Study on Activity Reduction in Concrete

Mahdi Bakhtiari* (a)

Leila Mokhtari Oranj(b), Nam-Suk Jung(b), Arim Lee(b), Hee-Seock Lee(a,b)

(a)Division of Advanced Nuclear Engineering (DANE), POSTECH, Korea

(b)Pohang Accelerator Laboratory (PAL), POSTECH, Korea

* bakhtiari@potech.ac.kr

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Contents

1. Introduction

2. Monte Carlo simulations

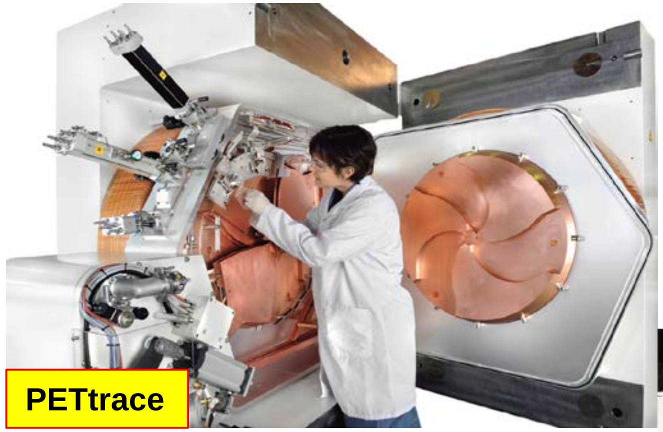
3. Results for neutron absorbing materials

4. Conclusions and remarks

1. Introduction

❑ Cyclotrons in Korea

- Totally 41 cyclotrons
- 31 cyclotrons are operating
- 20 non-self-shielded & 21 self-shielded
- Majority of cyclotrons for ^{18}F production



PETtrace

RDS Eclipse

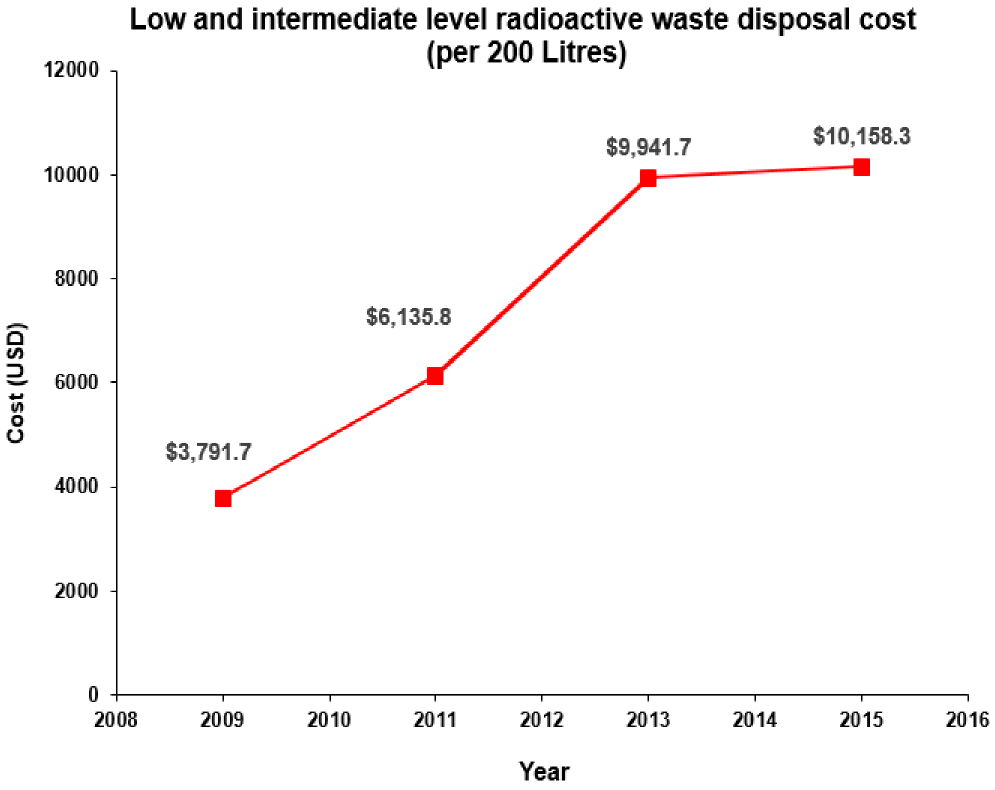
Cyclotron model name	Number
Cyclone 18/9	6
Cyclone 30	1
CYPRIS HM-7/12S	5
KIRAMS-13/30	8
MC50	1
MINItrace	1
PETtrace	11
RDS 111	1
RDS Eclipse	7
Total	41

