Measurement of the neutron capture cross section for ^{157}Gd and ^{155}Gd relevant to Nuclear Technology

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Motivations

The main motivation is related to the use of **"burnable neutron poisons"** in nuclear reactors

- To increase the **efficiency** and economic performances of **reactor fuel**, it is necessary to **increase** the initial **enrichment of ^{235}U** in the fuel itself.
- However high enrichments pose severe safety problems due to the **high initial excess reactivity**.
- This can be **inherently compensated** by loading the fuel with **"burnable neutron poisons"**, i.e. isotopes with very high capture cross section, that are depleted together with the fissile isotopes.

It is very important to assess the **capture behavior of burnable poisons** in order to evaluate:

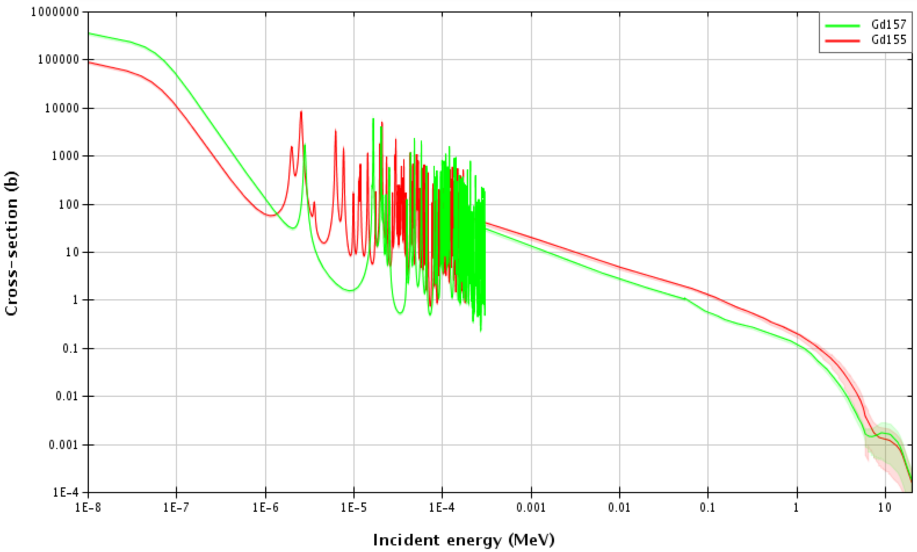
- the **economic gain due to the extension** of fuel life;
- the **residual reactivity penalty** at EndOfLife, in terms of reactor days lost (16 pins Gd-doped FAs for PWR = 5 full power days lost/year = 8 M€ for the electricity market in France);
- the **reactivity peak** for partially spent fuel for the criticality safety evaluations of Spent Fuel Pools.

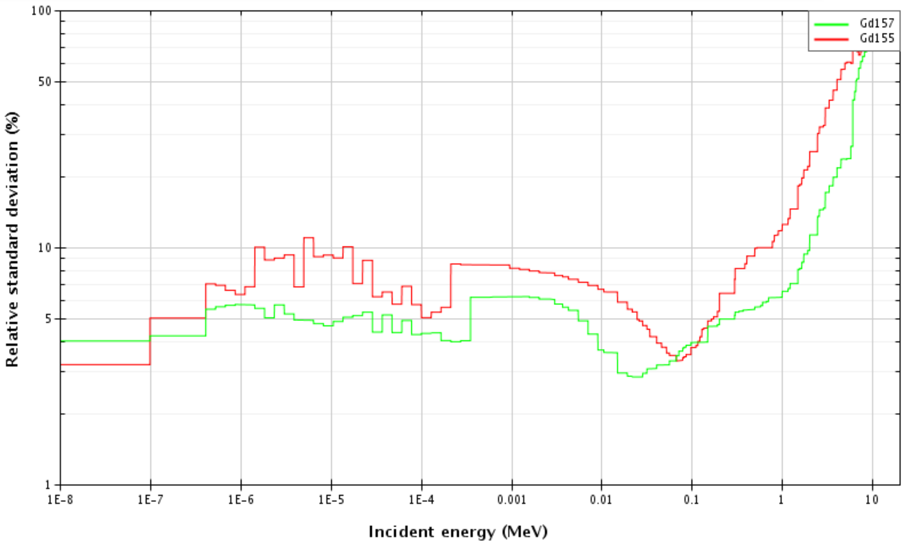
Gd in Gen. II and Gen. III Reactors

Current **Gen. II and Gen. III** nuclear reactors make **extensive use of Gadolinium** as:

- **burnable neutron poison** (Gadolinia: Gd_2O_3) for **PWR, BWR, VVER** fuels
- **emergency shutdown poison** (Gadolinium nitrate, $GdNO_3$), for **CANDU.**

The reason of this choice is the **extremely high neutron capture cross sections** of the odd Gd isotopes (**^{155}Gd and ^{157}Gd**) for low energy neutrons (thermal to ≈10 eV).



