Roman Sagaidak

Flerov Laboratory of Nuclear Reactions, Joint Institute for Nuclear Research, Dubna, Moscow region, Russian Federation

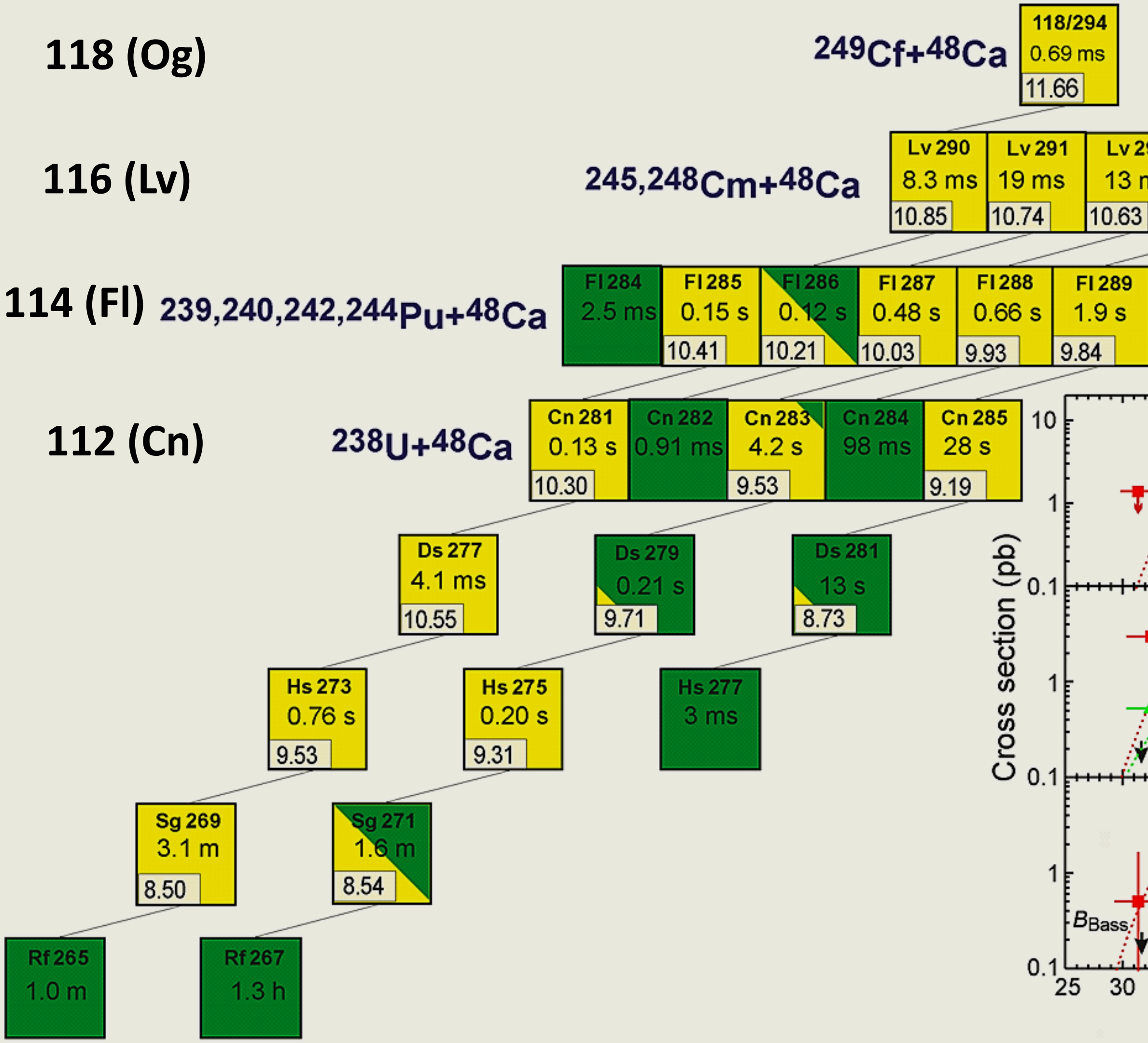
Durability of actinide targets and metallic foils in experiments on synthesis of superheavy nuclei

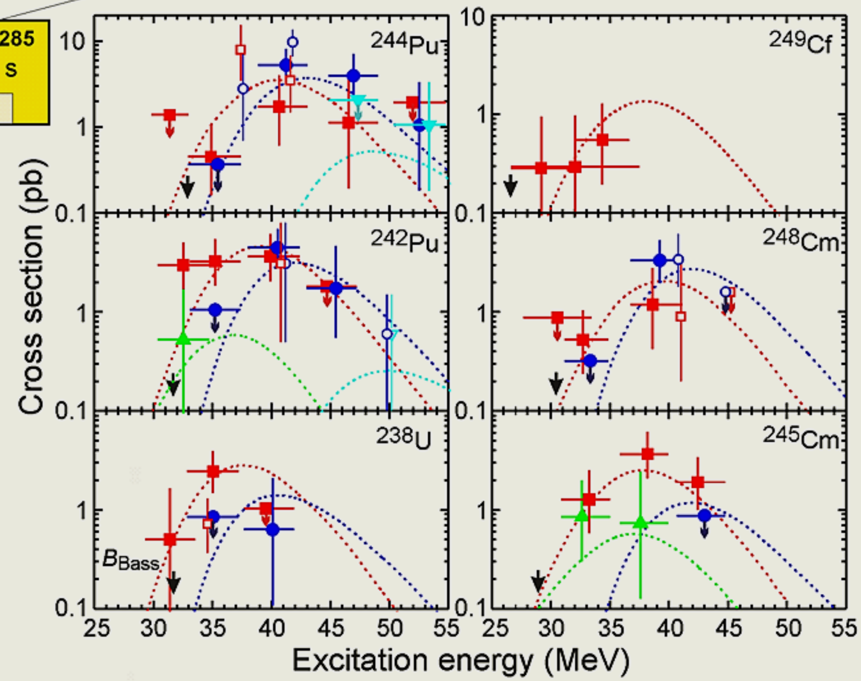
- **Motivation**
- **Sputtering of target (foil) atoms**
- **Target temperature**
- **Summary**

Superheavy nuclei production in fusion-evaporation reactions

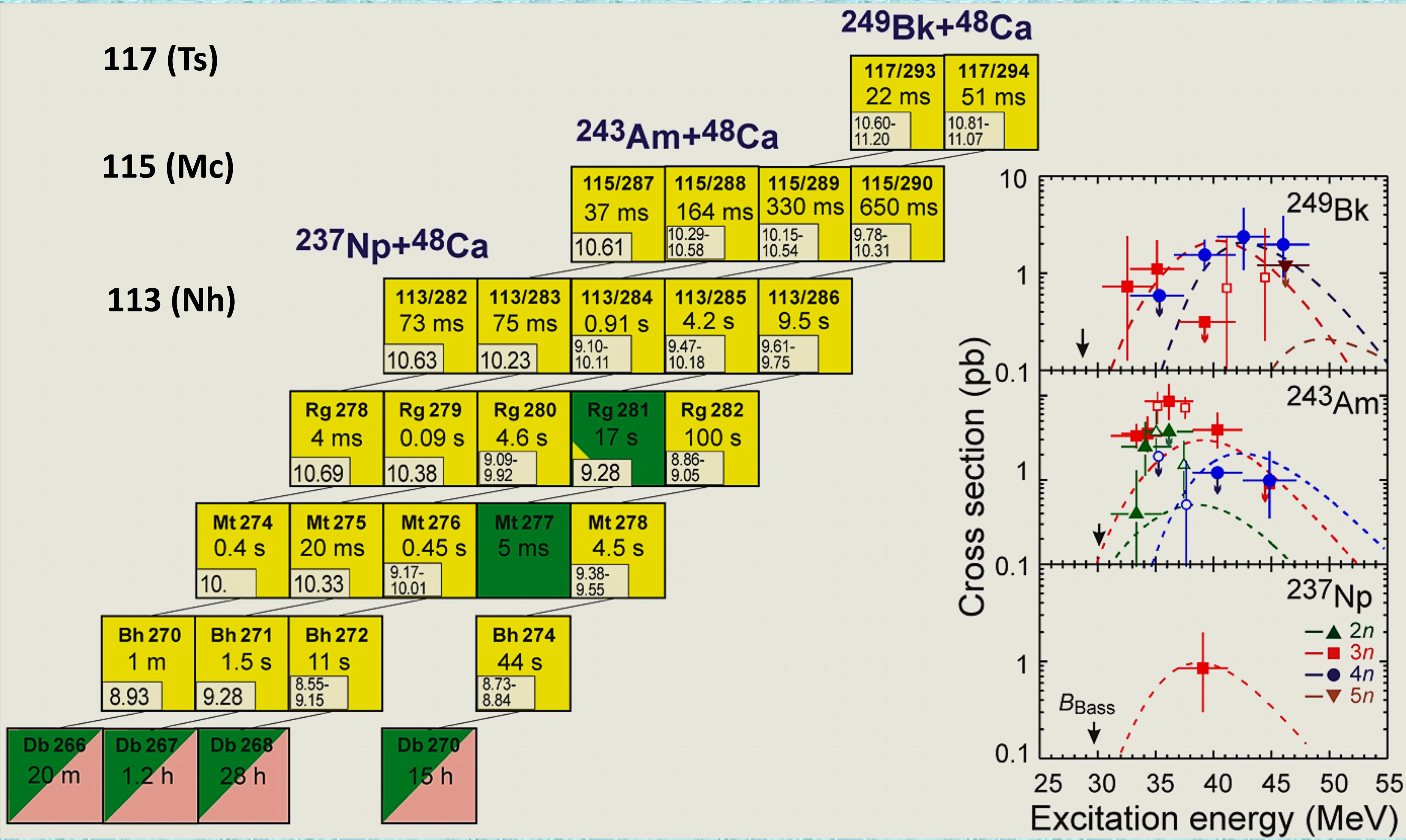
Even-Z nuclei – products of U,Pu,Cm,Cf(^{48}Ca,*xn*) reactions