❑ Production amount of radioactive waste

Radioactive Waste Generation during Operation [kg/month]	Shielding wall thickness [m]	
	Self-Shielded	Non-self-shielded
0 ~ 32	0.25 ~ 1	1.5 ~ 1.8

❑ Costs of the radioactive waste

- Radioactive waste cost has almost tripled between 2009 and 2015
- Practical measured must be taken to reduce the amount of wastes



❑ Major radionuclides in concrete

- During cyclotron operation, secondary neutrons from (p, n) reaction can activate the concrete
- A huge amount of radioactive waste is generated → increasing the cost of management
- This radioactive waste should be reduced for decommissioning phase

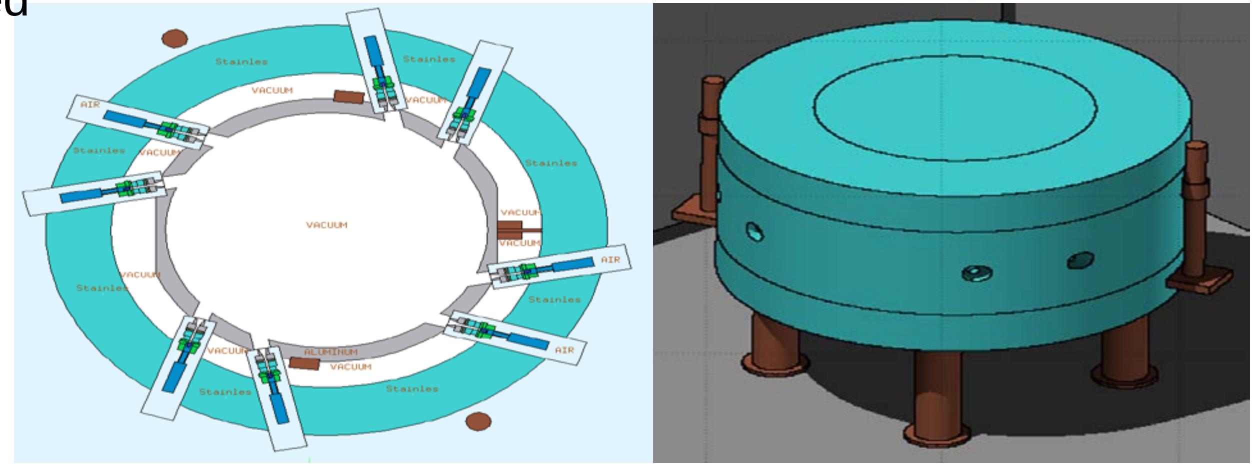
Radionuclide	Reaction	Half-life	Thermal neutron CX [barn]	Abundance [%]	Clearance Level [Bq/g]
Sc-46	**$^{45}Sc(n,\gamma)^{46}Sc$**	**83.79 d**	**26.5**	**100**	**0.1**
Mn-54	$^{55}Mn(n,2n)^{54}Mn$	312.20 d	0.785	100	0.1
	$^{54}Fe(n,p)^{54}Mn$		0.386	5.84	
Fe-59	$^{58}Fe(n,\gamma)^{59}Fe$	44.495 d	1.15	0.31	1
Co-60	**$^{59}Co(n,\gamma)^{60}Co$**	**5.271 y**	**37**	**100**	**0.1**
Zn-65	$^{64}Zn(n,\gamma)^{65}Zn$	243.93 d	0.78	48.89	0.1
Cs-134	**$^{133}Cs(n,\gamma)^{134}Cs$**	**2.0652 y**	**29**	**100**	**0.1**
Eu-152	**$^{151}Eu(n,\gamma)^{152}Eu$**	**13.517 y**	**5900**	**47.7**	**0.1**
Eu-154	$^{153}Eu(n,\gamma)^{154}Eu$	8.601 y	390	52.23	0.1