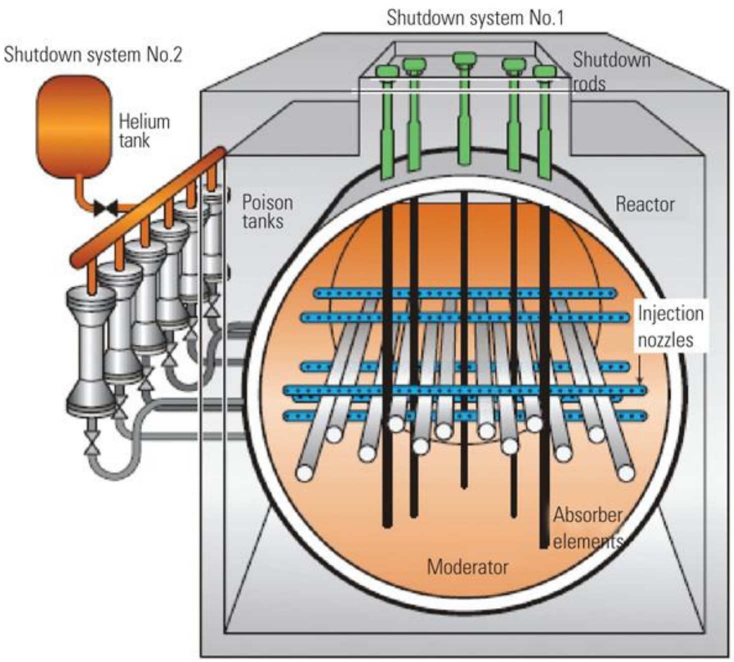


More proposals and investigations are on the way for **new reactor concepts** and for **new fuels** which involve massively **Gadolinium**.

Gadolinium in Gen. II and Gen. III Reactors

Emergency Shutdown Poison

- In CANDU reactors, **in case of severe accidents** due to or leading to criticality excursions, **Gadolinium nitrate** is injected into the moderator heavy water, to reduce (eliminate) **criticality risks** or excursions.
- However, uncertainties in the (n,γ) cross section of Gd odd isotopes may **impose special care** in the **safety calculations** for the licensing of CANDU reactors.



The $^{157}Gd(n,\gamma)$ thermal cross section

Despite their importance, the capture cross section of the odd Gd isotopes have not been extensively studied and are **not known with the accuracy required** by nuclear industry.

Reference	Year	Thermal xs (b)	Deviation from ENDF
Pattenden *2nd At. En. Conf. Geneva, 16*	1958	264000	+3.9%
Tattersall *Jour. Nucl. Ener. A 12, 32*	1960	213000	-20%
Moller *Nucl. Sci. Eng. 8, 183*	1960	254000	=
Groshev *Izv. Akad. Nauk, SSSR, 26, 1118*	1962	240000	-6%
Sun *J. Radioanal. Nucl. Chem. 256, 541*	2003	232000	-9%
Leinweber *Nucl. Sci. Eng. 154, 261*	2006	226000	-12%
Mughabghab *Evaluation (adopted in ENDF/B-VII)*	**2006**	**254000 ± 0.3%**	**=**
Choi *Nucl. Sci. Eng. 177, 219*	2014	239000	-6%

CEA Melusine/GEDEON-II results

CEA **Qualification Program** for French LWR using the **Melusine** reactor in Grenoble (2015 re-analysis based on JEFF 3.1.1 evaluations)

Isotope Concentration (Calc./Exp. - 1.)

Nature	Position	Consumption [%] [^{155}Gd]	$^{152}Gd/^{238}U$ [%]	$^{154}Gd/^{238}U$ [%]	$^{155}Gd/^{238}U$ [%]	$^{156}Gd/^{238}U$ [%]	$^{157}Gd/^{238}U$ [%]
$UO_2 + Gd_2O_3$ 5%	D07-270 [front $UO_2 + Gd_2O_3$ 5%]	26	−3 ± 5	−2 ± 1	1 ± 3	−3 ± 1	7 ± 5
	D07-100 [front $UO_2 + Gd_2O_3$ 5%]	58	−4 ± 5	−1 ± 1	−2 ± 2	−1 ± 1	2 ± 3
	G04-270 [front H_2O]	23	−11 ± 5	−1 ± 1	0 ± 6	−2 ± 1	9 ± 10
	G04-100 [front H_2O]	32	−12 ± 5	−1 ± 1	0 ± 3	−2 ± 1	8 ± 5
$UO_2 + Gd_2O_3$ 8%	D04-270 [angle H_2O]	40	−7 ± 5	−1 ± 1	4 ± 2	−3 ± 1	10 ± 2
	D04-100 [angle H_2O]	33	−8 ± 5	−1 ± 1	6 ± 1	−4 ± 1	14 ± 2
	G04-270 [front H_2O]	40	−8 ± 5	0 ± 1	1 ± 1	−1 ± 1	6 ± 2
	G04-080 [front H_2O]	34	−6 ± 5	−1 ± 1	0 ± 1	−1 ± 1	3 ± 2
	D07-270 [front $UO_2 + Gd_2O_3$ 8%]	53	−11 ± 5	−1 ± 1	3 ± 2	−3 ± 1	13 ± 3
	D07-100 [front $UO_2 + Gd_2O_3$ 8%]	46	−8 ± 5	−1 ± 1	2 ± 2	−2 ± 1	10 ± 2
	D10-270 [angle H_2O]	66	−12 ± 5	−1 ± 1	3 ± 3	−2 ± 1	13 ± 4
	D10-100 [angle H_2O]	58	−9 ± 5	0 ± 1	1 ± 3	−2 ± 1	8 ± 3
	K10-270 [angle H_2O]	87	−15 ± 6	−1 ± 1	17 ± 9	−3 ± 1	43 ± 15
	K10-100 [angle H_2O]	78	−15 ± 5	0 ± 1	1 ± 4	−1 ± 1	10 ± 6
	G10-270 [front H_2O]	88	−17 ± 6	0 ± 1	9 ± 10	−2 ± 1	24 ± 17
	G10-100 [front H_2O]	78	−18 ± 5	0 ± 1	4 ± 5	−1 ± 1	16 ± 7
	K04-270 [angle H_2O]	85	−15 ± 5	0 ± 1	7 ± 9	−1 ± 1	25 ± 16
	K04-100[angle H_2O]	75	−14 ± 5	−1 ± 1	−4 ± 6	−1 ± 1	3 ± 8
	K07-270 [front $UO_2 + Gd_2O_3$ 8%]	97	−17 ± 5	0 ± 1	56 ± 30	−1 ± 1	9 ± 40
	K07-100 [front $UO_2 + Gd_2O_3$ 8%]	89	−15 ± 5	0 ± 1	9 ± 11	−1 ± 1	24 ± 20