Plenary talk of Yu.Ts. Oganessian and review paper Yu.Ts. Oganessian, V.K. Utyonkov, Nucl. Phys. A944 (2015) 62



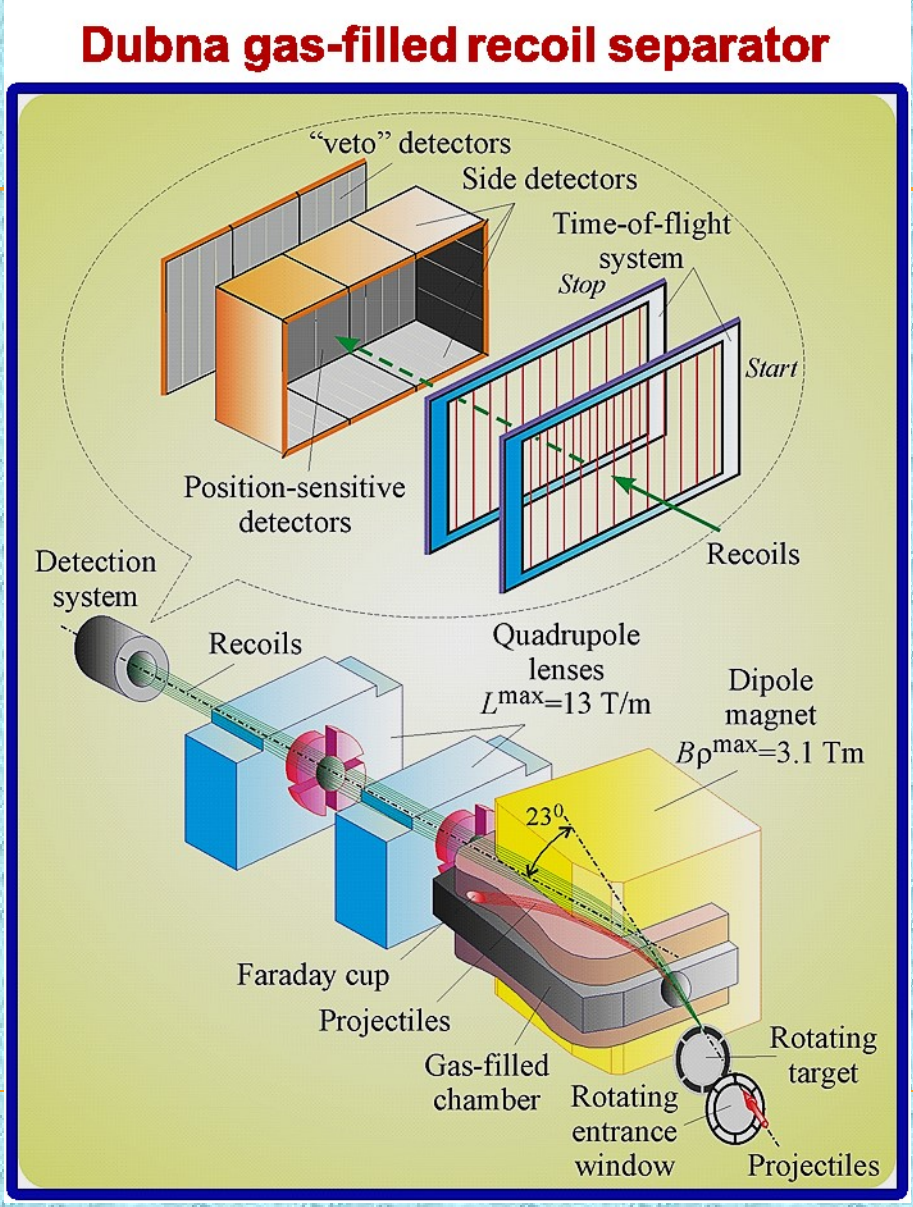


Odd-Z nuclei – products of Np,Am,Bk(^{48}Ca,*xn*) reactions



Cyclotron U400:
I(^{48}Ca)≃1 pμA
ε_{tr}(DGFRS)≃35%
Suppression factors:
^{48}Ca: ~10^{17}
PLP: ~$6\cdot10^{14}$
TLP: 10^4–10^6

Yields of SHN in
^{48}Ca+AT experiments:
(0.6–6) events/month
at CS=(1–10) pb



Dubna gas-filled recoil separator (DGFRS)

Cyclotron U400:
$I(^{48}Ca) \simeq 1$ pμA
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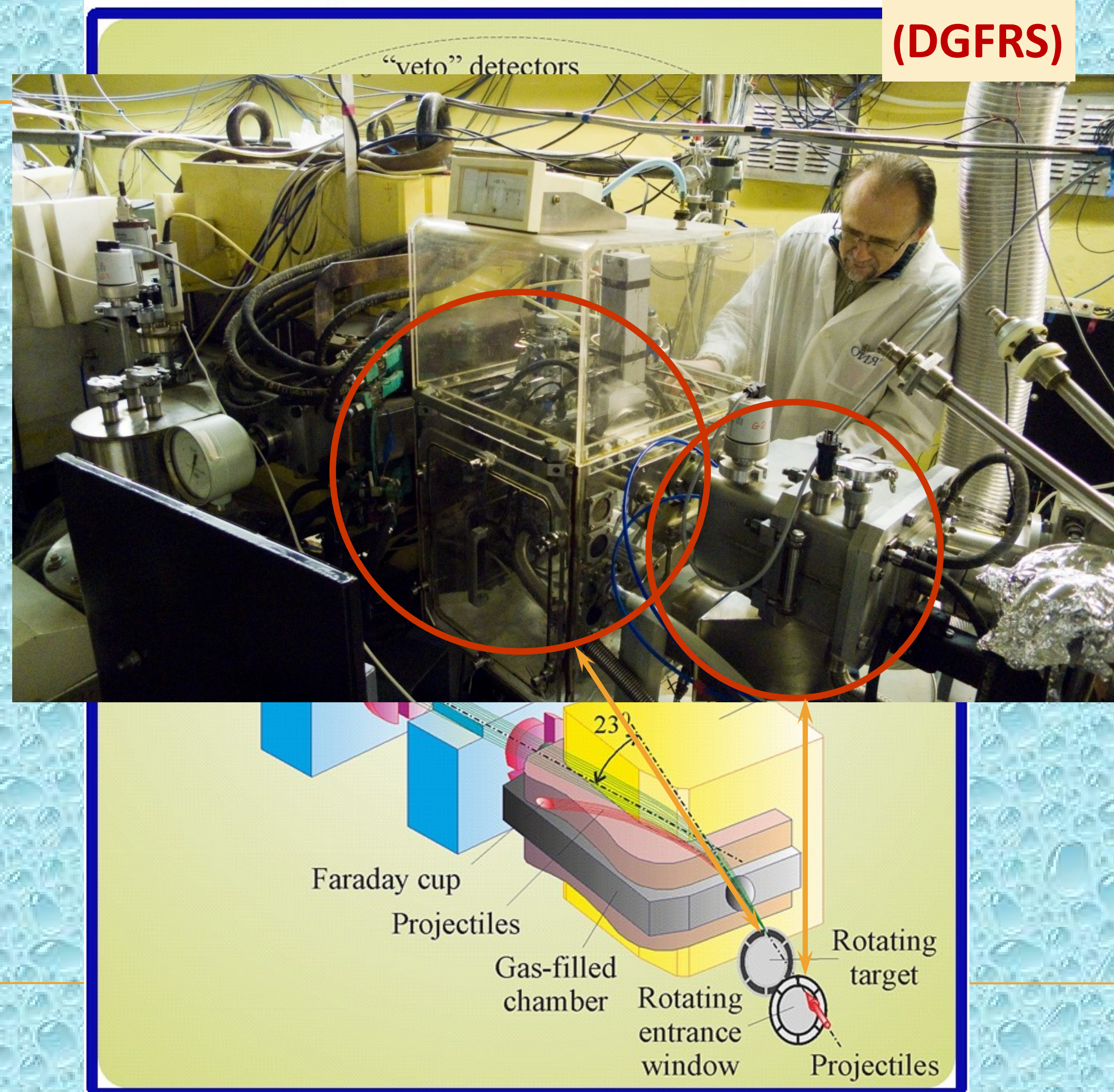


