HZDR HELMHOLTZ ZENTRUM DRESDEN ROSSENDORF

ACTIVATION OF THE CONSTRUCTING MATERIALS OF ACCELERATORS

10th – 12th October 2016

TECHNISCHE UNIVERSITÄT DARMSTADT

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GSI

ACTIVATION **EXPERIMENTS**

FAIR @ GSI

Irradiation > ^{238}U [200 MeV/u and 125 MeV/u],
^{124}Xe [300 MeV/u]

Experimental data for code validating:
> depth profiles of the residual activity
> fragmentation
> stopping range

> transition between
BME and RQMD

BEAM LOSSES **CRITERIA**

"SATIF–12* promised work"

> comparison of FLUKA2008 and FLUKA2011

> validation of the "Generic curve" approach for lower energies

> beam losses criteria for long-term irradiation ~ 20 years

* P. Katrík, E. Mustafin, D.H.H. Hoffmann, I. Strašík, M. Pavlovic - Proceedings of the 12th Shielding Aspects of Accelerators, Targets and Irradiation Facilities SATIF 12 (2014) 124-134

photo > www.gsi.de

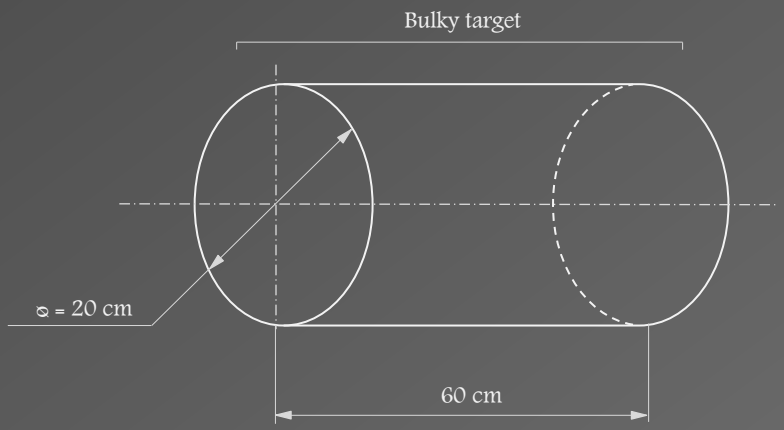
WORKING GROUP REPORT

BEAM LOSS

"To allow hands-on maintenance of accelerator components without unreasonable constraints, dose levels should be below 1 mSv/h (measured 30 cm from component surface). This corresponds to beam loss of about 1 W/m along the beam enclosure; this figure is roughly independent of beam energy, for energies above 100 MeV. Thus a greater number of particles can be lost at lower energies, as activation is less efficient. Note: the beam loss goal for the APT linac design was lower, 0.1 W/m above 100 MeV, to provide adequate margin for meeting the high availability requirement.*"

* N.V. Mokhov, W. Chou: "Proceedings of the 7th ICFA Mini-Workshop on High-Brightness Hadron Beams", FermiLAB, (2000) 51-61

SIMULATION **PARAMETERS**



target materials: stainless steel, cooper
target geometry: bulky target

beam: ^{1}H, ^{4}He, ^{12}C, ^{20}Ne, ^{40}Ar, ^{84}Kr, ^{132}Xe, ^{197}Au, ^{238}U
beam energies: 1GeV/u ÷ 200 MeV/u ÷ 25 MeV/u

normalized induced activities > beam power of 1 W

3 months of irradiation
several time points after irradiation

> 20 years irradiation + 20 years cooling time. (NEW)

	FLUKA 2008.3.6*	FLUKA 2011.2b.5**
	Tolerable beam losses for Uranium beam [W/m]	
1GeV/u	5	5
500 Mev/u	12	12
200 MeV/u	60	40
150 MeV/u		52

* I.STRASIK et al.: Residual activity induced by heavy ions and beam-loss criteria for heavy-ion accelerators, Phys. Rev. ST Accel. Beams 13,(2010)
** P. Katrík, et al. - Proceedings of the 12th Shielding Aspects of Accelerators, Targets and Irradiation Facilities SATIF 12 (2014) 124-134

GENERIC **CURVE**

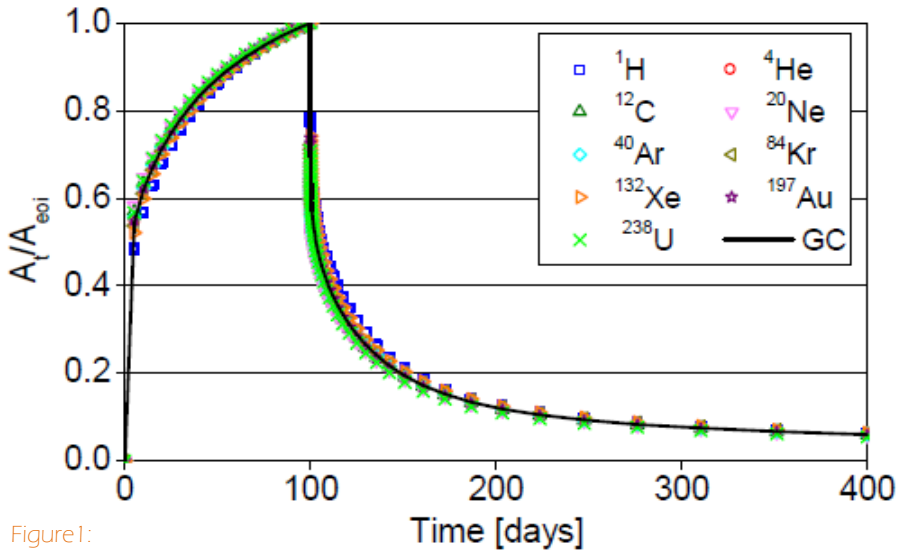


Figure1:

> time evolution of the activity
> independent of the projectile mass

> activity normalized to the activity at the end of the irradiation

> obtained by averaging the data points of the individual curves corresponding to different primary ions

Figure1: Time evolution of the relative activity in the Beam pipe by different projectiles at 500 MeV/u, calculated by FLUKA2008.*

Figure2: Inventory of nuclides produced in the Beam pipe by different projectiles at 500 MeV/u, calculated by FLUKA2008.*

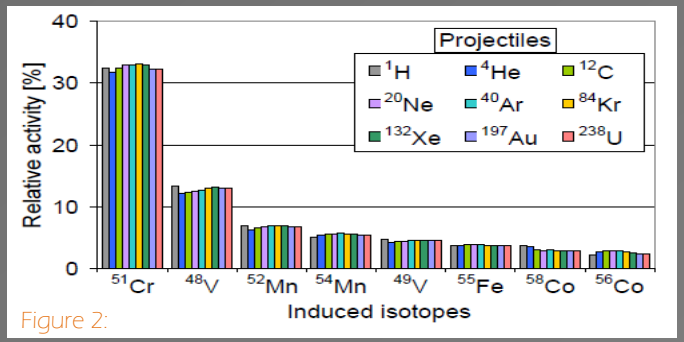


Figure 2:

* I.STRASIK et al.: Residual activity induced by heavy ions and beam-loss criteria for heavy-ion accelerators, Phys. Rev. ST Accel. Beams 13, (2010)

SCALING **LAW**

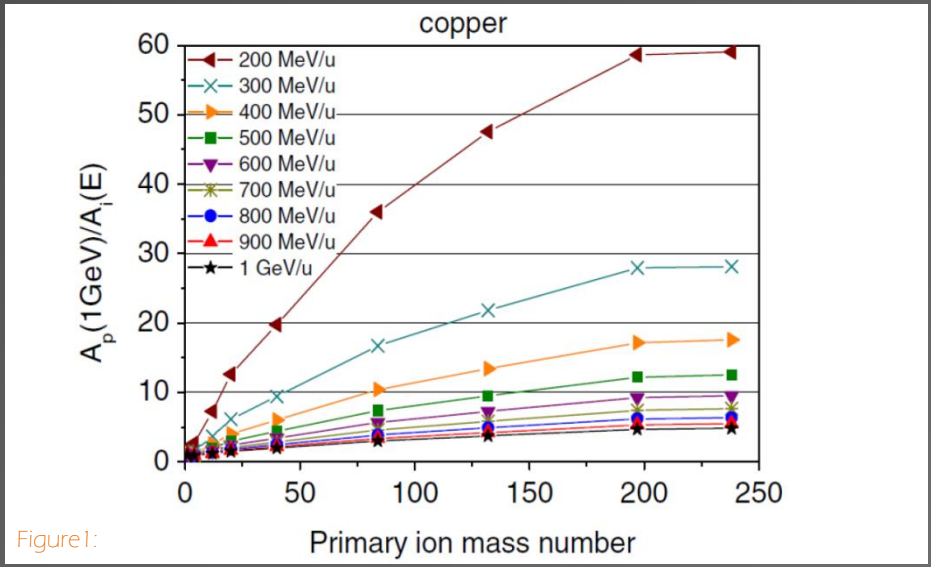


Figure1:

relative activities

> not strong dependence on the projectile species

> dependent on the target material (isotopes produced mostly by secondary particles)

the activity induced by 1 W/m of beam losses is decreasing

> with increasing ion mass

> with decreasing energy

Figure1: The scaling factor is represented by the ratio of the normalized activity induced by a 1 GeV proton beam, A_p (1GeV), to the normalized activity induced by the beam of interest at a given energy, A_i(E). Corresponding activities were calculated by FLUKA 2008.*

The activities used for the scaling law determination are calculated at the end of the irradiation.

