

The nELBE neutron beamline at HZDR: overview and shielding calculations

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- > The Transmutation Research at HZDR: MYRRHA and nELBE
- A quick look at the measurement program at nELBE
- An overview of the first (2007-2011) nELBE neutron beamline and of the new beamline and experimental room
- Monte Carlo studies for the nELBE upgrade: radiation field characterization
- Shielding calculations
- Signal/background optimization
- Some considerations about activation







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Motivation for the transmutation research (I)

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Source: Salvatores, Palmiotti, Progress in Part. And Nucl. Phys. 66 (2011)

From the IAEA FR13 Conference (March 2013, Paris):

-USA produces <u>~2000 MT</u> spent fuel per year;

estimated inventory by the end of
2012: <u>70000 MT</u> spent fuel

U Pu **Minor Actinides** Long-lived Fission Short-lived Fission **Stable Isotopes** Products Products 0.5 kg ²³⁷Np 1.0 kg ¹³⁷Cs 955.4 kg 8.5 kg 0.2 kg ¹²⁹ 10.1 kg Lanthanides 0.7 kg ⁹⁰Sr 0.8 kg ⁹⁹Tc 0.6 kg Am 21.8 kg other 0.7 kg ⁹³Zr 0.02 kg Cm 0.3 kg ¹³⁵Cs



Separated transuranic elements can be "transmuted" in shorter-lived elements in a neutron field, where fission and neutron capture processes are always in competition

> Fast neutron spectrum appears to be the most adapted to support transmutation

Different strategies can be followed... one is the ADS option



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(*) The nELBE group at HZDR:

- R. Beyer, E. Birgersson, A. Ferrari, E. Grosse, R. Hannaske, A.Junghans, T. Koegler,
- I. Koesterke, R. Massarczyk, A. Matič, K.-D. Schilling, R. Schwengner, G.Schramm, A. Wagner

A look at the research program at nELBE



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Table 32. Summary of Highest Priority Target Accuracies for FastReactors

		Energy Range	Current Accuracy (%)	Target Accuracy (%)	
11228	σ_{inel}	6.07 ÷ 0.498 MeV	$5.07 \div 0.498 \text{ MeV}$ $10 \div 20$ $2 \div 3$		
0238	σ_{capt}	24.8 ÷ 2.04 keV	3 ÷ 9	1.5 ÷ 2	
Du241		1.25 MoV $\div 454$ eV	8 ÷ 20	2÷3 (SFR,GFR, LFR)	
Pu241	o _{fiss}	1.35MeV ÷ 454 eV	8 ÷ 20	$5 \div 8$ (ABTR, EFR)	
Pu239	σ_{capt}	498 ÷ 2.04 keV	7 ÷ 15	4 ÷ 7	
Du240	$\sigma_{\rm fiss}$	1.35 ÷ 0.498 MeV	6	1.5 ÷ 2	
r u240	ν	1.35 ÷ 0.498 MeV	4	1 ÷ 3	
Pu242	$\sigma_{\rm fiss}$	2.23 ÷ 0.498 MeV	19÷21	3 ÷ 5	
Pu238	$\sigma_{\rm fiss}$	1.35 ÷ 0.183 MeV	17	3 ÷ 5	
Am242m	$\sigma_{\rm fiss}$	1.35MeV ÷ 67.4keV	17	3 ÷ 4	
Am241	$\sigma_{\rm fiss}$	6.07 ÷ 2.23 MeV	12	3	
Cm244	$\sigma_{\rm fiss}$	1.35 ÷ 0.498 MeV	50	5	
Cm245	σ_{fiss}	183 ÷ 67.4 keV	47	7	
Fe56	σ_{inel}	2.23 ÷ 0.498 MeV	16÷25	3 ÷ 6	
Na23	σ_{inel}	1.35 ÷ 0.498 MeV	28	4÷10	
Pb206	$\sigma_{\rm inel}$	2.23 ÷ 1.35 MeV	14	3	
Pb207	σ_{inel}	1.35 ÷ 0.498 MeV	11	3	
G:20	σ_{inel}	6.07 ÷ 1.35 MeV	14 ÷ 50	3 ÷ 6	
5128	σ_{capt}	19.6 ÷ 6.07 MeV	53	6	

Goal: Measuring fast neutron reactions with the required accuracy needed to develop Nuclear Trasmutation Facilities (structural & coolant materials)

- Neutron Inelastic scattering (n,n 'γ) for ⁵⁶Fe, Mo, Pb, Na and <u>Total neutron cross sections</u> σ_{tot} (Ta, Au, Al, C, H)
- Fission cross sections of minor actinides (radioactive targets)
 Collaboration with n-TOF at CERN

Based on the NEA high priority request list and on the OECD Working Party on Evaluation Co-operation, subgroup 26

http://www.nea.fr/html/science/wpec/volume26/volume26.pdf

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ELBE: Electron Linear accelerator with high Brilliance and low Emittance

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1: Diagnostic station, IR-imaging and biological IR experiment

- 2: Femtosecond laser, THz-spectroscopy, IR pump-probe experiment
- 3: Time-resolved semiconductor spectroscopy, THz-spectroscopy

- 4: FTIR, biological IR experiment
- 5: Near-field and pump-probe IR experiment
- 6: Radiochemistry and sum frequency generation experiment, photothermal deflection spectroscopy

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The old experimental halls (2007-2011)

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Photo-neutron source



Experimental room





²³⁵U fission chamber H19 from PTB.

