(3)	29,3(6,3)

A(Cs)	E gamma	br %, NDS	br %, present work
140	602,35	53,3(2,0)	32,9(5,7)
142	359,60	27,2(2,7)	21,0(3,8)
143	195,55	12,6(1,9)	11,1(2,4)
144	199,33	only relative	31,9(7,3)
145	175,40	19,8(2,4)	10,7(2,4)
146	181,02	only relative	27,1(6,3)

after branching correction



black -- old branchings
red -- new branchings
(beta corrected)