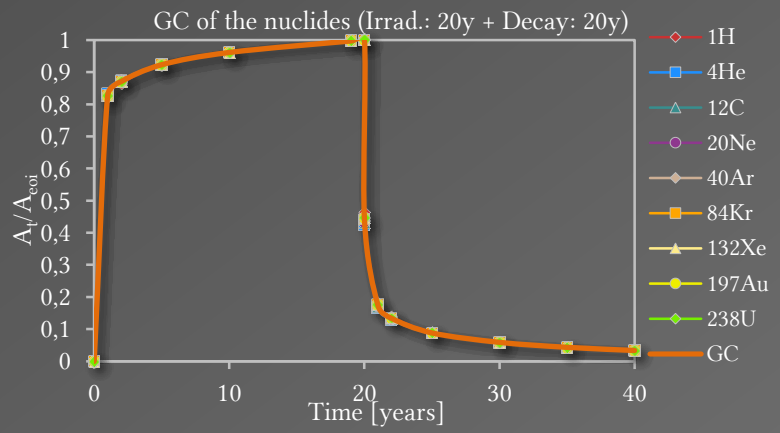
* I.STRASIK et al.: Residual activity induced by heavy ions and beam-loss criteria for heavy-ion accelerators, Phys. Rev. ST Accel. Beams 13, (2010)

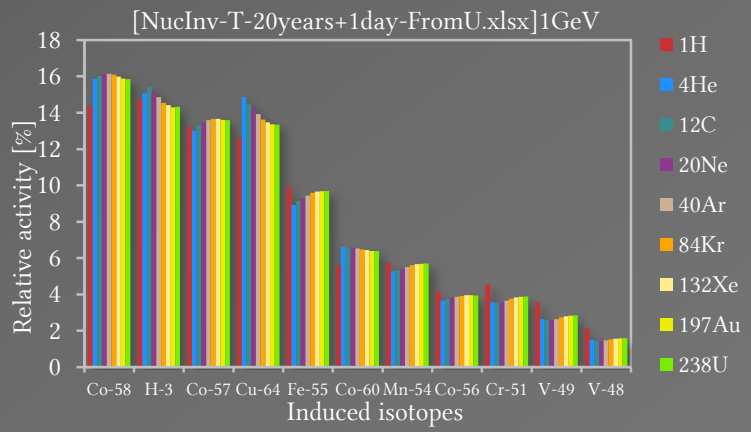
SIMULATION **PARAMETERS**

Irradiation time > 20 years

Beam energy > 1 GeV/u

Nuclide inventory > 1 day after irr.





Relative activities of nuclides produced by different beam species [%]

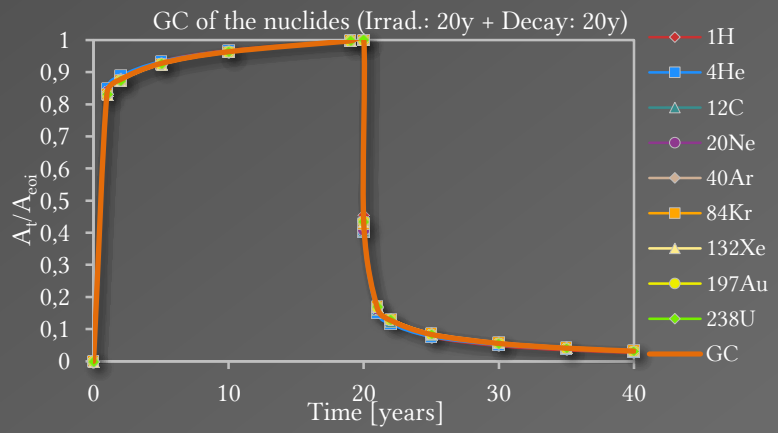
	^{1}H	^{4}He	^{12}C	^{20}Ne	^{40}Ar	^{84}Kr	^{132}Xe	^{197}Au	^{238}U
Co-58	14.38	15.86	16.02	16.14	16.14	16.09	15.99	15.89	15.85
H-3	14.72	15.07	15.42	15.17	14.86	14.55	14.43	14.30	14.33
Co-57	13.23	13.02	13.31	13.47	13.59	13.67	13.66	13.61	13.60
Cu-64	12.60	14.87	14.46	14.25	13.93	13.63	13.48	13.37	13.35
Fe-55	9.93	8.93	9.14	9.27	9.44	9.60	9.67	9.68	9.69
Co-60	5.59	6.62	6.57	6.56	6.53	6.48	6.44	6.39	6.39
Mn-54	5.74	5.28	5.34	5.40	5.51	5.62	5.67	5.68	5.71
Co-56	4.17	3.65	3.76	3.82	3.87	3.93	3.95	3.95	3.95
Cr-51	4.55	3.57	3.54	3.57	3.66	3.77	3.84	3.87	3.89
V-49	3.58	2.65	2.57	2.59	2.65	2.74	2.80	2.83	2.85
V-48	2.13	1.51	1.44	1.44	1.48	1.53	1.57	1.58	1.60

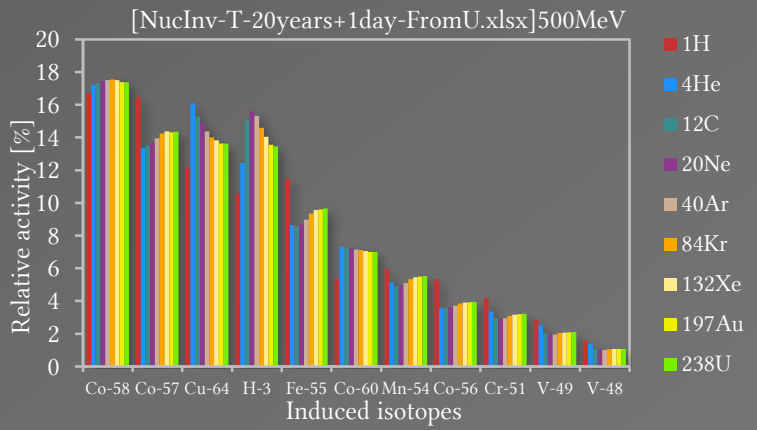
SIMULATION **PARAMETERS**

Irradiation time > 20 years

Beam energy > 500 MeV/u

Nuclide inventory > 1 day after irr.





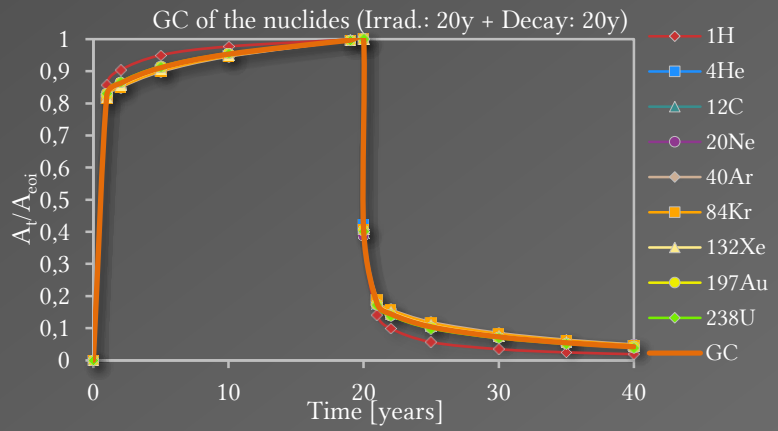
Relative activities of nuclides produced by different beam species [%]									
	^{1}H	^{4}He	^{12}C	^{20}Ne	^{40}Ar	^{84}Kr	^{132}Xe	^{197}Au	^{238}U
Co-58	16.72	17.21	17.34	17.43	17.51	17.54	17.52	17.39	17.37
Co-57	16.47	13.37	13.52	13.68	13.94	14.25	14.37	14.33	14.37
Cu-64	12.16	16.10	15.28	14.87	14.39	14.02	13.84	13.64	13.63
H-3	10.58	12.44	15.05	15.59	15.32	14.60	14.05	13.57	13.45
Fe-55	11.48	8.64	8.56	8.68	8.97	9.35	9.56	9.61	9.67
Co-60	5.39	7.32	7.25	7.20	7.16	7.12	7.07	7.00	7.00
Mn-54	5.96	5.13	4.92	4.93	5.10	5.34	5.46	5.49	5.54
Co-56	5.33	3.56	3.58	3.63	3.72	3.86	3.92	3.93	3.95
Cr-51	4.17	3.36	2.94	2.90	2.95	3.09	3.17	3.20	3.23
V-49	2.85	2.52	2.04	1.97	1.96	2.04	2.08	2.10	2.12
V-48	1.57	1.41	1.08	1.04	1.02	1.05	1.07	1.08	1.08

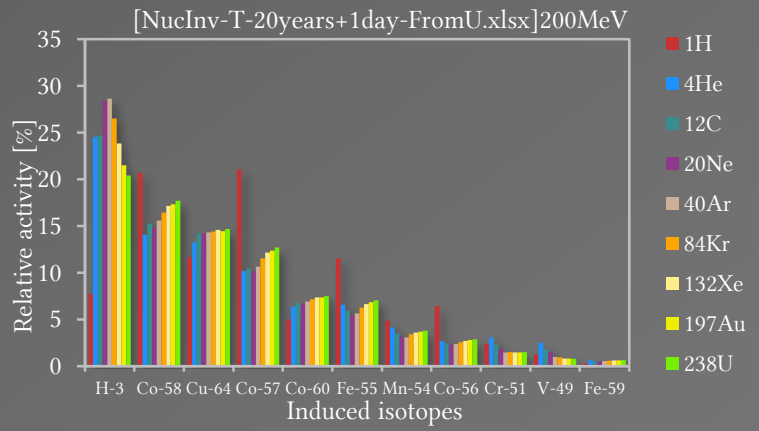
SIMULATION **PARAMETERS**

Irradiation time > 20 years

Beam energy > 200 MeV/u

Nuclide inventory > 1 day after irr.