Synthesis of SHN with $Z>118$ implies the use of the heavier than ^{48}Ca beam particles (^{50}Ti, ^{54}Cr etc.). According to theoretical predictions, one may expect the production cross section ≤50 fb for SHN formed in fusion-evaporation reactions with these projectiles. It means that at least 10^{20} beam particles should be passed through the target waiting for decays of 2 atoms of these nuclei (one year experiment at 1 pμA beam intensity !?).

High-current cyclotron DC-280

G.G. Gulbekian *et al.*, Proc. 21st Intern. Conf. on Cyclotrons and their Applications, 2016

DC-280 (expected) E=4÷8 MeV/amu		
Ion	Ion energy (MeV/amu)	Output intensity (s^{-1})
^{7}Li	4	1×10^{14}
^{18}O	8	1×10^{14}
^{40}Ar	5	6×10^{13}
^{48}Ca	5	$0.6\text{-}1.2\times10^{14}$
^{54}Cr	5	2×10^{13}
^{58}Fe	5	1×10^{13}
^{124}Sn	5	2×10^{12}
^{136}Xe	5	1×10^{14}
^{238}U	7	5×10^{10}

Powers generated inside actinide oxide target P_{targ} and Ti target backing P_{back} at HI beam intensity I_{beam} and energy E_{lab}. The values correspond to the beam energy absorbed by the target of W_{targ} thickness and by the 0.71 mg/cm² Ti target backing. Reactions leading to SHN with $112 \le Z \le 120$ are considered.

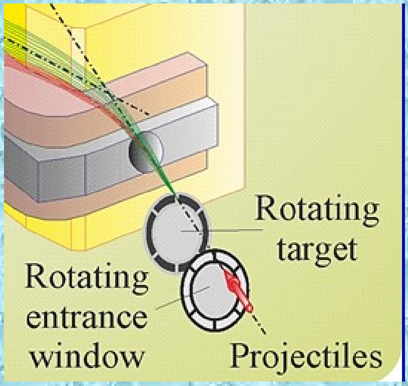
Reaction	I_{beam} (s^{-1})	E_{lab} (MeV)	W_{targ} (mg/cm²)	P_{targ} (W)	P_{back} (W)
$^{48}Ca+^{238}UO_2$	$7\cdot10^{13}$	235	0.4	65	189
$^{48}Ca+^{243}AmO_2$	$7\cdot10^{13}$	241	0.4	64	187
$^{54}Cr+^{238}U_3O_8$	$2\cdot10^{13}$	285	0.6	44	81
$^{58}Fe+^{244}PuO_2$	$1\cdot10^{13}$	311	0.7	29	45

- Heat load ↔ heat removal processes
- Beam dose load → sputtering of atoms

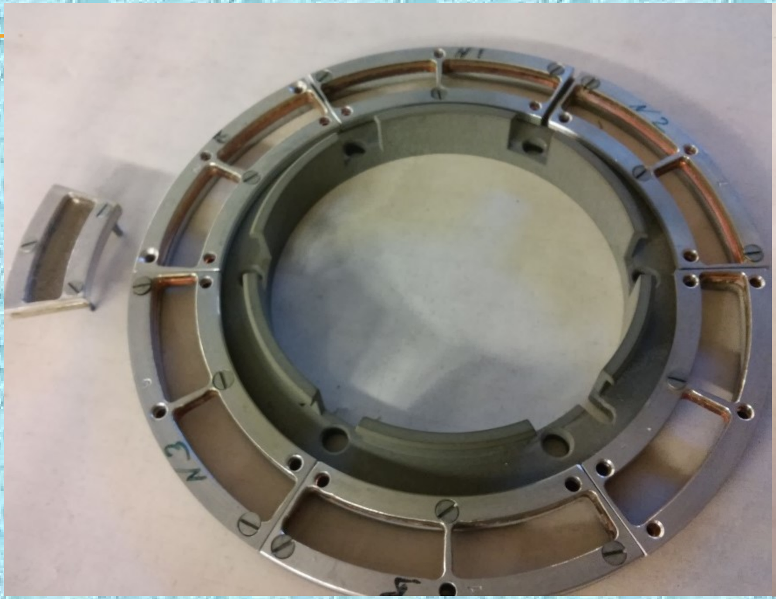
↓

Radiation damages

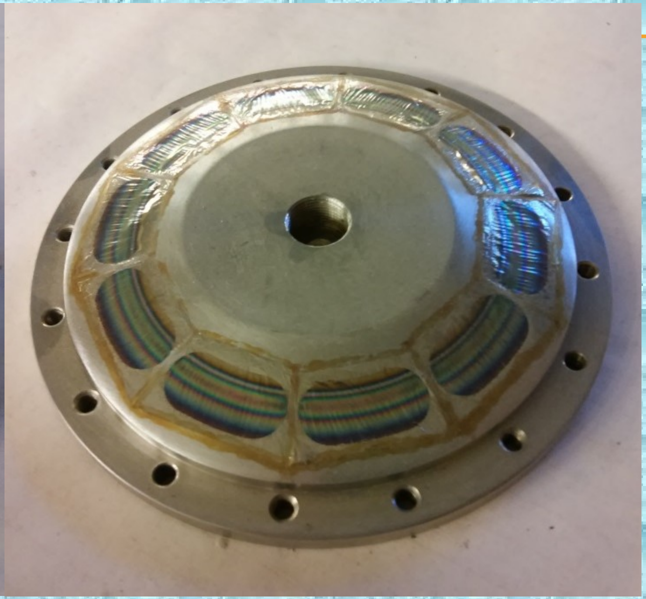
Rotating target and window is a way to reduce heat and dose loads

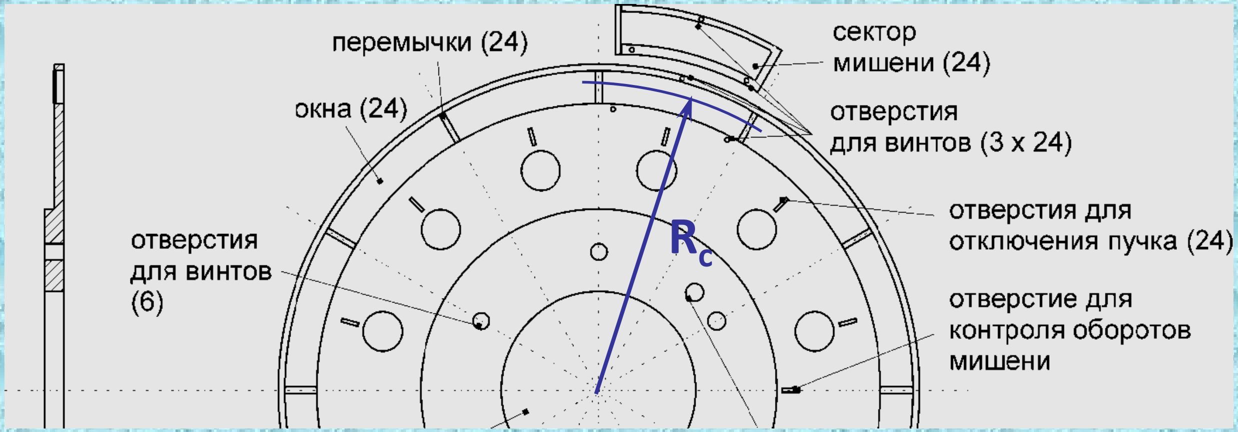


Target: R_c=60 mm, 6/12 cells. Reduction factor≈60 (d_b=8 mm)