2. Monte Carlo simulations

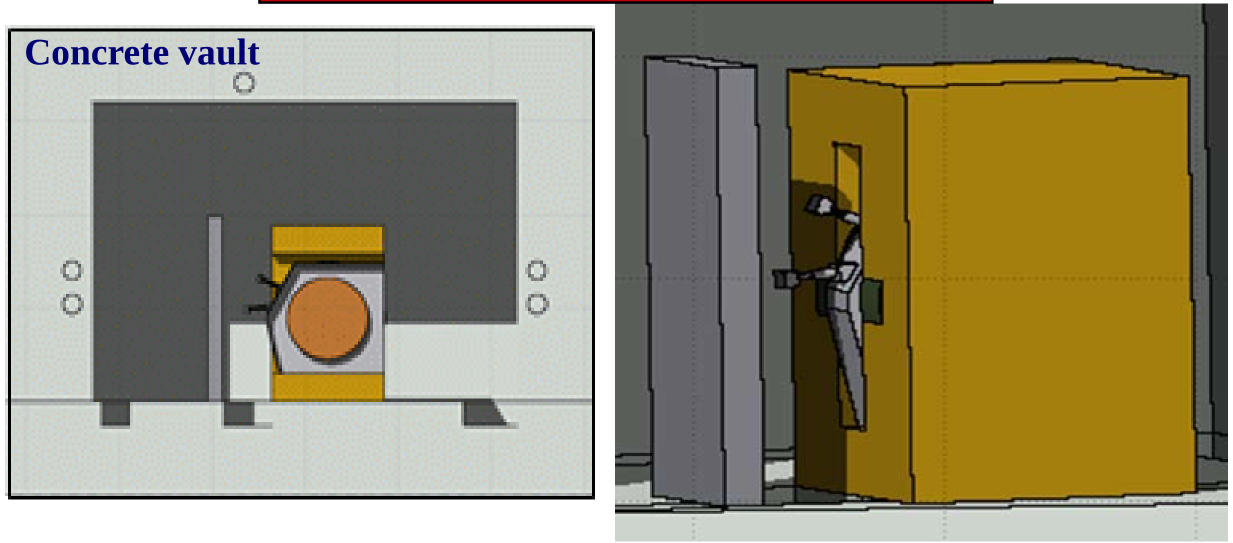
❑ FLUKA [1]

- A detailed geometry of different cyclotrons were simulated
- Neutron distributions inside the vault were estimated