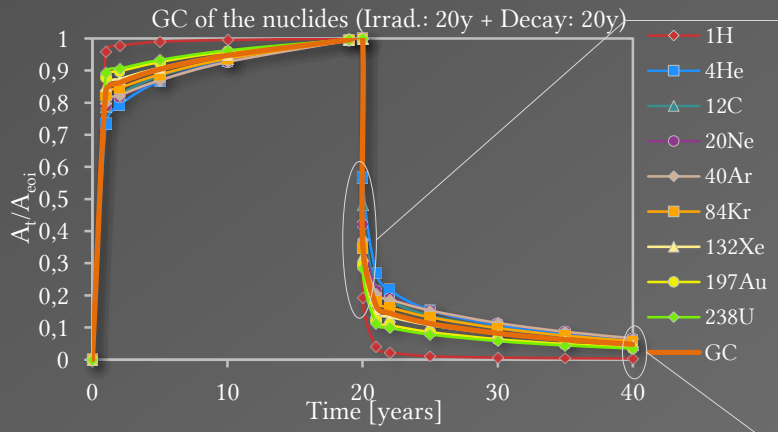
Relative activities of nuclides produced by different beam species [%]									
	^{1}H	^{4}He	^{12}C	^{20}Ne	^{40}Ar	^{84}Kr	^{132}Xe	^{197}Au	^{238}U
H-3	7.74	24.56	24.67	28.35	28.62	26.50	23.82	21.51	20.41
Co-58	20.69	14.08	15.23	14.87	15.58	16.44	17.14	17.33	17.70
Cu-64	11.62	13.25	14.16	14.17	14.31	14.41	14.59	14.45	14.67
Co-57	21.06	10.17	10.43	10.13	10.63	11.52	12.14	12.36	12.71
Co-60	4.99	6.41	6.74	6.72	6.93	7.16	7.37	7.36	7.50
Fe-55	11.55	6.55	5.96	5.43	5.63	6.25	6.63	6.86	7.06
Mn-54	4.85	4.14	3.49	3.12	3.09	3.42	3.59	3.68	3.78
Co-56	6.44	2.67	2.48	2.29	2.34	2.56	2.71	2.79	2.87
Cr-51	2.38	3.09	2.29	1.84	1.46	1.49	1.45	1.46	1.49
V-49	1.21	2.50	1.79	1.45	0.99	0.94	0.83	0.80	0.80
Fe-59	0.31	0.61	0.54	0.51	0.54	0.59	0.60	0.61	0.63

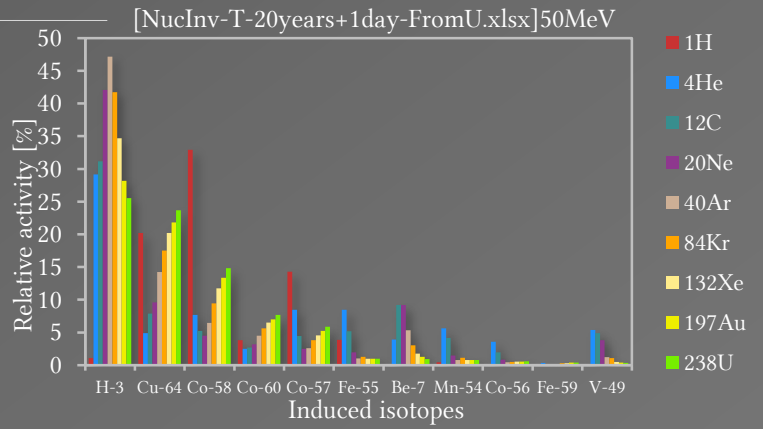
SIMULATION **PARAMETERS**

Irradiation time > 20 years

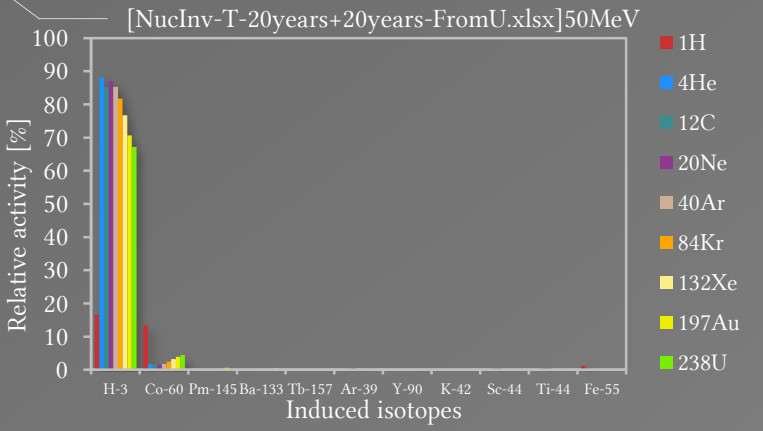
Beam energy > 50 MeV/u

Nuclide inventory > 1 day after irr.





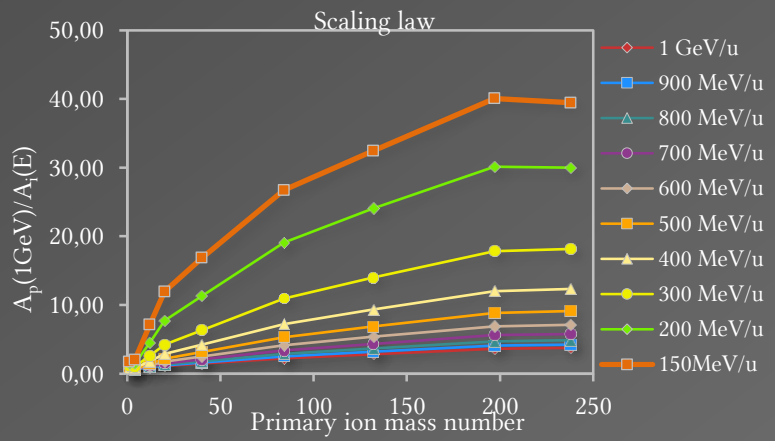
Nuclide inventory > 20 years after irr.



Relative activities of nuclides produced by different beam species [%]

	^{1}H	^{4}He	^{12}C	^{20}Ne	^{40}Ar	^{84}Kr	^{132}Xe	^{197}Au	^{238}U
H-3	1.06	29.15	31.16	42.09	47.14	41.74	34.66	28.18	25.52
Cu-64	20.20	4.90	7.85	9.57	14.24	17.50	20.22	21.80	23.65
Co-58	32.92	7.65	5.22	4.51	6.43	9.45	11.73	13.37	14.80
Co-60	3.83	2.51	2.71	3.16	4.49	5.64	6.49	7.00	7.67
Co-57	14.26	8.44	4.44	2.50	2.60	3.82	4.57	5.21	5.85
Fe-55	3.85	8.44	5.16	1.95	1.02	1.28	0.98	0.99	0.98
Be-7	0.01	3.92	9.18	9.17	5.34	3.05	1.76	1.27	0.92
Mn-54	0.43	5.60	4.15	1.47	0.78	1.14	0.75	0.79	0.79
Co-56	0.09	3.58	1.97	0.80	0.44	0.50	0.51	0.52	0.56
Fe-59	0.02	0.34	0.13	0.13	0.13	0.28	0.30	0.36	0.41
V-49	0.00	5.35	4.86	3.81	1.23	1.08	0.48	0.40	0.35

SIMULATION **RESULTS**



> Activities were calculated immediately after the end of the irradiation.

> A_P(1GeV) is a normalised activity induced by protons with the energy of 1GeV during 100 days of the irradiation.