Time resolution from peak width = 3.8 ns FWHM



The nELBE neutron spectrum

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Measurement time : 49.4 h $I_{e_{-}}$ = 15 µA, $E_{e_{-}}$ = 31 MeV Flight path 618 cm total uncertainty 3.5%

Absorption dips : 78,117, 355, 528, 722, 820 keV ²⁰⁸Pb scattering resonances Emission peaks: 40,89,179, 254, 314, 605 keV near threshold photoneutron emission In ²⁰⁸Pb (strong capture resonances of ²⁰⁷Pb)

R. Beyer et al. submitted to NIM A (2012)



<u>New Neutron Time-of-Flight Facility for Transmutation Studies</u>

nELBE is a partner in the EURATOM FP7 <u>ERINDA</u> and (from 2013) CHANDA projects Transnational access is supported. External users are very welcome

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Time-of-flight hall 6 m x 6 m x 9 m





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Strength of the improved source

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Neutron and photon total yields at $E_{e^{-}} = 50 \text{ MeV}$ (FLUKA Simulation)



At the entrance of the collimator:

 $n_{yield} = 4.34 \ 10^{-8} \ n \ per \ cm^2 \ per \ primary \ e^{-8}$

A deeper look at the photoproduction simulation

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Comparison of the total cross sections in FLUKA and in the MCNP code

An important difference:

- **MCNP** works with differential cross sections, for each reaction channel
- FLUKA uses the total cross sections to determine where the interaction occurs, then proceeds with the models (PEANUT[pre-equilibrium], evaporation and Fermi break-up)







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Calculation of the total yields (Source Strength)

Electron Energy (MeV)	Neutron Yield (n/e ⁻)	Neutron Source Strength at the radiator (n/s)		
	FLUKA	MCNP (*)	FLUKA	
20	1.205 10-3	7.9 10 ¹²	7.52 1012	
30	3.108 10-3	1.9 10 ¹³	1.94 1013	
40	4.51 10-3	2.7 10 ¹³	2.81 1013	
50	5.67 10-3		3.54 1013	

Hyp: 1 mA current $\rightarrow 6.24 \ 10^{15} \ e^{-/s}$

(*) nELBE published results: Ann. of Nucl. En. 34 (2007) 36-50

FLUKA statistical accuracy: < 1%

MCNP and FLUKA agree at the level of few percent in the yield calculation

An additional check: the comparison with the Swanson evaluations

Swanson (SLAC-PUB-2042, 1978)

calculated the neutron yields from semiinfinite slabs of materials by folding the measured photoneutron cross sections with the numerical integration of the photon track length distributions (derived from the analytical theory of the showers)

9.3·10¹⁰ Z^(0.73 ± 0.05) neutrons s⁻¹ kW⁻¹

The formula gives, for the asymptote of the curve of the lead: $2.32 \cdot 10^{12}$ n s⁻¹ kW⁻¹

This value is valid for semiinfinite slabs and at high energies: we have to correct in real cases.

By correcting for the energy (for ex. @ 30 MeV) and for the finite dimensions of the slab ($2X_0$ in our case) we get:

1.92 10¹³ n s⁻¹ @1 mA and @30 MeV

in perfect agreement with both MCNP and FLUKA



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- 1. Complete description of geometry and materials of the photo-neutron target, the Al-Lead beam dump, the Lead/Poly-Bor collimator, the walls and the final Lead/Poly-Bor Dump
- 2. To compute all the dose rates in the photo-neutron source hall:

We start from the photoproduction process, to have in each point the mixed field given by the neutron (isotrope) source + the Bremsstrahlung spectrum

3. To compute all the dose rates in the nELBE experimental room:

We use as source term the secondary neutron and photon spectra, calculated in the direction of the collimator



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Conversion coefficients from fluence to ambient dose equivalent are based on ICRP74 values and values calculated by M.Pelliccioni. They are implemented for protons, neutrons, charged pions, muons, photons, electrons (conversion coefficients for other particles are approximated by these).