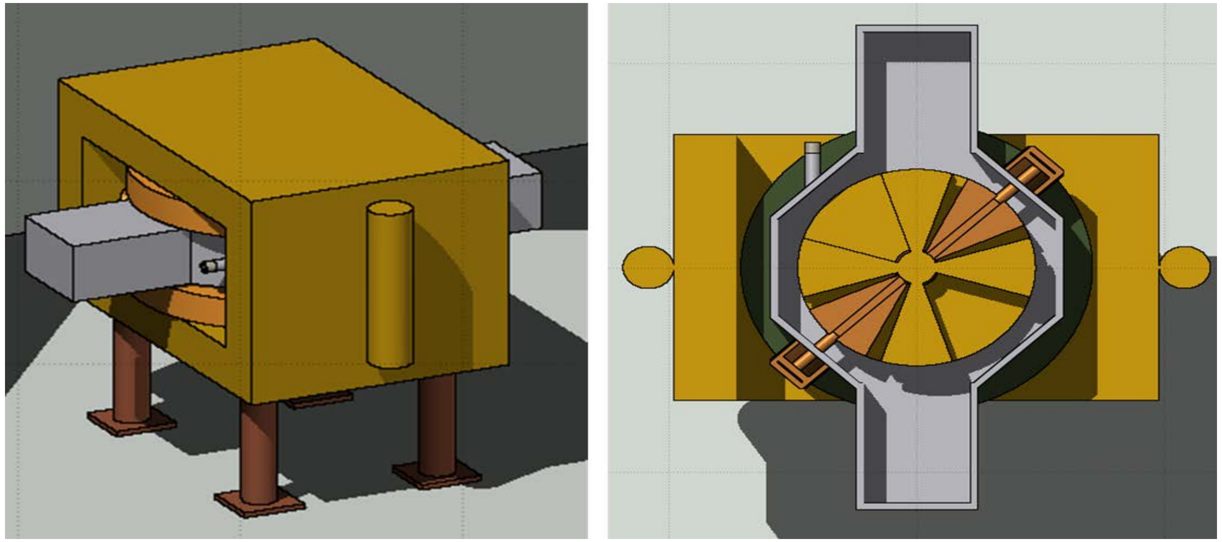
(IBA) Cyclone 18/9-Non-Self-Shielded



(GE) PETtrace Non-Self-Shielded type



KIRAMS-1318/9-Non-Self-Shielded

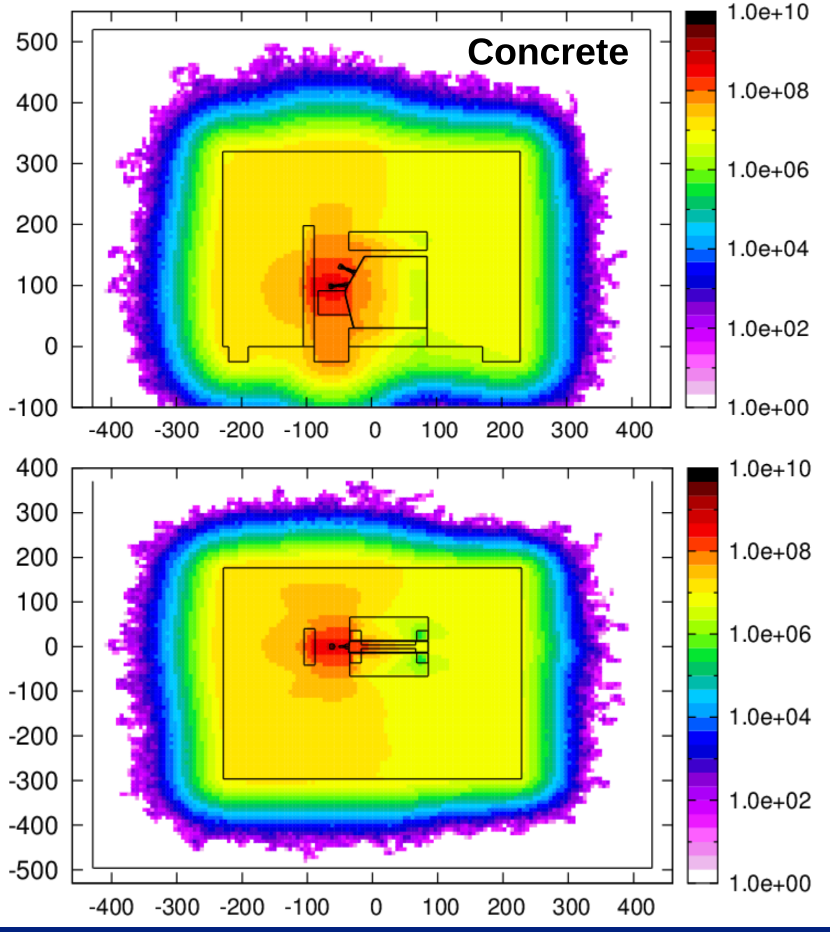


[1] A. Ferrari, et al, "FLUKA: a multi-particle transport code", CERN 2005-10 (2005), INFN/TC_05/11, SLAC-R-773.