Tolerable beam losses for different ion beams with the different energies [W/m]													
Energy Mass	1 GeV/u	900 MeV/u	800 MeV/u	700 MeV/u	600 MeV/u	500 MeV/u	400 MeV/u	300 MeV/u	200 MeV/u	150 MeV/u	100 MeV/u	50 MeV/u	25 MeV/u
^{1}H	0.73	0.74	0.75	0.77	0.80	0.85	0.94	1.11	1.45	1.77	~~2.32~~	~~3.83~~	~~7.88~~
^{4}He	0.66	0.66	0.66	0.67	0.70	0.75	0.84	1.03	1.47	2.00	~~3.05~~	~~5.91~~	~~13.14~~
^{12}C	0.88	0.92	0.98	1.07	1.21	1.43	1.82	2.58	4.54	7.11	~~12.04~~	~~25.14~~	~~53.85~~
^{20}Ne	1.12	1.21	1.33	1.50	1.76	2.18	2.87	4.24	7.71	11.94	~~19.99~~	~~44.65~~	~~104.20~~
^{40}Ar	1.46	1.61	1.81	2.09	2.50	3.16	4.25	6.35	11.38	16.84	~~26.71~~	~~58.16~~	~~153.44~~
^{84}Kr	2.24	2.51	2.88	3.40	4.15	5.31	7.25	10.94	19.13	26.86	~~39.76~~	~~83.64~~	~~209.61~~
^{132}Xe	2.83	3.20	3.68	4.36	5.36	6.87	9.40	14.06	24.13	32.55	~~47.29~~	~~95.15~~	~~226.33~~
^{197}Au	3.62	4.10	4.73	5.60	6.90	8.87	12.05	17.87	30.16	40.01	~~56.43~~	~~104.41~~	~~233.44~~
^{238}U	3.75	4.25	4.89	5.81	7.12	9.11	12.32	18.17	30.02	39.43	~~54.52~~	~~97.56~~	~~217.67~~

CRITERION **SUMMARY**

	FLUKA 2008.3.6*	FLUKA 2011.2b.5**	FLUKA 2011.2c.4
	Tolerable beam losses for Uranium beam [W/m]		
Irradiation >	100 days		20 years
1GeV/u	5	5	4
500 Mev/u	12	12	9
200 MeV/u	60	40	30
150 MeV/u		52	39

Beam losses criteria for heavy ion accelerators are **more strict** than considered before.

The nuclide inventory observed **heterogeneous** presence of nuclides after long-term accumulation.

Generic Curve (GC) is not fulfilling the main assumption of the stable production of the radioactive nuclei.

Using the beam losses criteria in the range of lower energies (**below ~ 200MeV/u**) is not recommended.

Calculations were provided for the copper bulky target, but similar results are expected for different materials as well, as it has been shown before on the stainless steel targets.

WHAT ARE THE POSSIBLE SOLUTIONS?

> Using the "GCs" and the scaling law only for high energy heavy ion accelerators.

> Search for a brand new approach of beam losses criteria determination for heavy-ions with the energies below 200 MeV/u.

* I.STRASIK et al.: Residual activity induced by heavy ions and beam-loss criteria for heavy-ion accelerators, Phys. Rev. ST Accel. Beams 13, (2010)

** P. Katrík, et al. - Proceedings of the 12th Shielding Aspects of Accelerators, Targets and Irradiation Facilities SATIF 12 (2014) 124-134

HHD BEAM DUMP ▸ GSI DARMSTADT

2 x CCD cameras
Lightening screen
Al 200
Cu 200
Al 100
Cu100
2 x platforms
Target holders
HHD DK 3

~ 80cm
ø=5–10 cm
Al 200
Cu 200
Al 100
Cu100
Target holders
60cm + 100um
(air gap + stainless steel vacuum window)
HHD DK 3

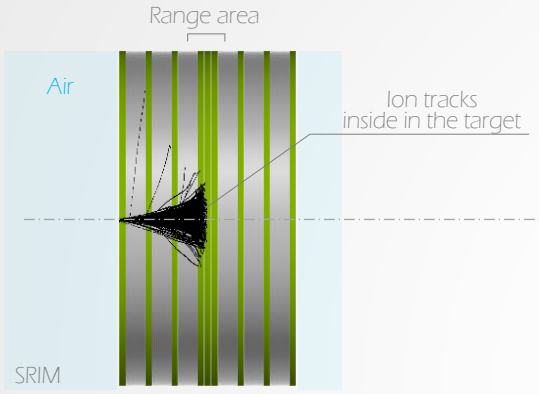
IRRADIATION

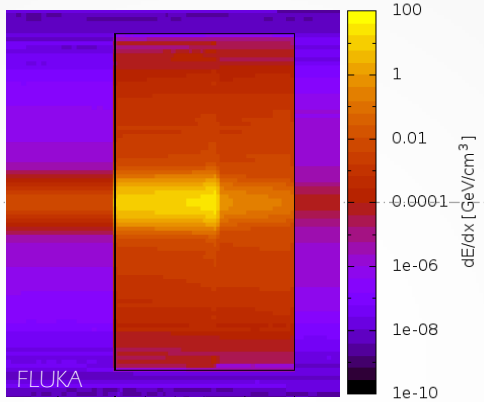
- ^{238}U [200 MeV/u] | NOI: 2.9 x10^{12}
- ^{238}U [125 MeV/u] | NOI: 1.2 x10^{12}
- ^{124}Xe [300 MeV/u] | NOI: 2.5 x 10^{12}

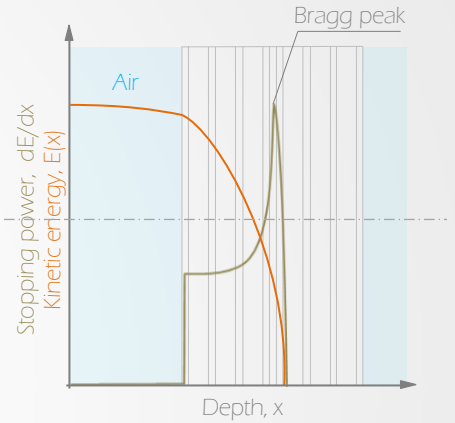
- DC Current transformers @ HHD

TARGETS

- The aluminium foils had a diameter of 5 cm - 10 cm
- Al - purity of 99.95% and density of 2.7 g/cm^3 at 20°C
- Assembled in "stacked-foil" geometry







Ion tracks are damage-trails created by swift heavy ions penetrating through solids.*

Stopping power in nuclear physics is defined as the retarding force acting on charged particles due to interaction with matter, resulting in loss of particle energy.**

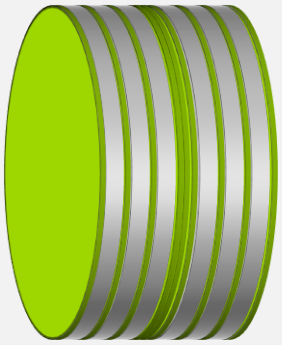
Bragg peak occurs because the interaction cross section increases as the charged particle's energy decreases.***

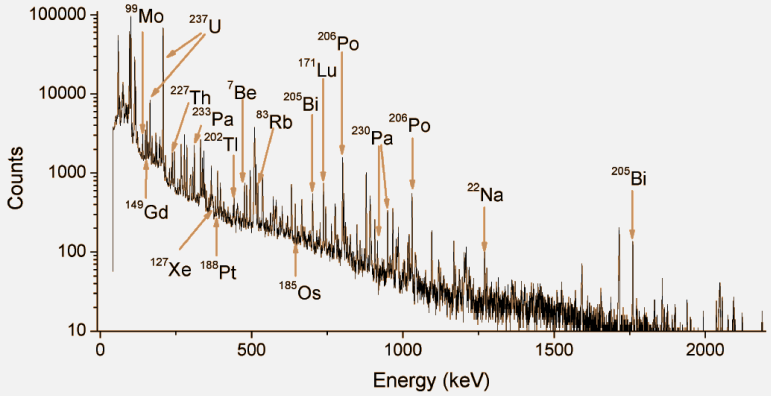
* D.A. Young "Etching of radiation damage in lithium fluoride". Nature. 182 (4632): 375–377 (1958). doi:10.1038/182375a0

** ICRU Report 73: Stopping of Ions heavier than Helium, Journal of the ICRU, 5 No. 1 (2005), Oxford Univ. ISBN 0-19-857012-0