In the card: AMB74 is the default choice for dose equivalent calculation



Dose profiles through the walls of the photo-neutron hall



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Horizontal section through the neutron hall



Definition of the volumes for the calculation:

Profile I (wall opposite to the neutron beamline)

a column along x, large: (-50 cm, 50 cm) in z (-50 cm, 50 cm) in y

Profile II (wall behind the beam dump)

a column along z, large: (-50 cm, 50 cm) in x (-50 cm, 50 cm) in y



Profile I. Wall opposite to the neutron beamline

Profile II. Wall behind the PNQ



Extrapolation: **1** μ **Sv/h** at ~ -525 cm (after 200 cm in the heavy concrete)



Extrapolation: **1** μ **Sv/h** at ~300 cm (after 150 cm in the heavy concrete)

Dose profiles through the roof of the photo-neutron hall



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Solution: 160 cm of heavy concrete



With this solution we find a dose rate of about $0.5 \mu Sv/h$ at the exit of the roof.



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Optimization of the longitidinal shielding in the neutron experimental room



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Dose profiles through the walls of the experimental room



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The H*(10) profile is calculated by averaging:

- on a quite narrow region above and below the beam level:

(-24 cm, 24 cm) in y

 on a large region in x (covering all the lateral walls)
and in steps of 2.5 cm in z.

The gamma component of the radiation is rapidly attenuated, faster then the neutron one.

This behaviour is different if compared with the (more critical) longitudinal radiation profiles

The reason is in the different energy spectrum of the photon radiation

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Photon energy spectra in the experimental room

H*(10) rate (uSv/h) from photons /acjual Beam Du/mp

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The photon energy spectrum has been studied considering three different detectors:

- Det.1 at the beam dump surface;
- Det. 2 at the surface of the wall, where the beam is impinging ;
- Det. 3 at the surface of the lateral walls and of the roof.

The radiation, that reaches the roof and the lateral walls, is essentially a low energy radiation, with the higher tail until only 6-7 MeV. The spectrum of the forward radiation is extended – as expected – until the higher energy value of the Bremsstrahlung (50 MeV).



Det 1 Det 2 Det 3

1 0+10





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Goal: choose a direction of the neutron beam-line that maximize the ratio $n_{yield}/\gamma_{yield,}$ taking into account the isotropy of the neutron production and the typical shape of the bremsstrahlung

The optimized direction is implemented in the new neutron beam-line by rotating the whole photo-neutron source (liquid lead target + dump)





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The real source





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In real life we must avoid the 'contamination' coming from the neutrons scattered on the beam dump (or photoproduced in the dump material).

A sizeable

contamination starts to be visible at 110° (around 1%). At 115° it is still at an acceptable level (around 3%)

	90°	95.5°	100°	105°	110°	115°
ldeal source n _{yield} /γ _{yield}	2.20 10 ⁻³	2.38 10 ⁻³	2.52 10 ⁻³	2.77 10 ⁻³	2.88 10 ⁻³	3.05 10 ⁻³
Real source n _{yield} /γ _{yield}	2.17 10 ⁻³	2.39 10 ⁻³	2.53 10 ⁻³	2.78 10 ⁻³	2.92 10 ⁻³	3.14 10 ⁻³

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The new nELBE experimental room

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Floor plan of the new *n*ELBE neutron source and low scattering experimental hall. Project at KAERI T.Y. Song et al., Journal of the Korean Physical Society, Vol. 59, No. 2, (2011) 1609

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Residual radiation in FLUKA: residual dose rates and specific activity



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✓ For an arbitrary irradiation pattern, the time evolution of the system (buildup and decay during the irradiation and cooling) is obtained <u>runtime</u> via the exact analytical solution of the **Bateman equations**:

$$\frac{dN_i}{dt} = -\sum_{\substack{j \neq i}} \left[\lambda_{ji}^d + \overline{\sigma}_{ji} \overline{\varphi} \right] N_i + \sum_{\substack{j \neq i}} \left[\lambda_{ij}^d + \overline{\sigma}_{ij} \overline{\varphi} \right] N_j \quad \text{for each radionuclide in the material}$$

where
$$\overline{\sigma}_{ji} = \frac{1}{\overline{\varphi}} \int \varphi(E) \sigma_{ji}(E) dE$$
 and $\overline{\varphi} = \int \varphi(E) dE$

The specific activity of each material at the cooling time t_{cool} is given by: $a(t_{cool}) = N\sigma\phi(1 - e^{-\lambda t_{irr}})e^{-\lambda t_{cool}}$

✓ At the same time FLUKA can perform the generation and transport of the decay radiation

In the same run we can obtain production of the residuals, their time evolution and the residual doses due to their decays