Large differences between calculations and experiment

CEA Melusine/GEDEON-II results

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Qualification of gadolinium burnable poison: Interpretation of MELUSINE/GEDEON-II spent fuel analysis

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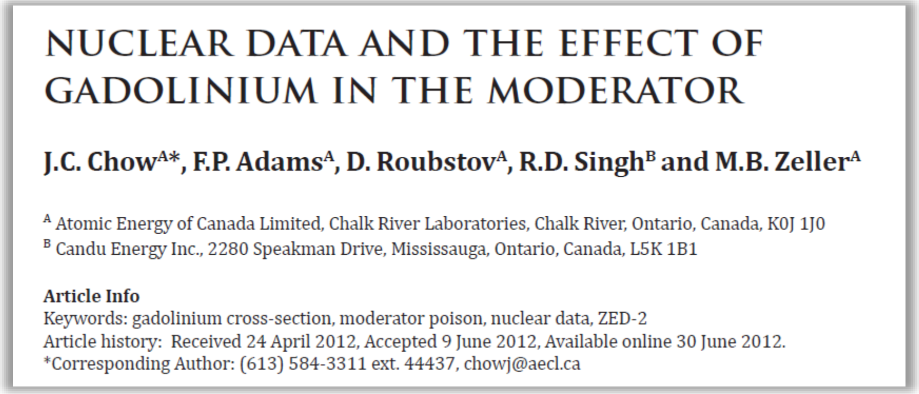
2015 !!

Table 6

REL2005-11 rings/experiment comparison for Gd consumption $C = (N^{Gdi}(0) - N^{Gdi}(BU))/N^{Gdi}(0)$.

Nature	Position	Consumption [%] [^{155}Gd]	Evol. ^{155}Gd [%]	Evol. ^{157}Gd [%]	
$UO_2 + Gd_2O_3$ 8%	D04-270 [angle H_2O]	40	−2.3 ± 2.2	−3.9 ± 2.1	Discharge 4GWd/t
	D04-100 [angle H_2O]	33	−3.7 ± 2.2	−6.3 ± 2.1	
	G04-270 [front H_2O]	40	−0.8 ± 2.3	−2.2 ± 2.2	
	G04-080 [front H_2O]	34	0.1 ± 2.5	−1.4 ± 2.4	
	D07-270 [front $UO_2 + Gd_2O_3$ 8%]	53	−1.4 ± 2.0	−3.5 ± 2.0	Discharge 7GWd/t
	D07-100 [front $UO_2 + Gd_2O_3$ 8%]	46	−0.9 ± 2.3	−3.3 ± 2.2	
	D10-270 [angle H_2O]	66	−1.1 ± 1.8	−2.2 ± 1.7	
	D10-100 [angle H_2O]	58	−0.2 ± 2.1	−1.8 ± 2.1	
	K10-270 [angle H_2O]	87	−1.9 ± 1.6	−1.3 ± 1.0	Discharge 10GWd/t
	K10-100 [angle H_2O]	78	−0.2 ± 1.5	−0.9 ± 1.5	
	G10-270 [front H_2O]	88	−1.0 ± 1.0	−0.7 ± 1.0	
	G10-100 [front H_2O]	78	−0.8 ± 1.0	−1.3 ± 1.4	
	K04-270 [angle H_2O]	85	−0.9 ± 1.0	−1.0 ± 1.0	Discharge 12GWd/t
	K04-100[angle H_2O]	75	1.1 ± 1.0	−0.4 ± 1.0	
	K07-270 [front $UO_2 + Gd_2O_3$ 8%]	97	−1.1 ± 0.5	0.0 ± 0.3	
	K07-100 [front $UO_2 + Gd_2O_3$ 8%]	89	−0.9 ± 1.0	−0.6 ± 1.0	