*** Charlie Ma, C-M; Lomax, Tony. Proton and carbon ion therapy. Boca Raton: CRC Press. p. 4. (2012) ISBN 9781439816073

Foils with different thicknesses

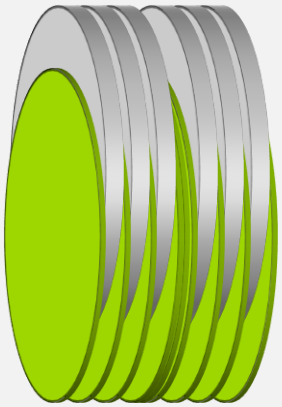


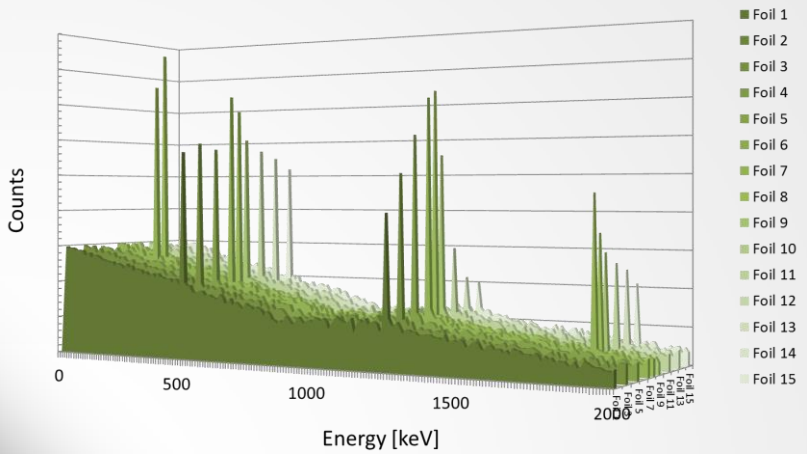


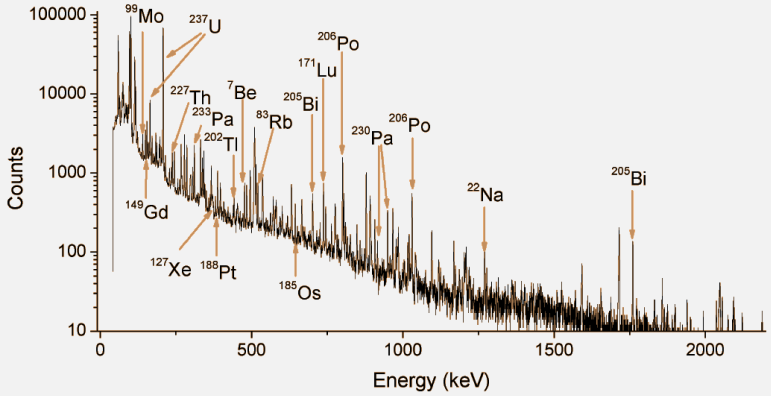
Gamma spectroscopy:

Detector Canberra® HPGe – GC3518 powered by Ortec HV-Supply (energy resolution of 1.8keV FWHM at the 1332 keV ^{60}Co). The detector was coupled with spectrometer ORTEC® DESPEC LF TM (8192 channels). Graphical interface of GammaVision® was used to analyse the Peak-Net-Area.

Foils with different thicknesses







Different shapes of depth-profiles:

> target- nuclei fragments
> primary-projectiles fragments

Gamma spectroscopy:

Detector Canberra® HPGe – GC3518 powered by Ortec HV-Supply (energy resolution of 1.8keV FWHM at the 1332 keV ^{60}Co). The detector was coupled with spectrometer ORTEC® DESPEC LF TM (8192 channels). Graphical interface of GammaVision® was used to analyse the Peak-Net-Area.

DEPTH PROFILES OF THE RESIDUAL ACTIVITY

Nuclide > ^{7}Be | Irr > ^{238}U | E_{ini} > 200 [Mev/u] | A_E [Bq/ion] $2.63x10^{-10} \pm 1.44x10^{-11}$ | A_S [Bq/ion] $3.72x10^{-10} \pm 8.91x10^{-12}$ | A_E/A_S: 0.71

TARGET-NUCLEI FRAGMENT

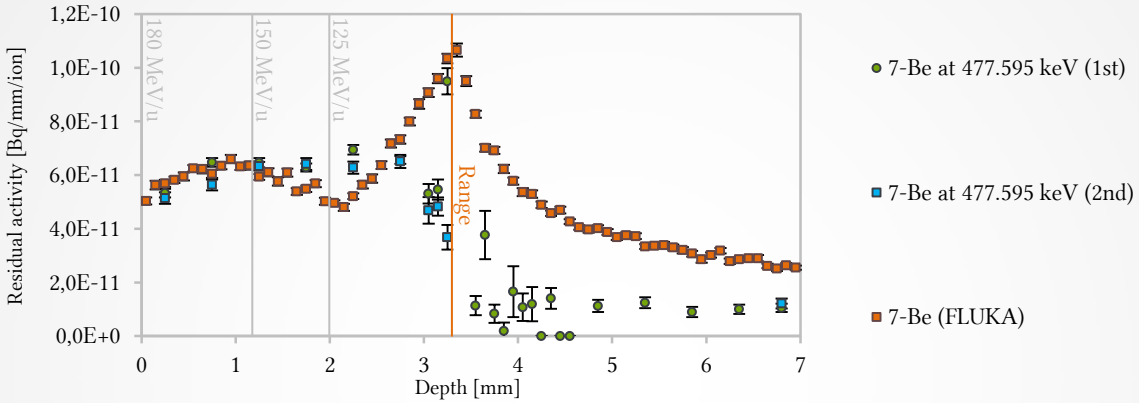
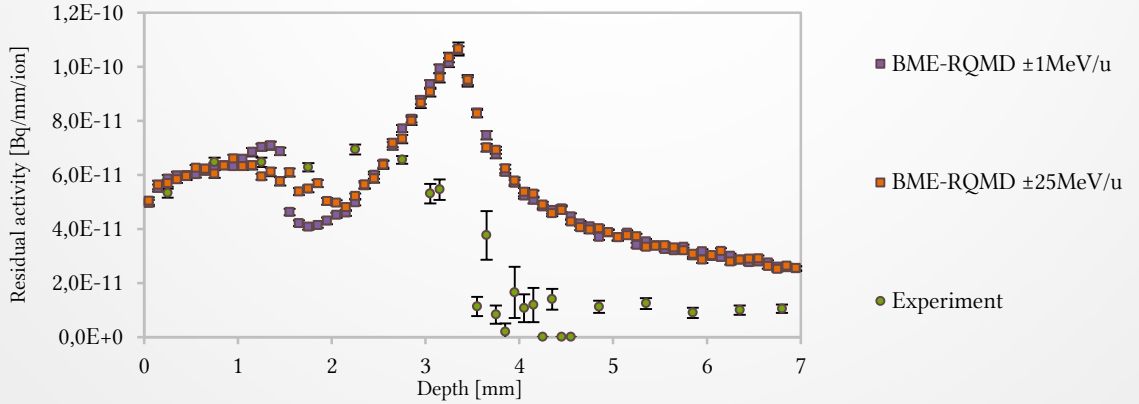


Figure: The depth profile of ^{7}Be in an aluminium target irradiated by ^{238}U with energy of 200 MeV/u.



Is the depth profile influenced by changing between two physical models around 125MeV/u ?

DEPTH PROFILES OF THE RESIDUAL ACTIVITY

TARGET-NUCLEI FRAGMENT

Nuclide > ^{7}Be | Irr > ^{238}U | E_{ini} > 125 [Mev/u] | [Bq/ion] $7.63\times10^{-11} \pm 1.16\times10^{-11}$ | [Bq/ion] $3.06\times10^{-10} \pm 1.84\times10^{-11}$ | A_E/A_S: 0.25 (10 / 0.1)

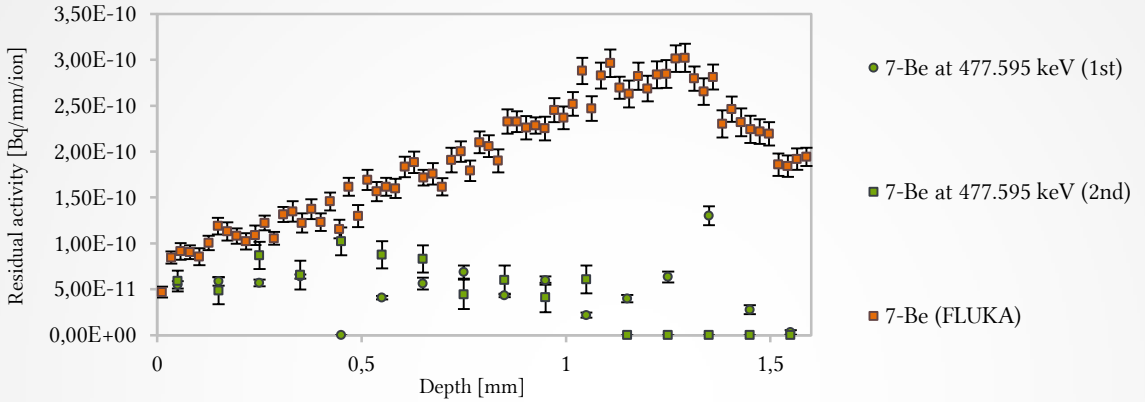


Figure: The depth profile of ^{7}Be in an aluminium target irradiated by ^{238}U with energy of 125 MeV/u.

Nuclide > ^{7}Be | Irr > ^{124}Xe | E_{ini} > 300 [Mev/u] | A_E [Bq/ion] $7.31\times10^{-10} \pm 3.15\times10^{-11}$ | A_S [Bq/ion] $1.24\times10^{-9} \pm 3.19\times10^{-11}$ | A_E/A_S: 0.59 (10 / 0.1)

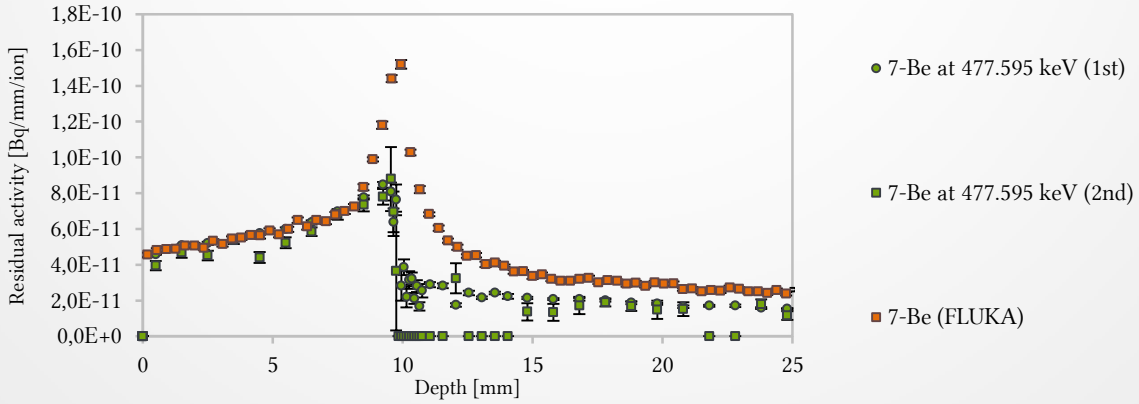


Figure: The depth profile of ^{7}Be in an aluminium target irradiated by ^{124}Xe with energy of 300 MeV/u.