FLUKA model used for the activation studies

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- a detailed model of the Photo-neutron source (Liquid Lead + Molybdenum around the target)
- Al-Pb beam dump. The lead of the beam dump has been realistically described as PbSb4 alloy
- the last 3 m of the electron beam-pipe
- shielding walls (the first 30-50 cm, then an infinite absorber is used) and collimator



(I) Prompt dose rate

 $I = 15 \ \mu A$



Along the axis x = -100 cm the prompt and the residual dose profiles have been evaluated

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(II) Residual dose rates

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Irradiation hypothesis: 6 weeks continous at 15 μ A Cooling times: end of irradiation (EOI), 24 h, 1 week

Ambient dose equivalent rate (µSv/h)



- At EOI we are dominated by radionuclides produced in the aluminum of the dump - After 24 h the contribution of the activated Molybdenum becomes dominant, due to the presence of the ⁹⁹Mo ($t_{1/2} = 65.9$ h)

HELMHOLTZ **Contribution of the single radionuclides to the total specific activity** in the central part of the Molybdenum tube around the liquid lead channel





Radionuclide	t _{1/2}	Production process	Decay mode	Specific activity [Bq/cm3] after 24 h cooling
⁹⁹ Mo	65.94 h	Thermal neutron activation	β-	4.4 E+3

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Contribution of the single radionuclides to the total specific activity in the PbSb4 alloy of the beam dump

Hypotheses: 6 weeks continous irradiation 24 h cooling time



Residual activity (Bq/cm3) - 24 h cooling after 6 weeks irradiation at 15 microA

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Thank you for your attention!

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Physics in the FLUKA code (in a glance)



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➤ Nucleus –nucleus interactions 100 MeV/n – 10⁵ TeV/n → New Model (BME) down to 20 MeV/n

- > Electromagnetic and μ interactions 1 keV 10⁵ TeV
- Hadron hadron and hadron nucleus interactions 0 10⁵ TeV
- Neutrino interactions
- > Charged particle transport, transport in magnetic field
- Full neutron transport above 20 MeV and multigroup transport under 20 MeV (260 energy groups)

FLUKA is able to compute, in the same simulation, the entire cascade of secondary particles, from TeV energies down to thermal neutrons

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The effect of a typical target





The "cross effect" visible for neutrons is due to the geometry of the target. Since I have used a slab with a square shape in the transversal plane (dimensions: 5 cm x 5 cm), the directions at 45° and 135° in θ with φ = 45°, 135°, 225° or 315° (*) are the directions, where the tracklength inside the target is bigger. As consequence, a secondary inelastic neutron emitted in that directions has a bigger probability to have a second interaction.

The behaviour at θ ~ 90° is due to the contribution of the ϕ components, described above (the plot is the result of the average in a region defined by z in (-75 cm, 75 cm) respect to the PNQ source).

(*) θ is the polar angle respect to the photoneutron beamline (x), ϕ the azimuthal angle in the yz plane

Contribution of the single radionuclides to the total specific activity in the PbSb4 alloy after 6 weeks irradiation and 24 h cooling

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Radionuclide	t _{1/2}	Production process	Main decay mode	Specific activity [Bq/cm3] after 24 h cooling
¹¹⁷ Sb	2.8 h	Charged particle reaction	β+	2.5 E-2
¹¹⁹ Sb	38.19 h	Charged particle reaction	e capture	1.1 E+3
¹²¹ Sn	27.06 h	Fast and thermal neutron activation	β-	0.3
¹²² Sb	2.72 d	Fast and thermal neutron activation	β·	4.7 E+4
¹²⁴ Sb	60.2 d	Fast and thermal neutron activation	β-	8.3 E+3
²⁰³ Hg	46.61 d	Fast and thermal neutron activation	β-	2.2
²⁰⁰ Pb	21.5 h	Charged particle reaction	e ⁻ capture	4.4
²⁰¹ Pb	9.33 h	Charged particle reaction	e ⁻ capture, β ⁺	10.3
²⁰³ Pb	51.87 h	Charged particle reaction and fast neutron activation [mainly (γ, n) from ²⁰⁴ Pb, (p,n) from ²⁰³ Tl]	e ⁻ capture, γ	6.5 E+3
²⁰⁹ Pb	3.25 h	Fast and thermal neutron activation [(n,γ) from ²⁰⁸ Pb]	β-	8.5
²⁰⁴ T1	3.78 y	Fast and thermal neutron activation	β ⁻	0.2
²⁰² T1	12.23 d	Charged particle reaction and fast neutron activation	e ⁻ capture, γ , β^+	32.2
²⁰¹ Tl	73.1 h	Charged particle reaction	e ⁻ capture, γ	54.7
²⁰⁰ T1	26.1 h	Charged particle reaction	e ⁻ capture, β^+ , γ	8.1
¹⁹⁹ Tl	7.42 h	Charged particle reaction	e ⁻ capture, β ⁺	0.3

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