AECL - Chalk River results



NUCLEAR DATA AND THE EFFECT OF GADOLINIUM IN THE MODERATOR

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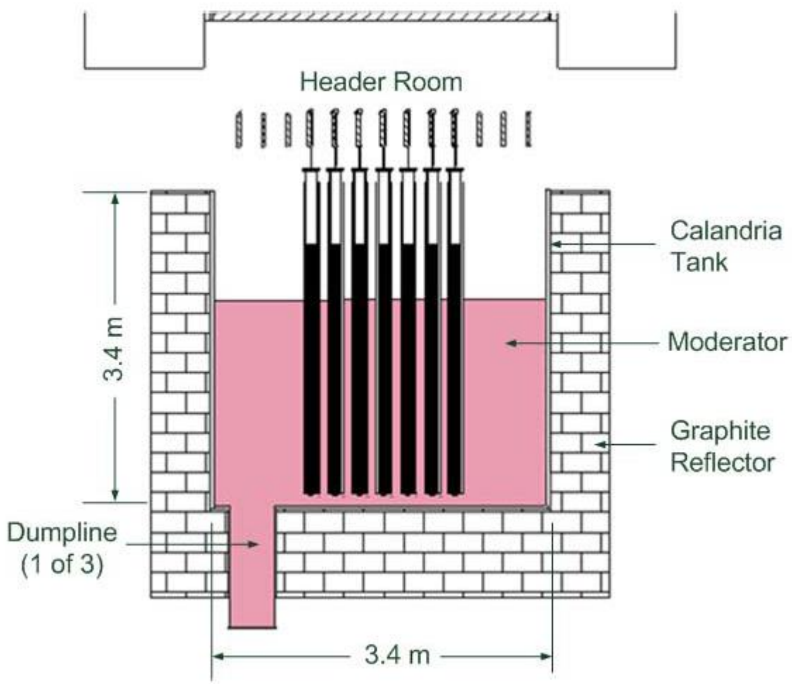
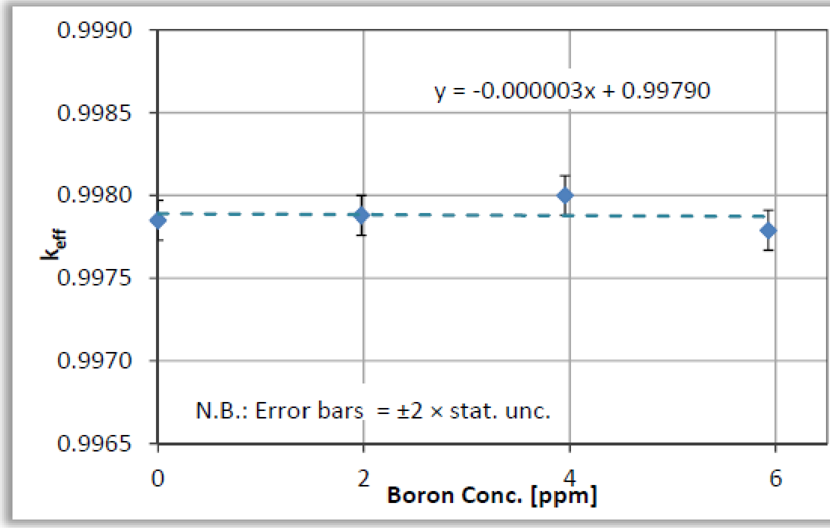
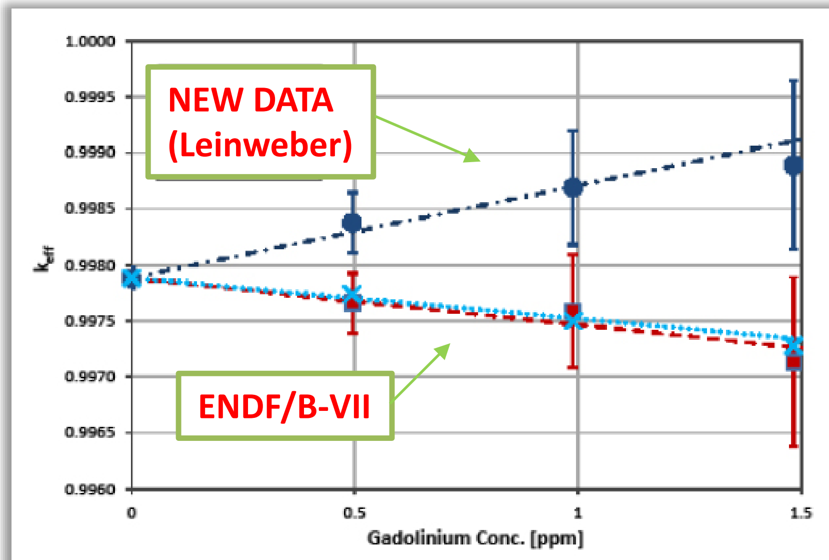






Sensitivity analysis (ENEA)