DEPTH PROFILES OF THE RESIDUAL ACTIVITY

Nuclide > ^{22}Na | Irr > ^{238}U | E_{ini} > 200 [Mev/u] | A_E [Bq/ion] $1.49x10^{-11} \pm 6.63x10^{-13}$ | A_S [Bq/ion] $2.36x10^{-11} \pm 5.72x10^{-13}$ | A_E/A_S: 0.63 (10 / 0.1)

TARGET-NUCLEI FRAGMENT

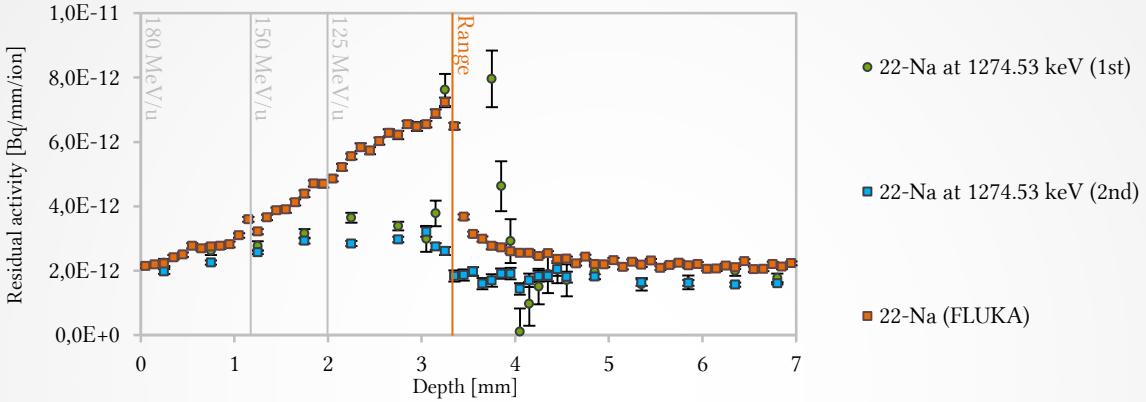
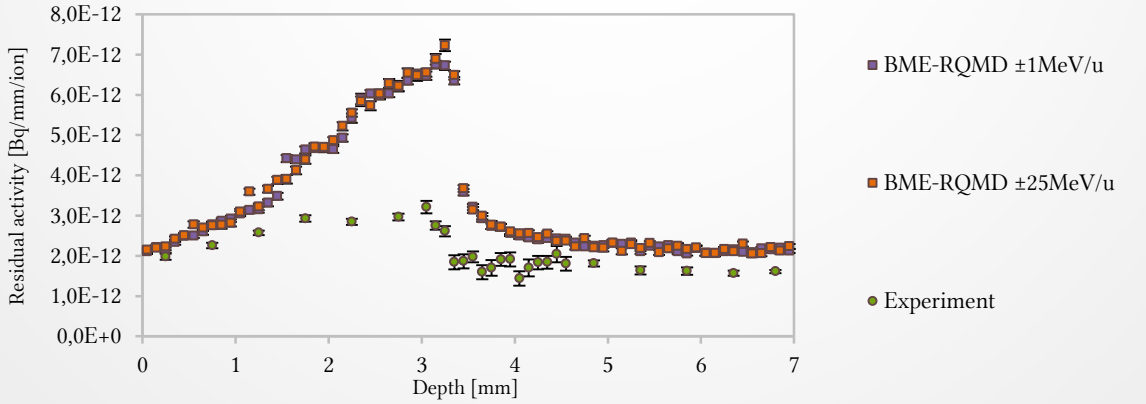


Figure: The depth profile of ^{22}Na in an aluminium target irradiated by ^{238}U with energy of 200 MeV/u.



DEPTH PROFILES OF THE RESIDUAL ACTIVITY

TARGET-NUCLEI FRAGMENT

Nuclide > ^{22}Na | Irr > ^{238}U | E_{ini} > 125 [Mev/u] | [Bq/ion] $3.44x10^{-12} \pm 4.04x10^{-13}$ | [Bq/ion] $2.10x10^{-11} \pm 1.20x10^{-12}$ | A_E/A_S: 0.16

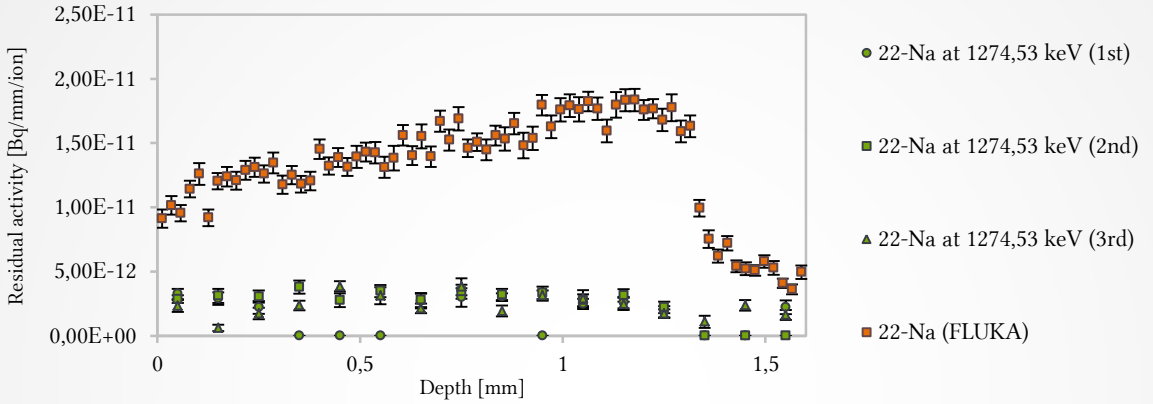


Figure: The depth profile of ^{22}Na in an aluminium target irradiated by ^{238}U with energy of 125 MeV/u.

Nuclide > ^{22}Na | Irr > ^{124}Xe | E_{ini} > 300 [Mev/u] | A_E [Bq/ion] $5.91x10^{-11} \pm 1.16x10^{-12}$ | A_S [Bq/ion] $1.00x10^{-10} \pm 2.32x10^{-12}$ | A_E/A_S: 0.59

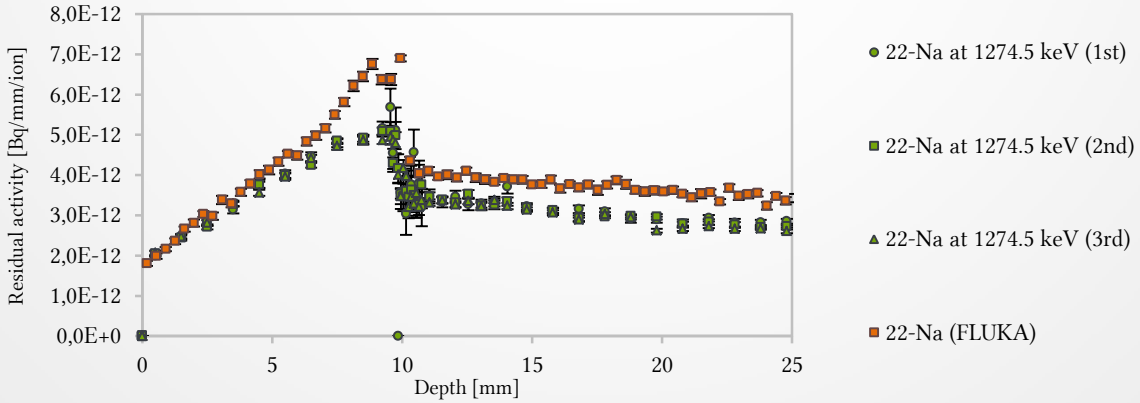


Figure: The depth profile of ^{22}Na in an aluminium target irradiated by ^{124}Xe with energy of 300 MeV/u.