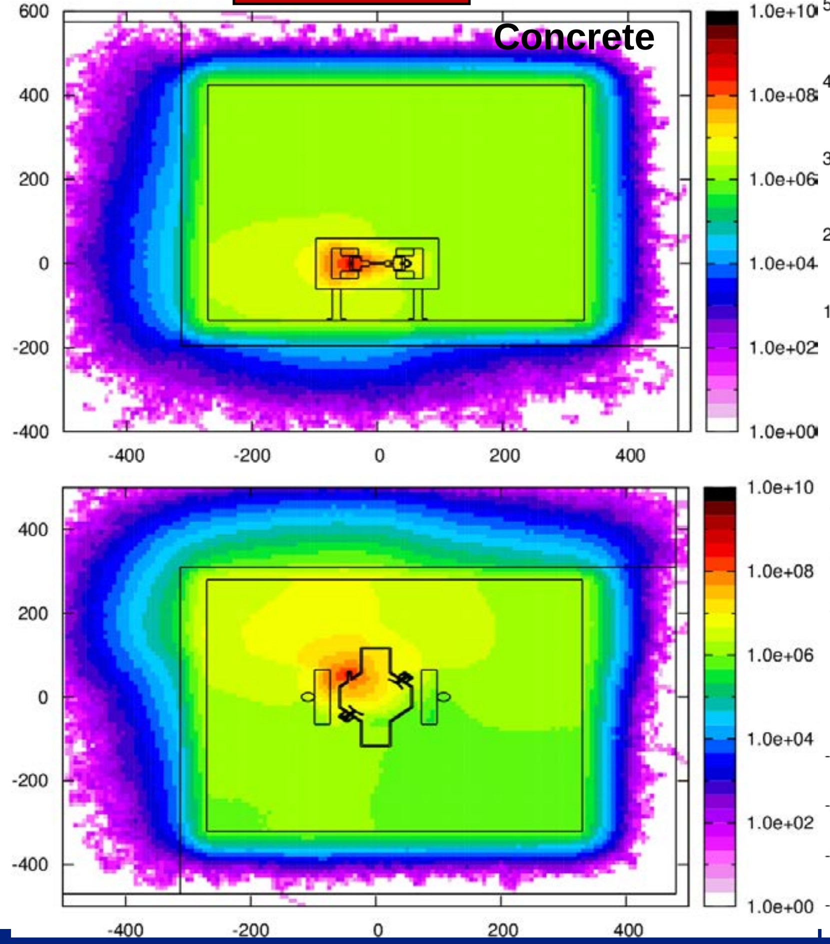
FLUKA

- Neutron flux distribution in different cyclotrons vault
- Walls of cyclotron rooms are expected to be radioactive wastes
- Concrete activation is more sever in non-self-shielded cyclotrons