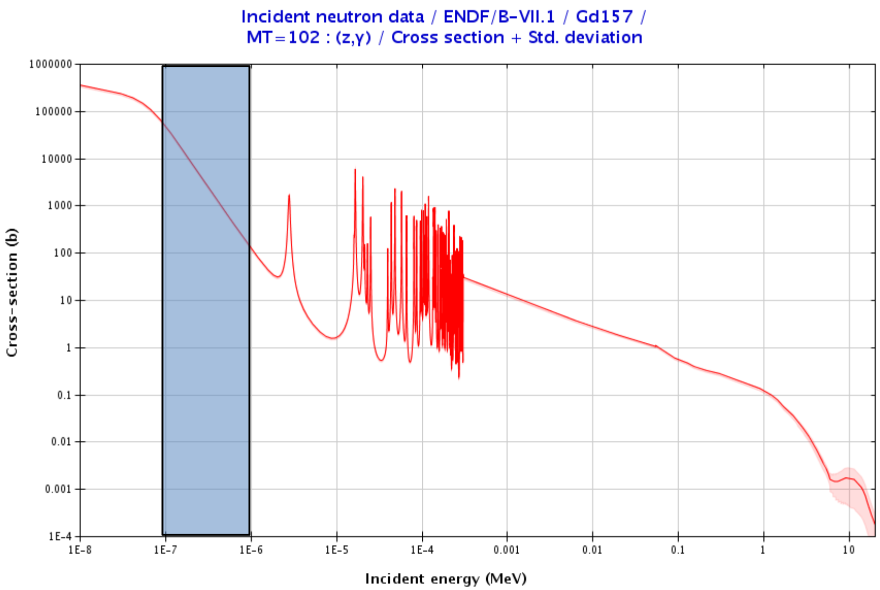
Nuclide-Reaction	Contrib. to Uncertainty in k *(% Δk/k)*	Rank
^{235}U $\bar{\nu}$ (ave. neut. mult.)	2.70E-01	1.00
^{238}U(n,γ)	1.97E-01	0.81
^{235}U(n,γ)	1.43E-01	0.64
^{235}U(n,f)	1.43E-01	0.56
^{235}U(n,f) vs ^{235}U(n,γ)	1.21E-01	0.54
^{238}U(n,n')	1.20E-01	0.51
^{235}U χ (fiss. neut. spec.)	1.13E-01	0.45
^{238}U $\bar{\nu}$	7.11E-02	0.32
^{157}Gd(n,γ)	6.03E-02	0.26
^{155}Gd(n,γ)	4.48E-02	0.20
^{92}Zr(n,γ)	4.29E-02	0.16
^{1}H(n,γ)	3.67E-02	0.14
^{91}Zr(n,γ)	3.48E-02	0.13
^{1}H(n,n)	3.13E-02	0.12
^{90}Zr(n,γ)	2.82E-02	0.10

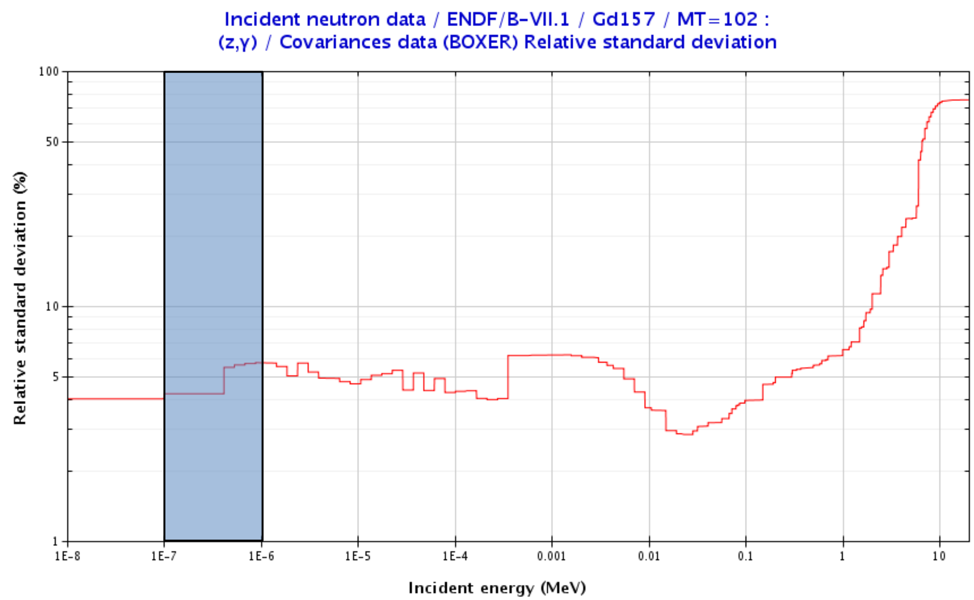
The **uncertainty** on **Gd** cross sections gives the **largest contribution** to the uncertainty on k **after** ^{235,238}**U**.

Several **cross sections** in this list have been **measured at n_TOF**.

Sensitivity analysis (ENEA)

Sensitivity analysis performed on a BWR (General Electric) with different configutions and moderator density.





The most important energy region for reactor applications is **between 100 meV and 1 eV**

Capture cross section of **^{154}Gd and ^{156}Gd** may also have an impact on the **predictions of ^{155}Gd and ^{157}Gd** (see Bernard and Santamarina, Annals of Nucl. Energy, 2015)

Gd in Neutron Capture Therapy

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A theranostic approach based on the use of a dual *boron*/Gd agent to improve the efficacy of Boron Neutron Capture Therapy in the lung cancer treatment

Diego Alberti, PhD[a], Nicoletta Protti, PhD[b,c], Antonio Toppino, PhD[d], Annamaria Deagostino, PhD[d], Stefania Lanzardo, PhD[a], Silva Bortolussi, PhD[b,c], Saverio Altieri, PhD[b,c], Claudia Voena, PhD[a,e], Roberto Chiarle, MD[a,e,f], Simonetta Geninatti Crich, PhD[a,*], Silvio Aime, PhD[a]