Window: R_c=50 mm, 10 cells. Reduction factor≈50 (d_b=8 mm)



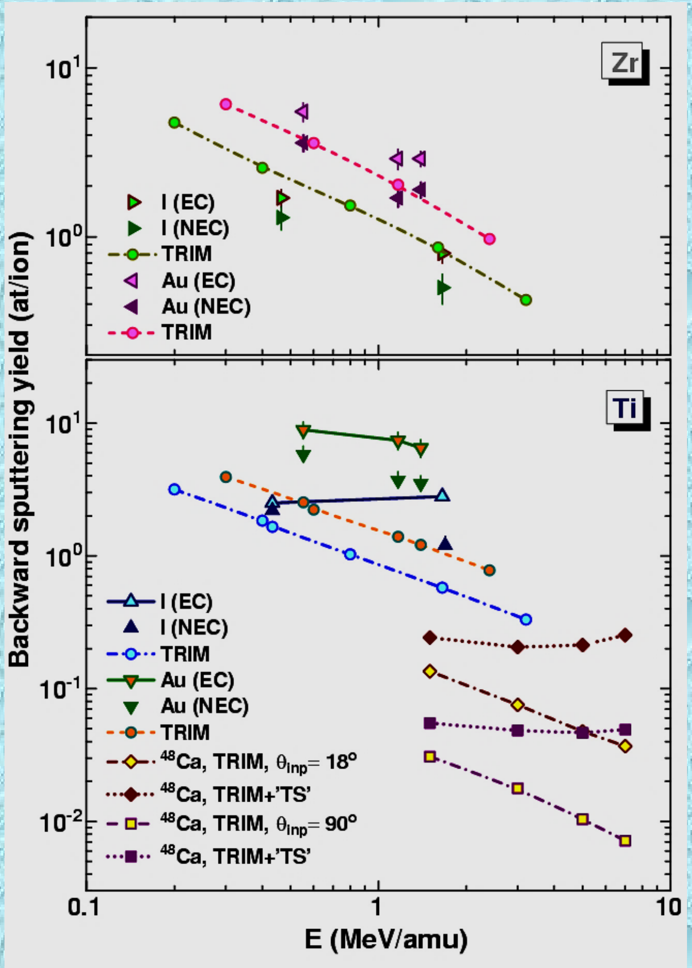


New large rotating target R_c=250 mm, 24 target cells. Reduction factor (RF) ≈ 250 (d_b=8 mm)

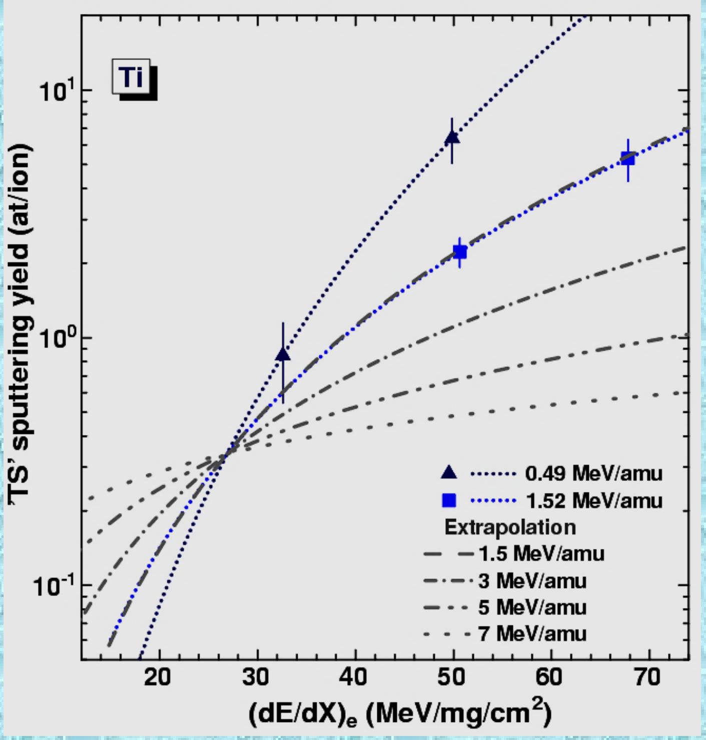
The observation of 2 events of SHN with Z=120 requires the beam dose of ~10^{20} particles (σ=50 fb, W_{targ}=0.4 mg/cm^2 and ε_{eff}=40%). Such dose may lead to the total loss of a target material toward the end of a run with a stationary target, if $S\approx$0.01 atom/ion (TRIM simulations). For rotating devices, the total yield of sputtered atoms is reduced by the corresponding RF. The question arises of the S values for actinide oxide targets and Ti target backing (window).

SPUTTERING YIELDS of Ti ATOMS of TARGET BACKING and WINDOW FOILS

H.D. Mieskes, et al, Phys. Rev. B 67 (2003) 155414



The reason for the discrepancy is an additional so-called "thermal spike ('TS') component" inherent in sputtering of such metals as Ti. It can be extracted from measured sputtering yields by the subtraction of those obtained with TRIM simulations. The 'TS' sputtering yields corresponding to the same velocity are usually fitted with the $a[(dE/dX)_e]^b$ function. Using HI $(dE/dX)_e$ at specified energies, 'TS' sputtering yields could be estimated.



SPUTTERING YIELDS of HEAVY ATOMS from UO_2 and Eu_2O_3 TARGETS

Backward sputtering yields of heavy atoms as a function of the beam energy for different HIs (full symbols) interacting with UO_2 [a,b] and Eu_2O_3 [c] targets.

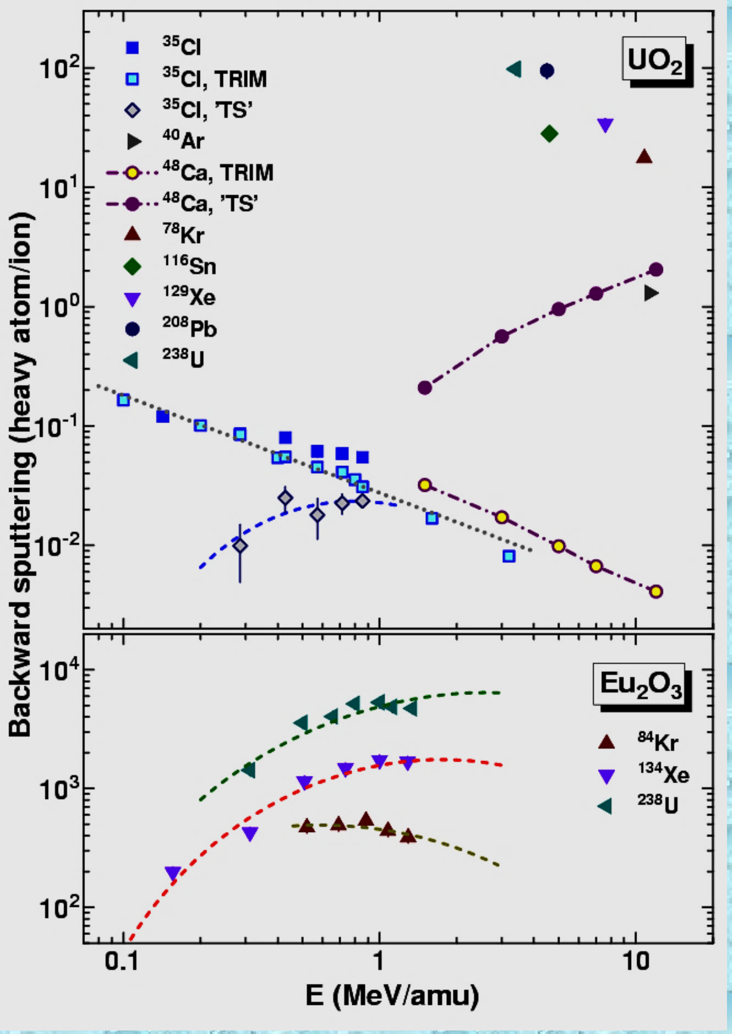


Figure gives

- the results of the data fits obtained with $S \sim [(dE/dX)_e]^4 =$ $= \{A[Q_{eq}(E)]^2\, ln(BE)/E\}^4$ [a] (dashed lines);
- the yields of U atoms sputtered by ^{35}Cl and ^{48}Ca ions from the UO_2 target as obtained with TRIM simulations [d];
- the smoothed-out approximation to the TRIM simulated data (the dotted line);
- the yields of U atoms under Cl ions, which are associated with thermal spike ('TS') effects (open diamonds);
- the yields of U atoms sputtered by ^{48}Ca ions as obtained with the approximations associated with 'TS' effects (full circles connected by the dash-dotted line).