DEPTH PROFILES OF THE RESIDUAL ACTIVITY

Nuclide > ^{127}Xe | Irr > ^{238}U | E_{ini} > 125 [Mev/u] | A_E [Bq/ion] $3.88 \times 10^{-11} \pm 1.67 \times 10^{-12}$ | A_S [Bq/ion] $1.89 \times 10^{-10} \pm 8.53 \times 10^{-12}$ | A_E/A_S: 0.21 (10 / 0.1)

PRIMARY-PARTICLE FRAGMENT

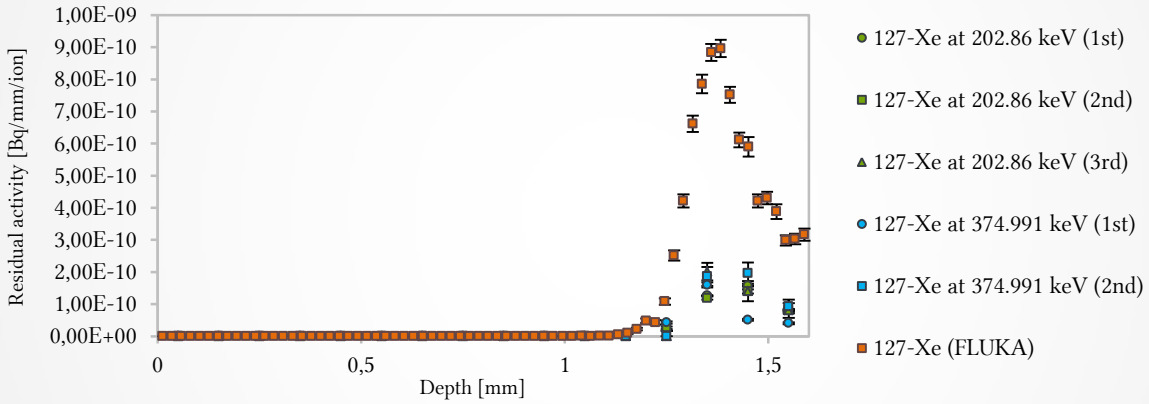


Figure: The depth profile of ^{127}Xe in an aluminium target irradiated by ^{238}U with energy of 125 MeV/u.

DEPTH PROFILES OF THE RESIDUAL ACTIVITY

Nuclide > ^{170}Lu | Irr > ^{238}U | E_{ini} > 200 [Mev/u] | A_E [Bq/ion] $7.78x10^{-10} \pm 2.70x10^{-11}$ | A_S [Bq/ion] $2.33x10^{-9} \pm 5.50 x10^{-12}$ | A_E/A_S: 0.33

PRIMARY-PARTICLE FRAGMENT

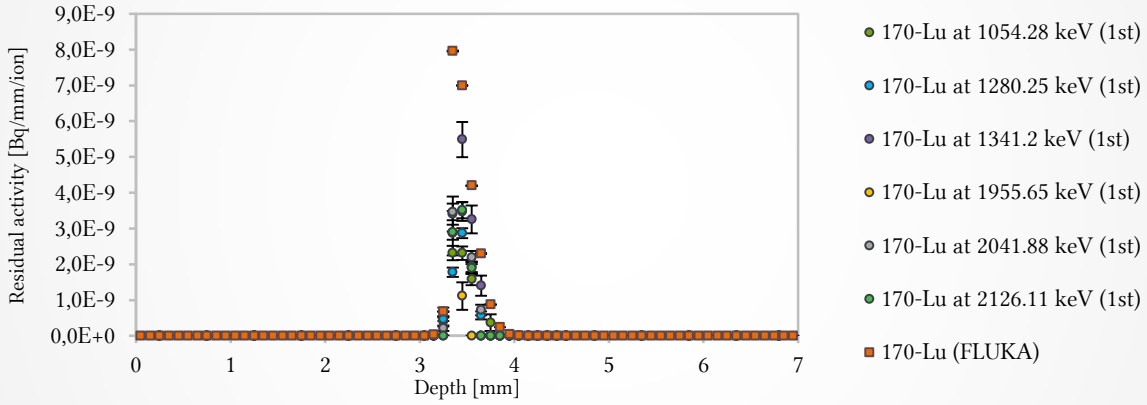


Figure: The depth profile of ^{170}Lu in an aluminium target irradiated by ^{238}U with energy of 200 MeV/u.

DEPTH PROFILES OF THE RESIDUAL ACTIVITY

Nuclide > ^{48}V | Irr > ^{124}Xe | E_{ini} > 300 [Mev/u] | A_E [Bq/ion] $1.28\times10^{-10} \pm 6.22\times10^{-12}$ | A_S [Bq/ion] $2.02\times10^{-10} \pm 1.32\times10^{-11}$ | A_E/A_S: 0.63 (10 / 0.1)

PRIMARY-PARTICLE FRAGMENT

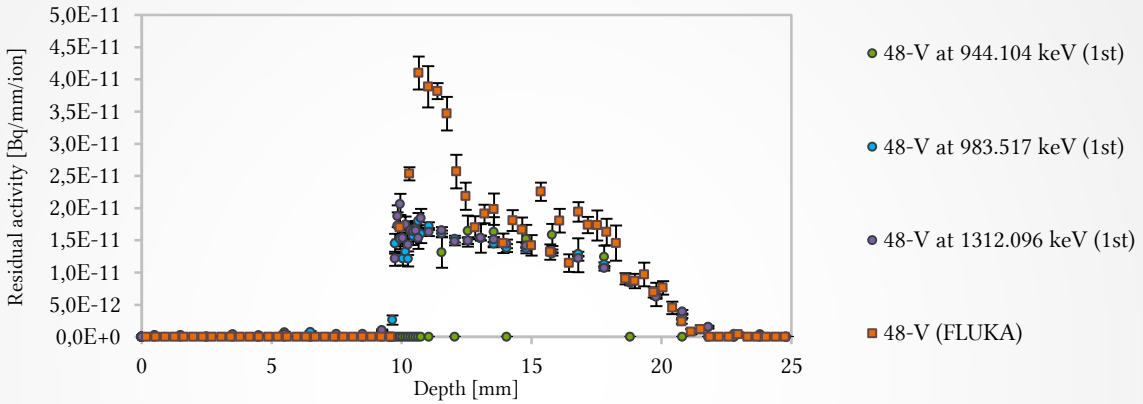
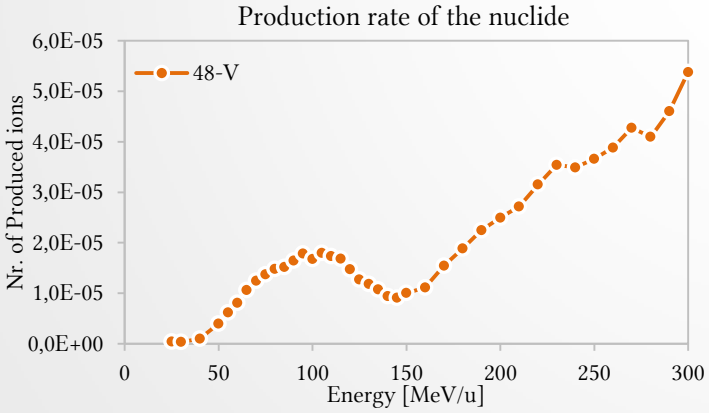
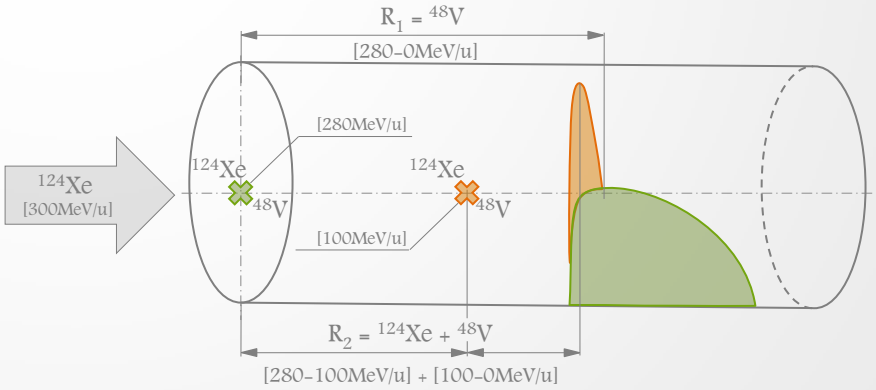


Figure: The depth profile of ^{48}V in an aluminium target irradiated by ^{124}Xe with energy of 300 MeV/u.





Observed in case of intermediate-Z fragments: ^{46}Sc, ^{48}V, ^{52}Mn, ^{54}Mn, ^{56}Co, ^{58}Co, ^{65}Zn

DEPTH PROFILES OF THE RESIDUAL ACTIVITY

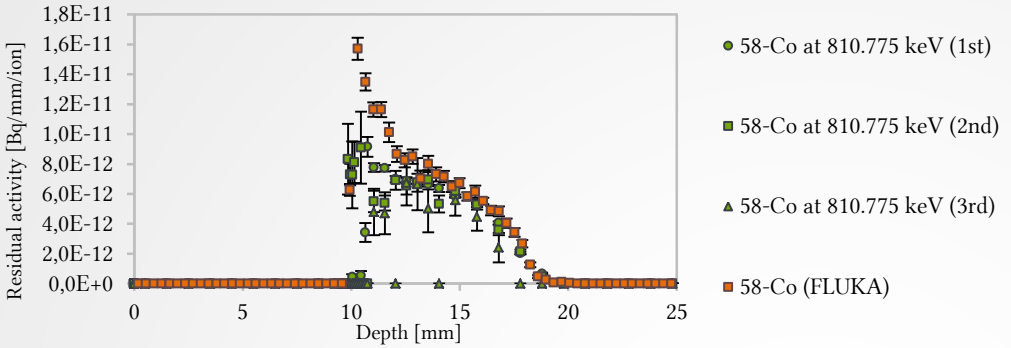


Figure: The depth profile of ^{58}Co in an aluminium target irradiated by ^{124}Xe with energy of 300 MeV/u.