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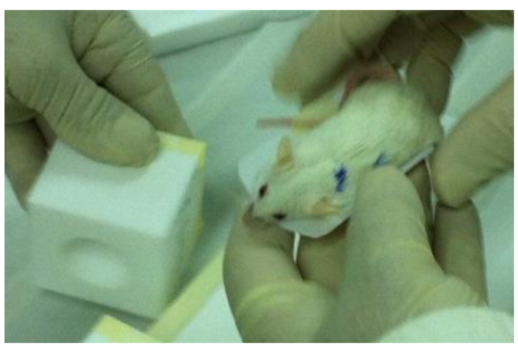
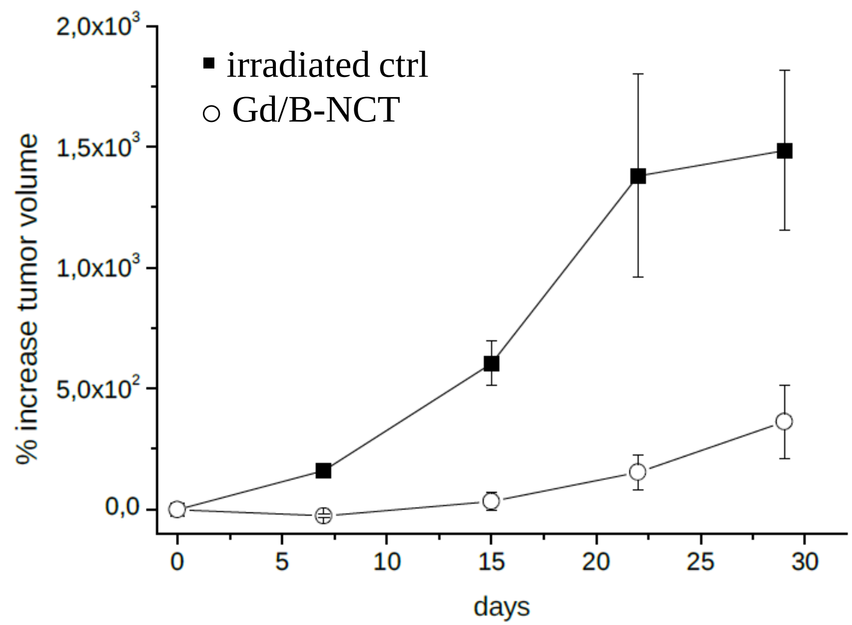
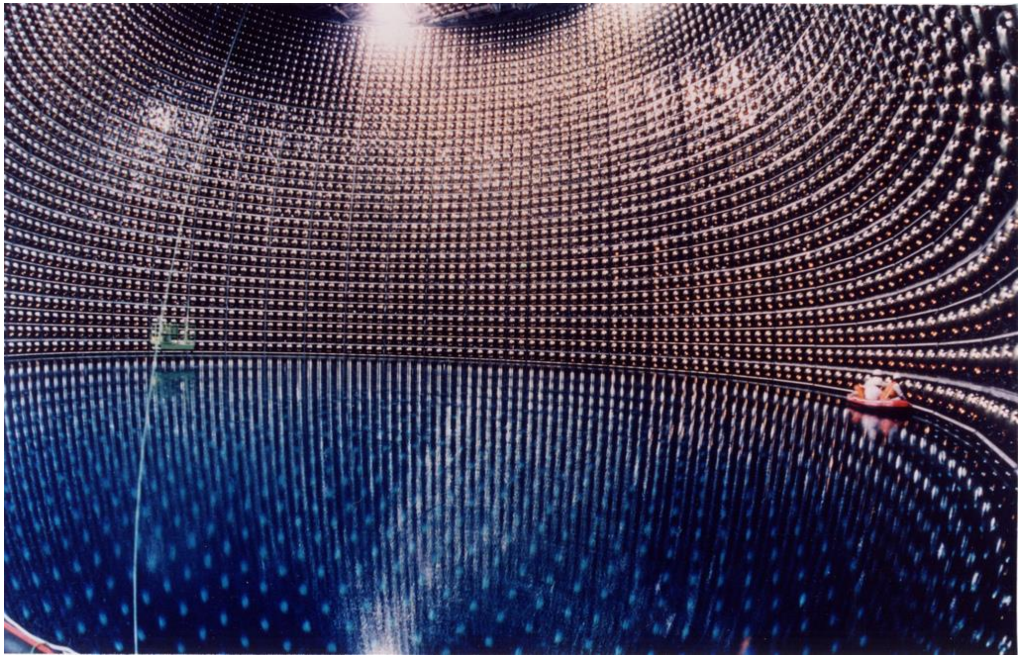
In vivo efficacy test of Gd/B/LDL-mediated BNCT on primary breast cancer lung metastases in BALB/C mice





N. Protti: *"Evaluation of the synergy effect of combined ^{10}B+^{157}Gd NCT"*

Gd in Neutrino detection



Super Kamiokande
50000 tons of pure water with more than 11000 PMT

A frontier development on **neutrino** detectors is related to **neutron** detection

$$\bar{\nu} + p \rightarrow \mu^{+} + n$$

An antineutrino can be distinguished from the neutrino counterpart without magnetizing the whole detector, by detecting a neutron.

To detect the produced neutron one can use a very tiny amount of Gd diluted in water.

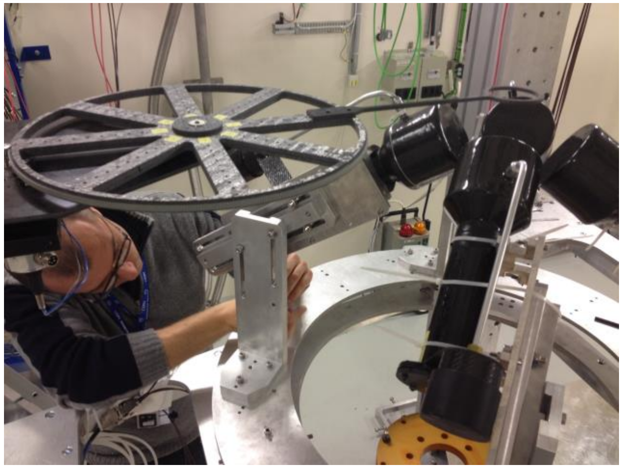
Need to accurately know neutron capture cross section of Gd.