[a] C.K. Meins, J.E. Griffith, Y. Qiu, M.H. Mendenhall, L.E. Seiberling, T.A. Tombrello, Radiat. Eff. Def. 71 (1983) 13.

[b] S. Schlutig, Ph.D. thesis, Univ. Caen, URL: http://tel.archives-ouvertes.fr/tel-00002110, 2001.

[c] W. Guthier, Springer Proc. Phys. 9 (1986) 17.

[d] J.F. Ziegler, J.P. Ziegler, J.P. Biersack, Code SRIM-2013, http://www.srim.org.

SPUTTERING YIELDS of HEAVY ATOMS from UF_4, UO_2 and Eu_2O_3

Backward sputtering yields of heavy atoms as a function of the electronic energy losses $(dE/dX)_e$ for different HIs interacting with UF_4 [a], UO_2 [b,c] and Eu_2O_3 [d] targets at definite velocities (symbols).

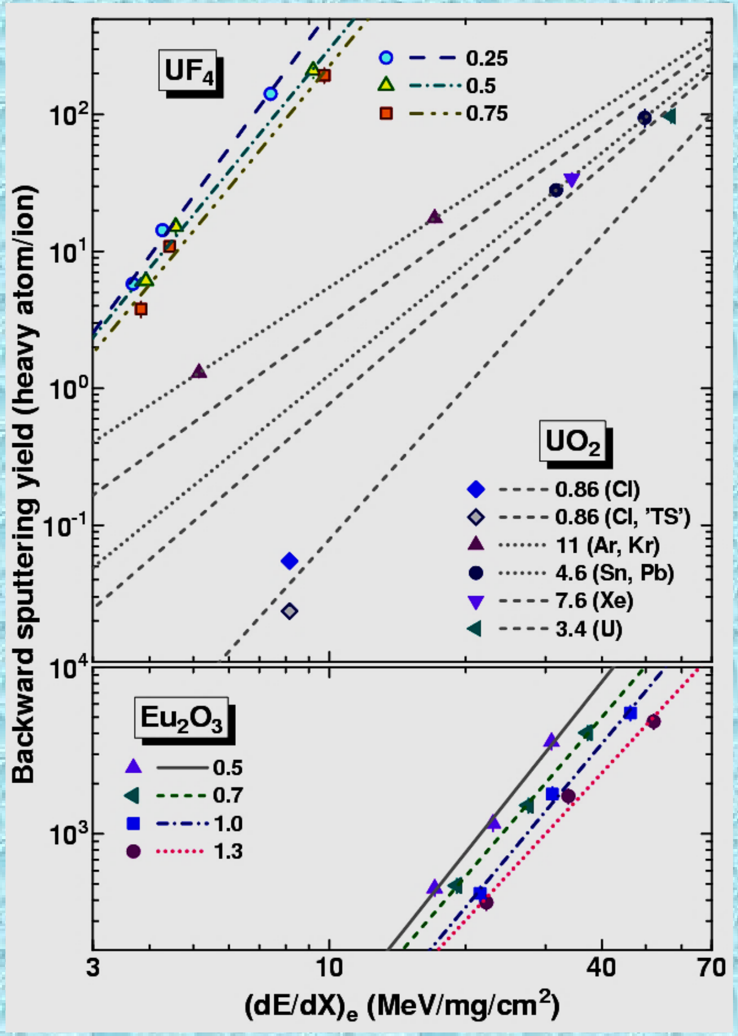


Figure gives

- the results of the $a[(dE/dX)_e]^b$ function fits to the data corresponding to the indicated different values of MeV/amu (different lines for UF_4 and Eu_2O_3, and dotted lines for UO_2);
- the results of the interpolation (extrapolation) of the U sputtering yields for UO_2 at energies corresponding to those used in the irradiations by Cl, Xe and U ions (dashed lines).

[a] J.E. Griffith, R.A. Weller, L.E. Seiberling, T.A. Tombrello, Radiat. Eff. 51 (1980) 223.

[b] C.K. Meins, J.E. Griffith, Y. Qiu, M.H. Mendenhall, L.E. Seiberling, T.A. Tombrello, Radiat. Eff. Def. 71 (1983) 13.

[c] S. Schlutig, Ph.D. thesis, Univ. Caen, URL: http://tel.archives-ouvertes.fr/tel-00002110, 2001.

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ESTIMATES and OBSERVATIONS regarding to the DGFRS experiments (Ti window foils)

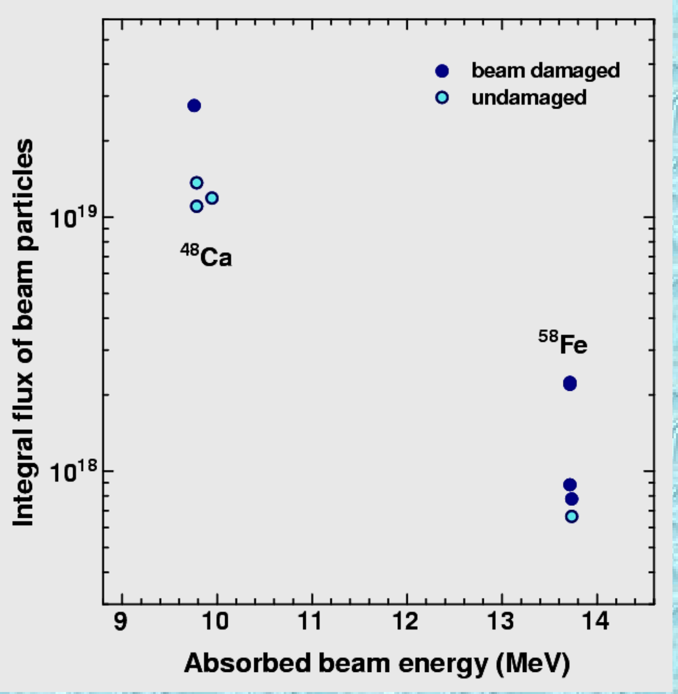
The expected thinning for the Ti foils δ_{TH} (in percent) according to the estimates of the backward sputtering yields against the beam doses of the ^{48}Ca and ^{58}Fe ions passed through the sets #1 – #10 of foils used in the experiments in 2005–2009. Average beam intensities $\langle I \rangle$ (in 10^{12} particle/s), foil thicknesses $\langle W_{Ti} \rangle$ (in 10^{18} atom/cm^2), beam power density $\langle P_{Ti} \rangle$ (in W/cm^2) released in foils at the intensity $\langle I \rangle$, beam radius r_b=0.4 cm and beam electronic energy losses in Ti $(dE/dX)_e$ [SRIM] (in MeV/mg/cm^2), and the backward sputtering yields S_b (in atom/ion) estimated according to the extrapolations are listed for the reference.

Set	beam	beam dose	$\langle I \rangle$	$\langle W_{Ti} \rangle$	$(dE/dX)_e$	$\langle P_{Ti} \rangle$	S_b	δ_{TH}
#1[a]	^{48}Ca	1.18×10^{19}	6.05	9.15	13.3	18.6	0.180	1.85
#2[b]		2.75×10^{19}	5.38	9.21	13.3	16.7	0.179	4.25
#3[b]	^{58}Fe	2.20×10^{18}	4.18	9.11	18.9	18.3	0.267	0.513
#4[b]		8.84×10^{17}	4.77			20.8		0.206
#5[b]		2.24×10^{18}	4.22			18.4		0.521
#6[b]		7.78×10^{17}	3.86	9.12		16.9		0.181
#7[a]		6.64×10^{17}	2.72			11.9		0.155
#8[a]	^{48}Ca	1.10×10^{19}	3.65	9.17	13.4	11.4	0.177	1.69
#9[a]		1.37×10^{19}	4.37			13.6		2.10
#10[a]		1.19×10^{19}	4.00		13.6	12.7	0.172	1.78

[a] Foils were not damaged during the irradiation.