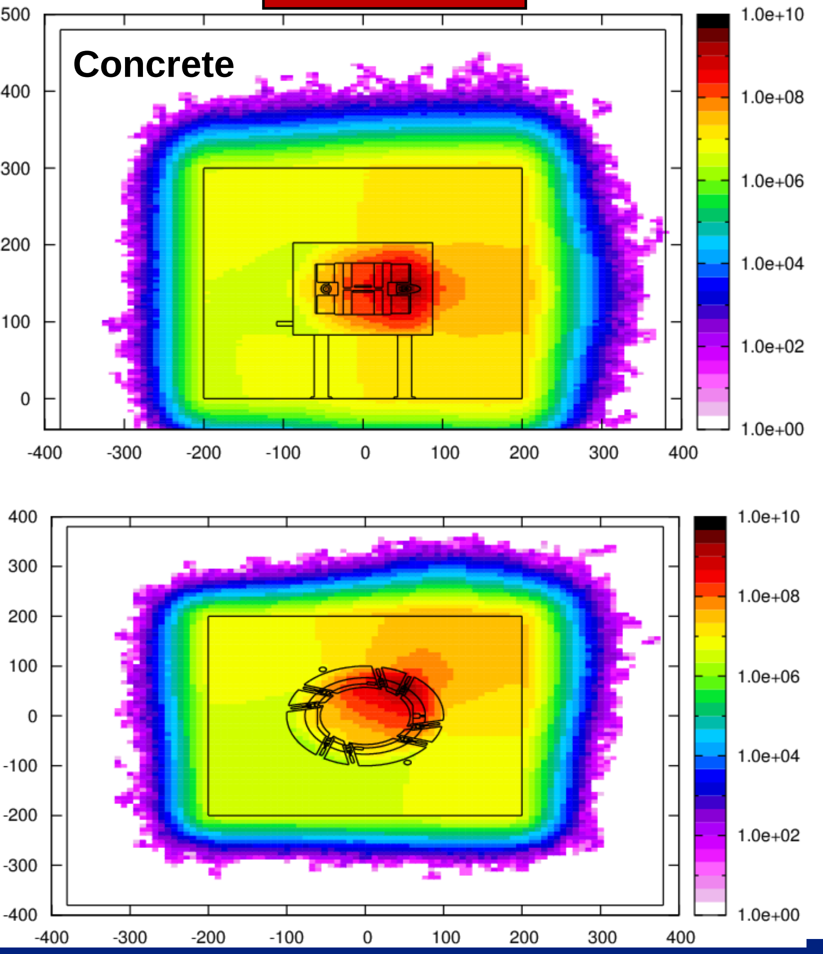
PETtrace



KIRAMS-13



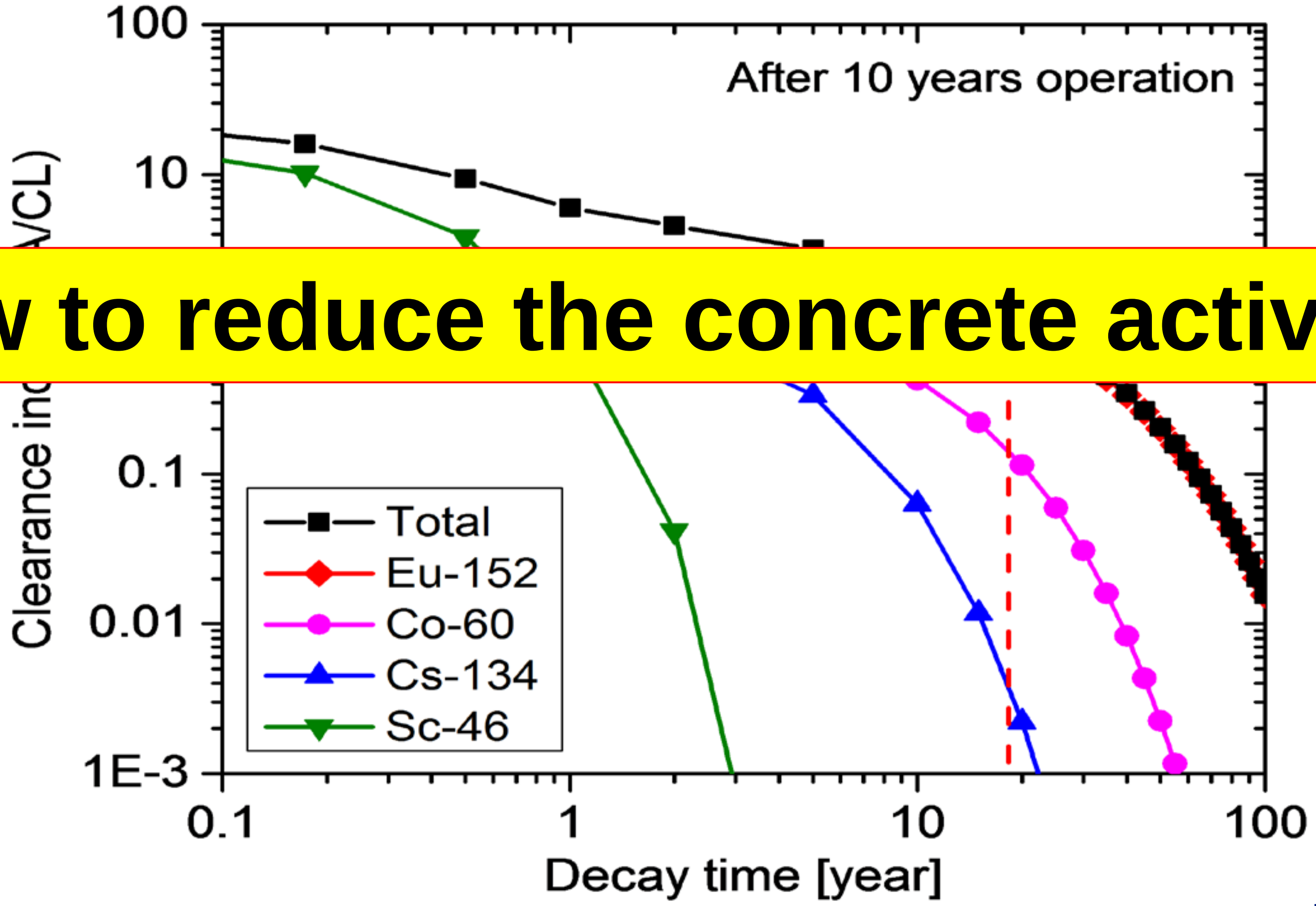
Cyclone18/9



Monte Carlo simulations

❑ Major radionuclides in concrete

- Eu-152 and Co-60 are the major radionuclides
- Eu-152 clearance index is higher than 1 by a factor of ~3 after shut down
- Around 20 years after shut down, Eu-152 clearance index is 1



How to reduce the concrete activation?