The n_TOF measurement

We aim at measuring the ^{155,157}Gd cross section from thermal to 1 MeV with 2% uncertainty

Pro's of n_TOF (EAR1)

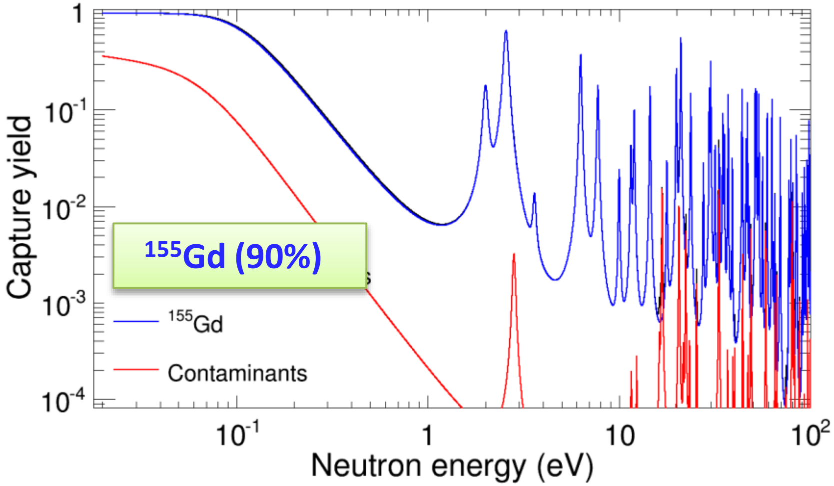
- Wide **energy range** *(from thermal to 1 MeV)*
- Very good **energy resolution**
- **High-performance detectors** and DAQ system
- Well established **analysis technique** *(PHWT)*
- **Well characterized** neutron beam *(flux, resolution function, beam profile, etc…)*



F. Mastinu et al., *"New C_6D_6 detectors: reduced neutron sensitivity & improved safety"*, CERN-n_TOF-PUB-2013-002

Requirements (samples)

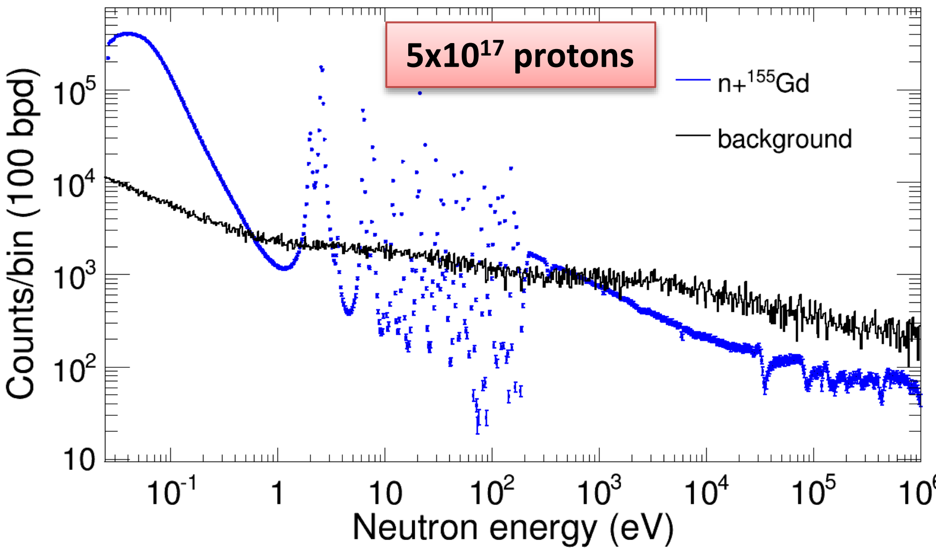
- **Highly enriched** samples (~90%)
- **Different thickness** (for high and low cross sections)
- **Well characterized** (mass, omogeneity and impurities)



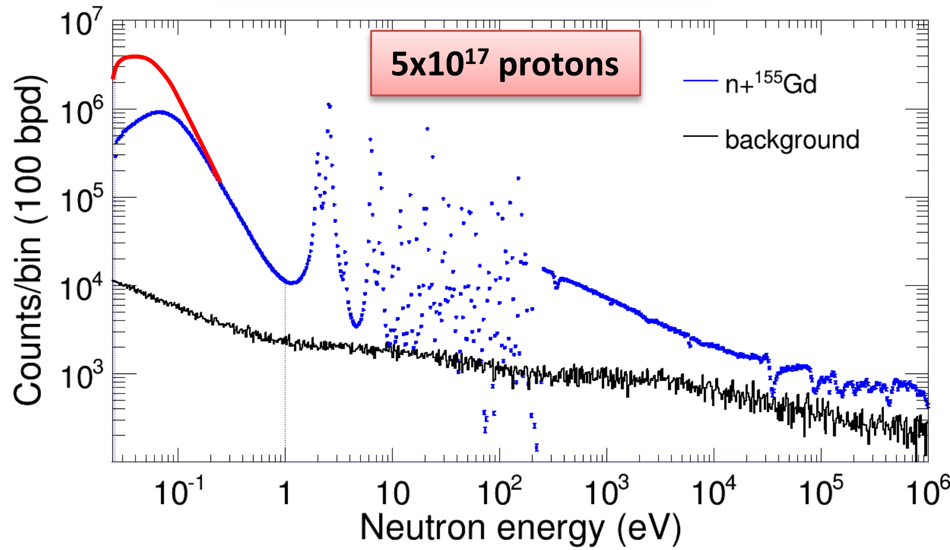
Expected count rate: ^{155}Gd

Two sample, one of which **very thin**, are needed to measure the cross section in the whole energy region **from thermal to 1 MeV** (the cross section **drops by more than two orders of magnitude** from thermal to 1 eV).