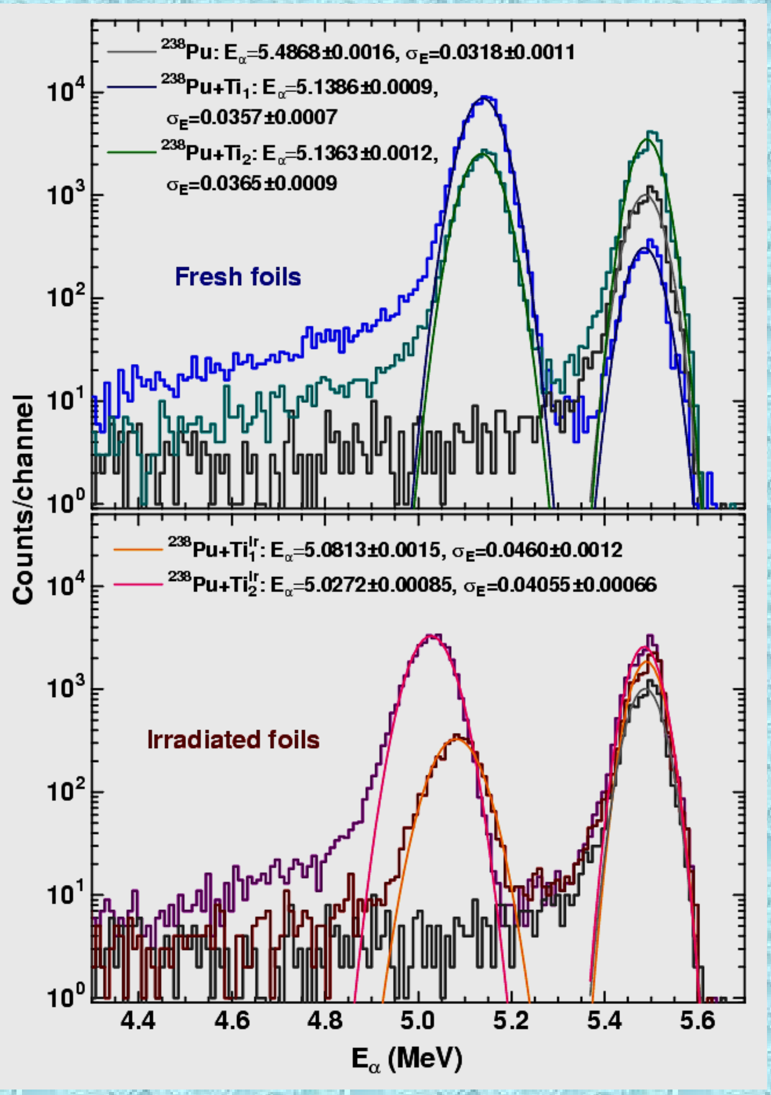
[b] One or several foils were damaged under the beam.

The ability of Ti foils to withstand a beam dose load



The beam doses of ^{48}Ca and ^{58}Fe ions that the Ti window foils were able to withstand in the DGFRS experiments as a function of the beam energy absorbed by the foils.

OBSERVATIONS in the DGFRS EXPERIMENTS
(energy absorption of α-particles in Ti WINDOW FOILS)



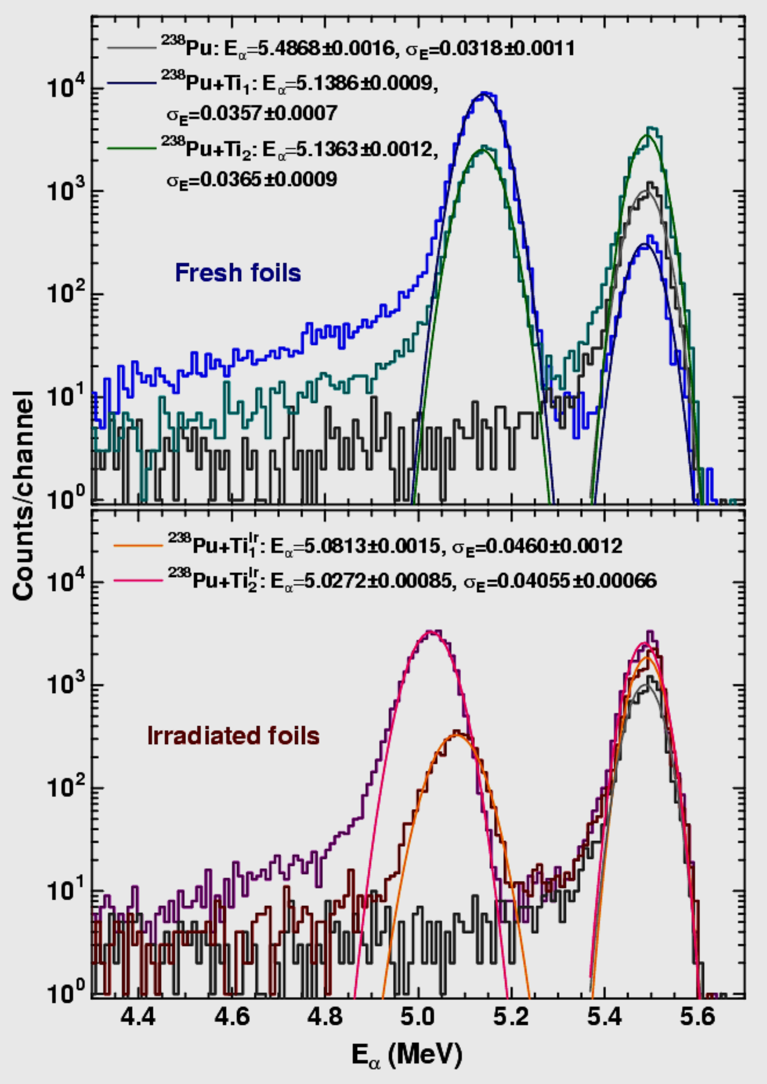
Thicknesses *W* of Ti window foils (in mg/cm²) as obtained with the measurements of the absorption energy for the ^{238}Pu *α*-particles: W_1 and W_2 for the fresh foils, and $W^{ir}_{\#1}$ and $W^{ir}_{\#2}$ for the irradiated foils (sets #1 and #2). $\langle E_{inp}\rangle$ is the input energy obtained in the direct measurements with the source (in MeV).

D	$\langle E_{\rm inp}\rangle$	W_1	W_2	$W^{\rm ir}_{\#1}$	$W^{\rm ir}_{\#2}$
2	$5.4844_{0.0020}$	$0.7187_{0.0062}$	$0.7389_{0.0061}$	$0.7712_{0.0062}$	$0.8108_{0.0062}$
3	$5.4852_{0.0017}$	$0.7165_{0.0054}$			
	$5.4904_{0.0019}$		$0.7321_{0.0064}$		
	$5.4895_{0.0021}$			$0.8411_{0.0074}$	
	$5.4846_{0.0017}$				$0.9390_{0.0044}$
weighting		$0.7268^{a}_{0.0053}$	$0.732^{b}_{0.008}$		

[a] The mean value resulting from weighting of a great number of large-size pieces of Ti foils, from which 5 pieces were selected for the Set #1 (see Table 2).

[b] The mean value resulting from weighting of 5 pieces of Ti foils selected for the subsequent irradiations (Set #2 in Table 2)

OBSERVATIONS in the DGFRS EXPERIMENTS (energy absorption of α-particles in Ti WINDOW FOILS)