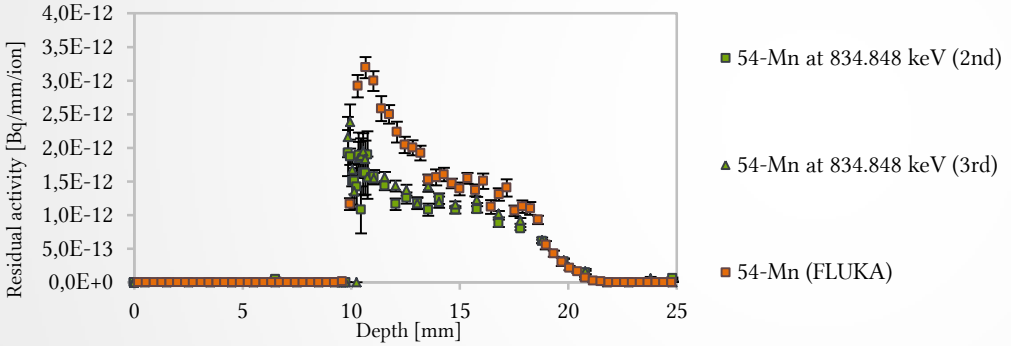


Figure: The depth profile of ^{54}Mn in an aluminium target irradiated by ^{124}Xe with energy of 300 MeV/u.

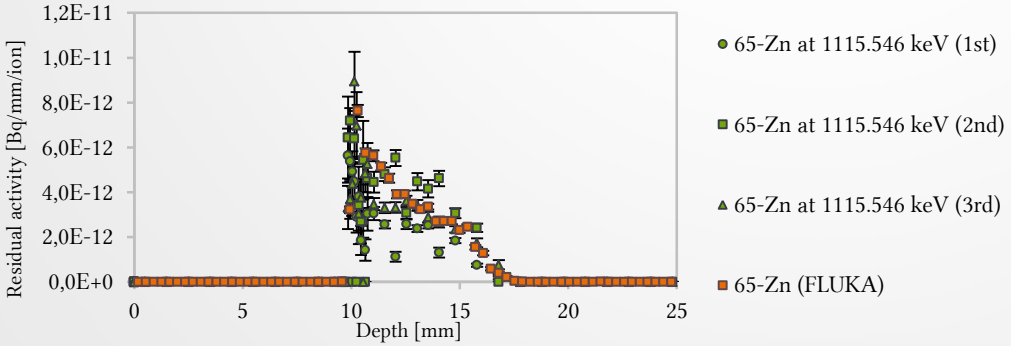


Figure: The depth profile of ^{65}Zn in an aluminium target irradiated by ^{124}Xe with energy of 300 MeV/u.

DEPTH PROFILES OF THE RESIDUAL ACTIVITY

Nuclide > ^{237}U | Irr > ^{238}U | E_{ini} > 200 [Mev/u] | A_E [Bq/ion] $6.74x10^{-9} \pm 1.35x10^{-11}$ | A_S [Bq/ion] $4.02x10^{-9} \pm 8.40x10^{-12}$ | A_E/A_S: 1.62 (10 / 0.1)

PRIMARY-PARTICLE FRAGMENT

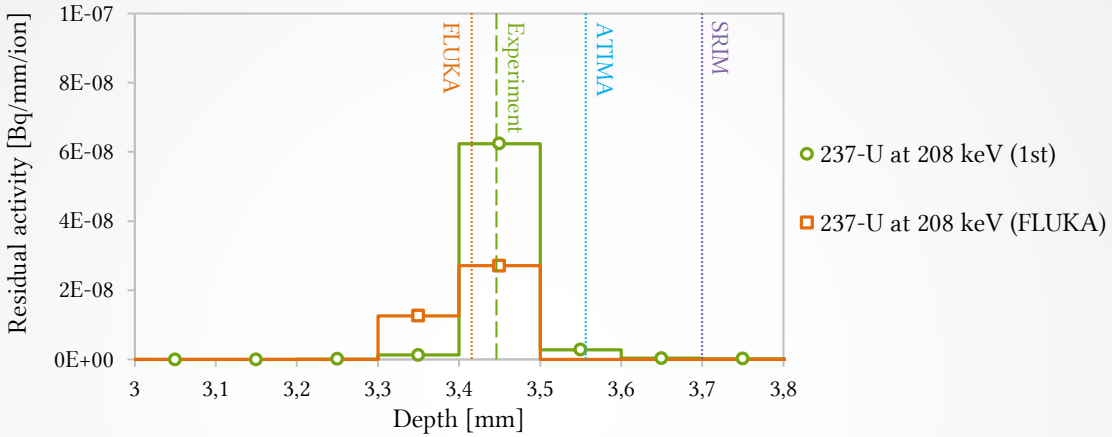


Figure: The depth profile of ^{237}U in an aluminium target irradiated by ^{238}U with energy of 200 MeV/u.

	ATIMA	SRIM	FLUKA	EXPERIMENT
	Range ± range straggling [mm]			
^{238}U	3.722 ± 0.003	3.874 ± 0.010	3.442 ± 0.004	- - - - - - -
^{237}U	3.569 ± 0.003	3.713 ± 0.007	3.423 ± 0.007	3.456 ± 0.004

The depth profile of ^{237}U can be used for crosschecking of the primary particles range calculations. The difference between simulated and experimentally measured range is less than 1%.

DEPTH PROFILES OF THE RESIDUAL ACTIVITY

Nuclide > ^{120m}Sb | Irr > ^{238}U | E_{ini} > 200 [Mev/u] | A_E [Bq/ion] $2.63\times10^{-10} \pm 8.62\times10^{-12}$ | A_S [Bq/ion] .... | A_E/A_S: 10 .... 0.1

PRIMARY-PARTICLE FRAGMENT

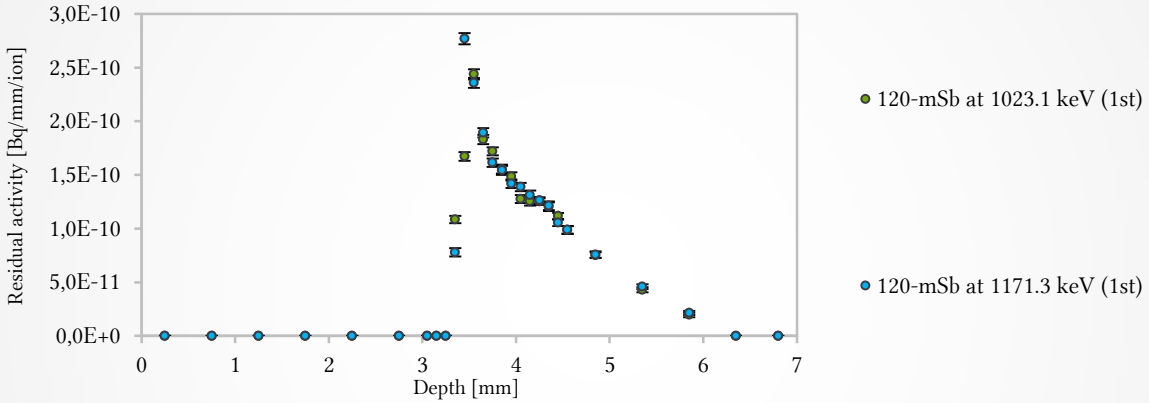


Figure: The depth profile of ^{120m}Sb in an aluminium target irradiated by ^{238}U with energy of 200 MeV/u.

Set of several nuclides in their metastable state were experimentally measured and their depth profiles were created:

> ^{101m}Rh, ^{106m}Ag, ^{110m}Ag, ^{119m}Te, ^{120m}Sb, ^{121m}Te, etc.

VALIDATION **SUMMARY**

Set of three experimental irradiations were performed for crosschecking and validation of the FLUKA code at GSI Darmstadt.

Range calculations: The calculated range of the primary particles was compared with the experimental data of aluminium target irradiated by ^{238}U with energy of 200 MeV/u. The observed difference is less than 1%.

Depth profile shapes: the shape of the depth profiles of the primary-particles fragments (intermediate-Z) simulated by FLUKA exhibits a difference compare to the experimentally measured ones.

BME – RQMD energy range: the results of the transportation of heavy ions in matter are facing some difficulties.

> target-nuclei fragments: ^{7}Be and ^{22}Na (residual activities of first foils are only correct)

> primary-ions fragments: intermediate-Z nuclei

OUTCOMING QUESTIONS:

> Is a transition between RQMD (Relativistic Quantum Molecular Dynamics) and the BME (Boltzmann Master Equation) sufficiently smooth?

> Why are the depth profiles of the target-nuclei fragments in agreement with the simulation only in the first foils of the target?

> Is there any option to calculate the residual activity of isomeric nuclides and score them with the RESNUCLEi detector?

THANK YOU

FOR YOUR ATTENTION

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