Thin sample
10 mg (1.2×10^{-5} at/b)



Thick sample
100 mg (1.2×10^{-4} at/b)

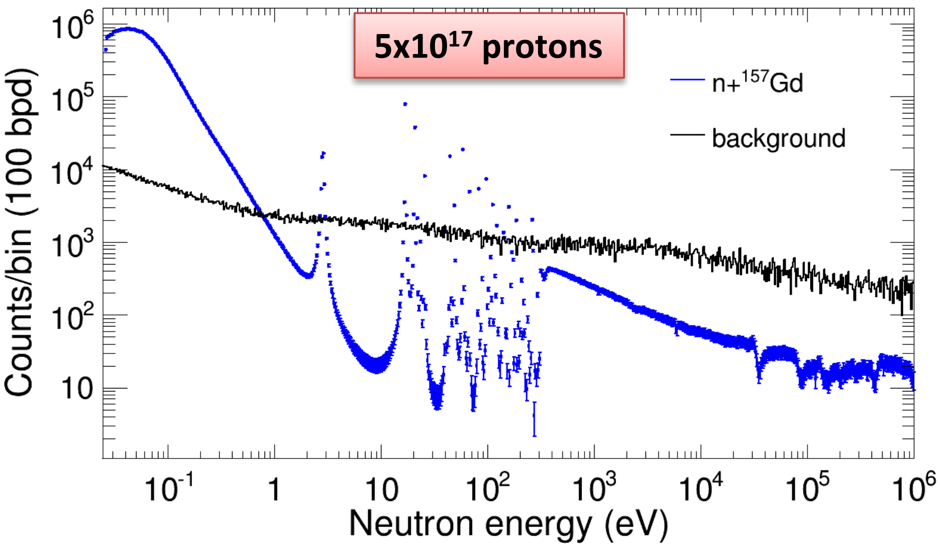


5×10^{17} protons per sample + 4×10^{17} for background ~ **1.4×10^{18} protons**

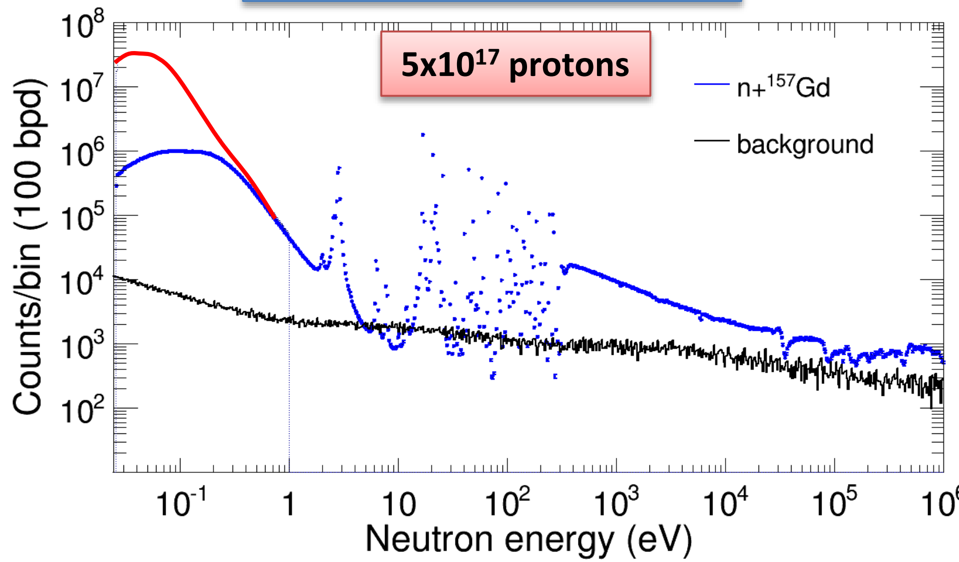
Expected count rate: ^{157}Gd

Two sample, one of which **very thin**, are needed to measure the cross section in the whole energy region **from thermal to 1 MeV** (the cross section **drops by more than two orders of magnitude** from thermal to 1 eV).

Thin sample
5 mg (6.1×10^{-6} at/b)



Thick sample
200 mg (2.4×10^{-4} at/b)



5×10^{17} protons per sample (use background from ^{155}Gd)~ **1.×10^{18} protons**

Summary

- The use of Gd as **burnable neutron poison** or **emergency shutdown poison** is becoming very attractive to **increase efficiency** and economic performance of current reactors. It may also be useful in future generation systems

- **HOWEVER**, the **uncertaint**y on the capture cross section of the two odd isotopes is still **too high** (~10%), and an intense experimental activity is ongoing to solve this problem.

- At **n_TOF** we have the **right facility, experimental setup, expertise** to solve this problem.

- **Samples** will be **provided by ORNL**, and will be **characterized by the PSI** participating group.

- We aim at measuring the capture cross section **from thermal to 1 MeV** with **2% accuracy**, thus solving this problem.

- The total number of **requested protons is 2.4x10^{18}**