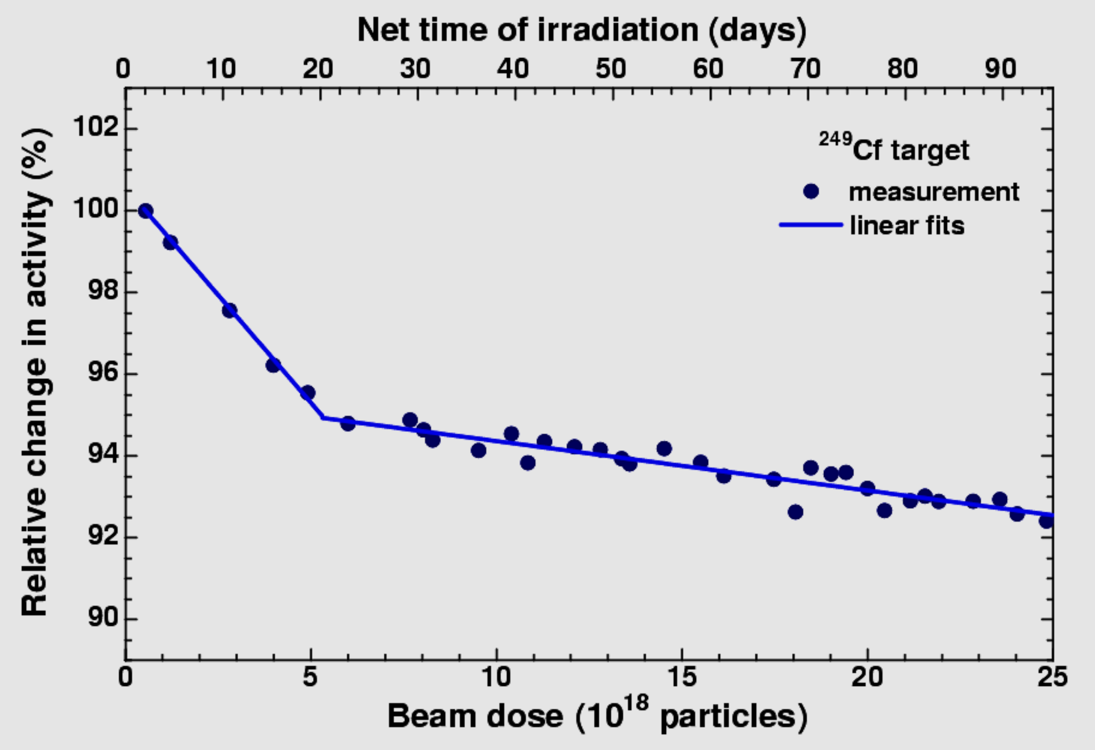
A thickening of the Ti window foils due to TiH_2 formation as deduced from the experiments with the ^{238}Pu *α*-source. $\langle E_{inp} \rangle$ and $\langle E_{out} \rangle$ are the measured input and output *α*-particle energies (in MeV). $\Delta W(TiH_2)$ is the TiH_2 thickness obtained with $\langle E_{inp} \rangle$ and $\langle E_{out} \rangle$ and the application of the range-energy curve for α-particles in TiH_2 [SRIM] (in mg/cm2). $\delta W(TiH_2)$ is the percentage of Ti atoms corresponding to $\Delta W(TiH_2)$, as obtained by comparison to thickness values obtained by weighting.

D	Set	$\langle E_{\rm inp} \rangle$	Set	$\langle E_{\rm out} \rangle$	$\Delta W(\rm TiH_2)$	$\delta W(\rm TiH_2)$
2	1	$5.1367_{0.0010}$	#1	$5.1107_{0.0010}$	$0.0460_{0.0035}$	$6.1_{0.5}$
	2	$5.1267_{0.0009}$	#2	$5.0910_{0.0010}$	$0.0630_{0.0034}$	$8.3_{0.5}$
3	1	$5.1386_{0.0009}$	#1	$5.0813_{0.0015}$	$0.1012_{0.0042}$	$13.4_{0.6}$
	2	$5.1363_{0.0012}$	#2	$5.0272_{0.0009}$	$0.1920_{0.0036}$	$25.2_{0.6}$

OBSERVATIONS in the DGFRS EXPERIMENTS
(α-activity measurement of the ^{249}Cf TARGET during irradiation)

The sputtering yield of U atoms from UO_2 is estimated as ~1 atom/ion for the 5 MeV/amu ^{48}Ca beam. This yield is quite detectable in our experiments on synthesis of SHN.

In these experiments we systematically monitored the integrity of radioactive targets by counting their α-activities. The results of such monitoring for the ^{249}Cf target ($T_{1/2}$=351 y):

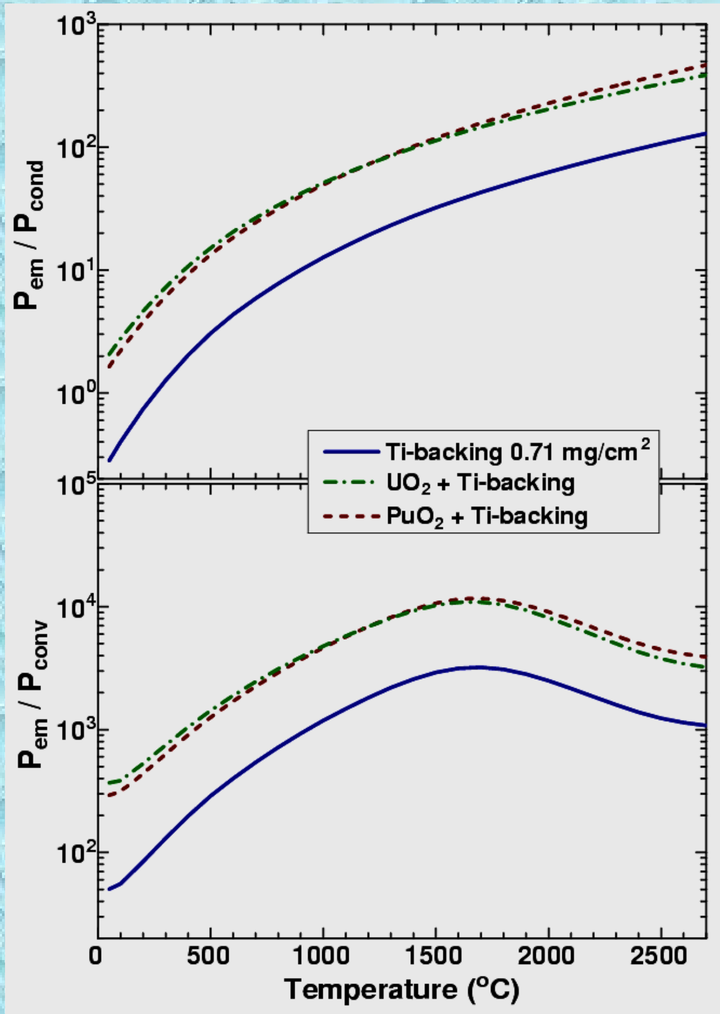


At the beginning of the irradiation, the sputtering of Cf atoms corresponds to S_{tr}=(0.187±0.003) atom/ion, then it reduces to S_{tr}=(0.021±0.002) atom/ion. For a total beam dose accumulated during the experiment, the reduced sputtering yield is estimated as 6.73×10^{-4} atom/ion/cm^2.

In a future experiment on synthesis of SHN with Z=120 in the ^{50}Ti+^{249}Cf reaction, the beam dose of (1–2)×10^{20} particles may allow us to observe few events of its decay. This dose should lead to the loss of (7–14)% of the 0.4 mg/cm^2 target toward the end of the experiment (assuming the same sputtering yield for ^{50}Ti, as obtained for ^{48}Ca beam).

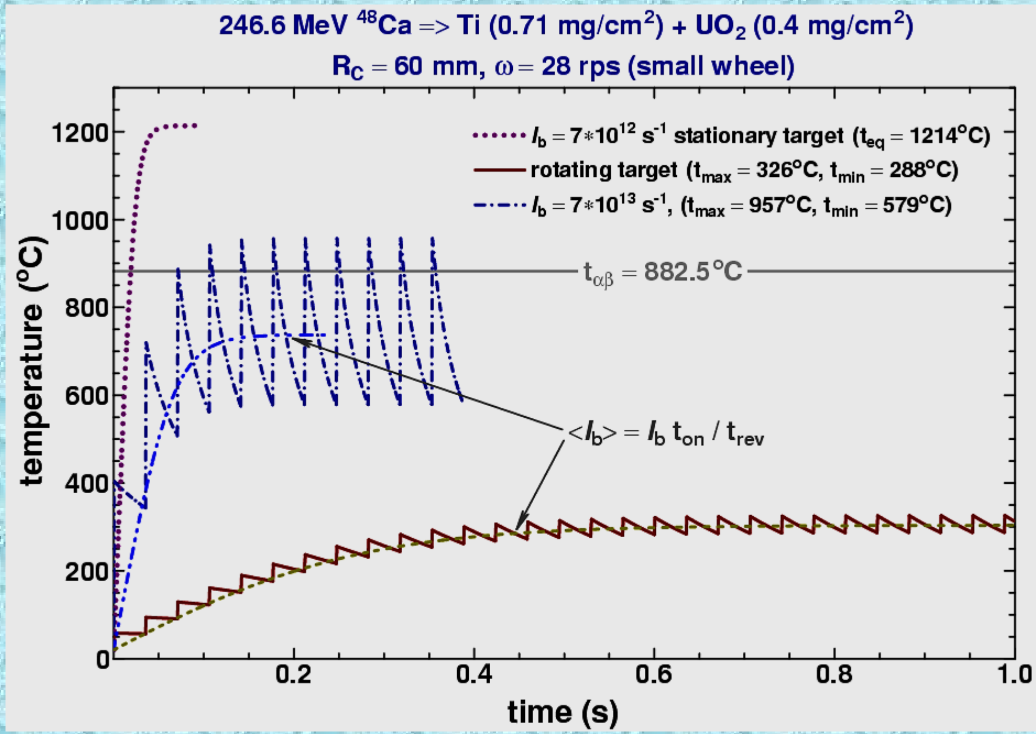
TEMPERATURE of the ROTATING TARGET

According to appropriate estimates, heat removal due to the radiation emitted from the surfaces of the target and target backing foils becomes the main process of their cooling at high temperatures.



With the assumption on the absence of the temperature difference between the target backing and target itself, the differential equation taking into account a radiation heat exchange only [J.L. Yntema & F. Nickel, *Lect. Notes Phys*, 83, 206 (1978)] can be rewritten as :

$$\Delta E \cdot I_b(t)/F + (\varepsilon_t+\varepsilon_b)\cdot\sigma_{SB}\cdot T_0^4 = \\ =(\varepsilon_t+\varepsilon_b)\cdot\sigma_{SB}\cdot T^4(t) + (c_t\cdot\rho_t\cdot d_t + c_b\cdot\rho_b\cdot d_b)\cdot dT(t)/dt$$



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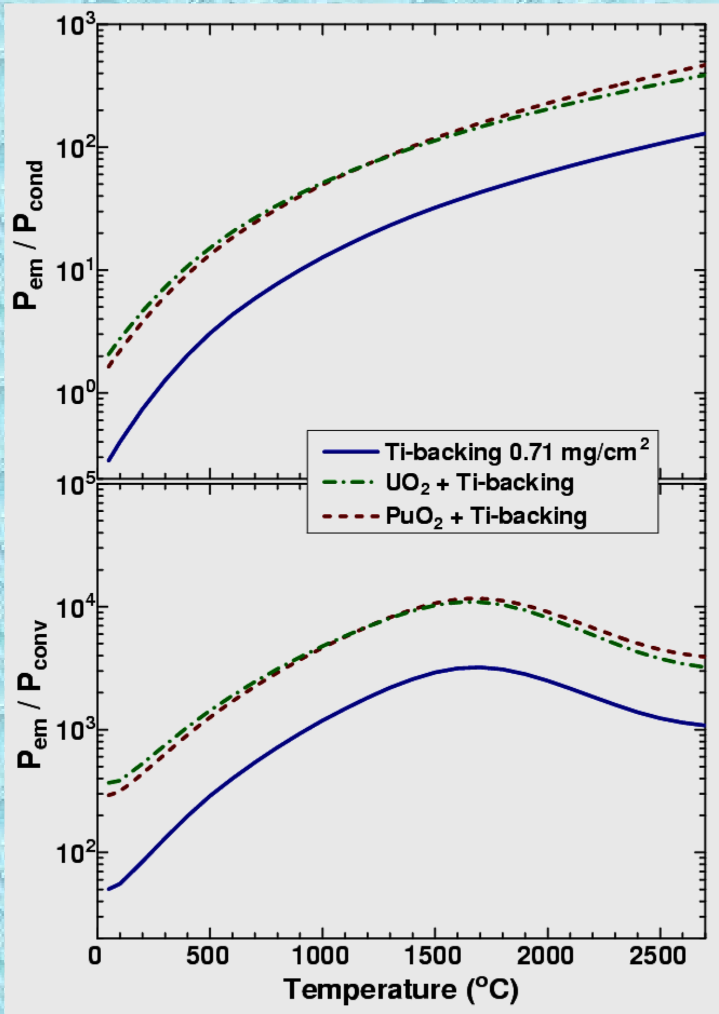

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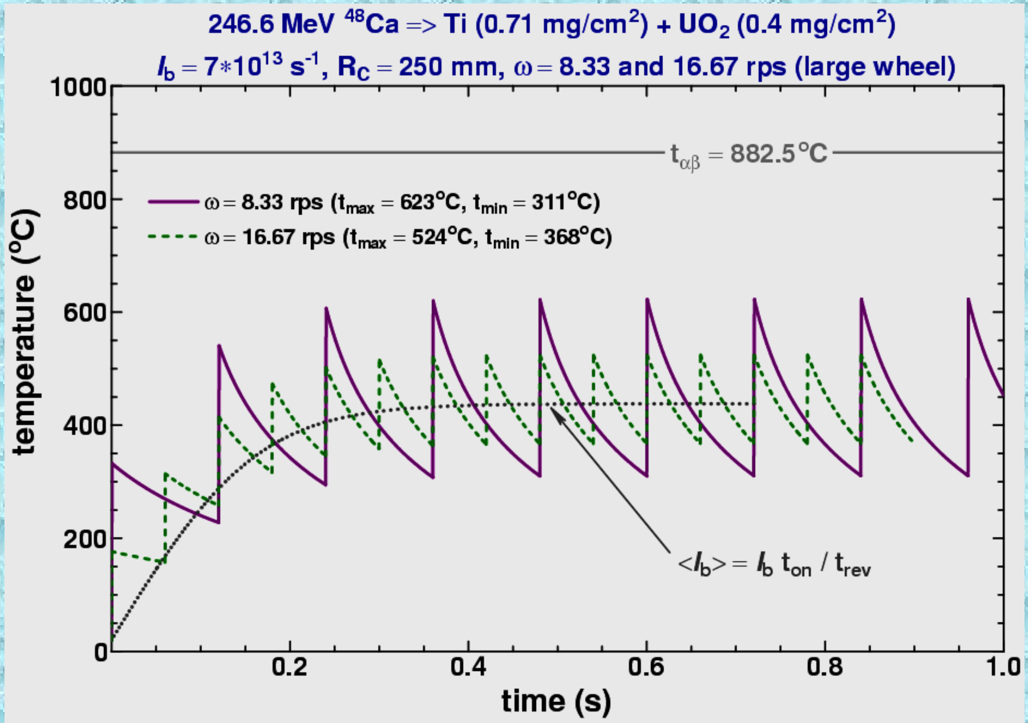

SUMMARY

Sputtering of the actinide targets and Ti window foils irradiated by intense heavy ion beams has been considered in the context of experiments on synthesis and studies of superheavy nuclei, which are carried out in Dubna with a gas-filled recoil separator (DGFRS) and will be continued with higher beam intensities. Targets and foils are loaded by the beam dose of $\sim 10^{20}$ particles and intensity of $\sim 7\times10^{13}$ s^{-1} in these experiments. Durability of targets and window foils become questionable in such conditions.

- Available data on sputtering yields for Ti and U atoms obtained at the same ion velocity and plotted as a function of the electronic energy losses of beam particles were approximated with relationship $a[(dE/dX)_e]^b$. With these approximations, the Ti and U sputtering yields for the 5 MeV/amu ^{48}Ca beam were estimated as ~0.05 and ~1 atom/ion, respectively. These values are ~5 times and ~2 orders of magnitude as high as those obtained with TRIM simulations.

- Attempts were made to trace sputtering of Ti atoms from window foils used in the DGFRS experiments with a high beam dose collection. The examination of these foils with the α-particle energy loss measurements revealed their apparent thickening (instead of thinning), as compared to the fresh foils. Such “thickening” could be explained by an increase in energy losses of α-particles due to the adsorption of hydrogen by the foil faced to the DGFRS volume.

- For the Cf target irradiated by the 245 MeV ^{48}Ca beam, the transmission sputtering yield of 6.73×10^{-4} atom/ion/cm^2 was obtained with the measurements of its α-activity during a long-term experiment. This value is ~50 times as low as the yield obtained by the approximation of the UO_2 data, but it is only ~2 times as high as the value obtained with TRIM simulations.

- The temperature of the rotating target and target backing as a whole was estimated as a function of time in the conditions of their pulse heating by the beam and radiative cooling of their surfaces. This way of heat removal might be effective at temperatures of several hundreds of Celsius degrees which are reached in ^{48}Ca beam irradiations at the intensity of $\sim 7\times10^{13}$ s^{-1}, as implied by these estimates.


THANK YOU
FOR YOUR
ATTENTION!
Flerov Laboratory of Nuclear Reactions , JINR, Dubna