Monte Carlo simulations

- **PHITS-3.02 [1]**

  - **For saving the time, a simple geometry of PETtrace cyclotron was simulated in PHITS**
  - **INCL4.6 [2] followed by GEM [3] were applied for proton-induced reactions on $H_2{}^{18}O$**
  - **JENDL-4.0 [3] cross sections library was used for neutron interactions**
  - **An optimum neutron absorbing material was determined**

[1] T. Sato et al, Features of Particle and Heavy Ion Transport code System (PHITS) version 3.02, J. Nucl. Sci. Technol. 55, (2018)
[2] A. Boudrad, et al, New potentialities of the lieège intranuclear cascade model for reactions induced by nucleons and light charged particles, Phys. Rev. C 87, (2013)
[3] S. Furihata, Statistical analysis of light fragment production from medium energy proton-induced reactions, Nucl. Instrum. Meth. B171, (2000)

Monte Carlo simulations

❑ Concrete composition

- Several types of concrete have been analyzed [1]
- PAL concrete (2017) is used in the simulation

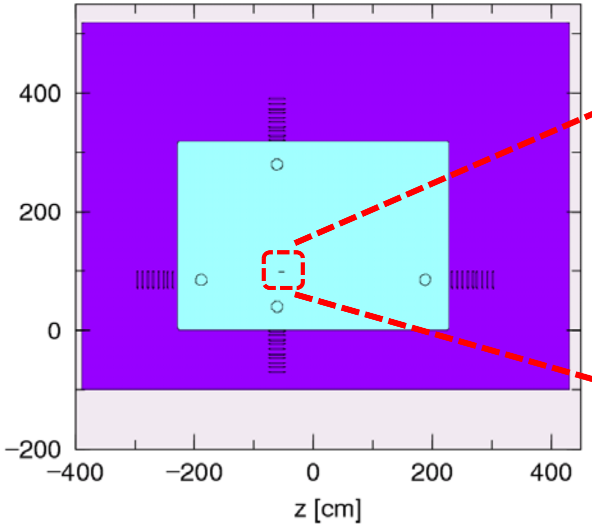
Concrete composition

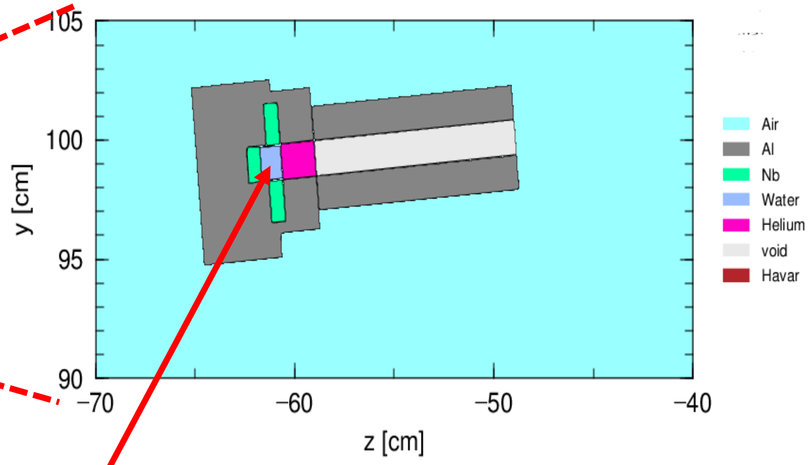
	18 Mpa	21 Mpa	27 Mpa	27 Mpa		2017	2018	2016		
원소	Conc. 1	Conc. 2	Conc. 3	Conc. 4	KOMAC Conc.	PAL Conc.	IBS Conc.	PAL Conc.		
	wt%									
O	#########	43.2462	45.6507	45.7017	44.4873	43.1286	45.4904	43.5674	Ir	0.0761
Si	#########	29.3021	29.8340	28.9250	27.5505	25.6873	28.9840	26.7925	Ho	0.0689
Al	6.867994	6.3052	6.2499	6.8007	7.1568	5.6702	7.0504	5.6657	Er	0.0547
Ca	6.139519	8.9241	8.4203	6.4807	10.8648	13.5019	7.5489	9.4130	Pt	0.0462
Fe	3.175370	1.5846	2.0201	3.0730	2.3268	2.3696	2.3042	3.5867	Re	0.0260
Na	2.374914	4.2661	1.9034	2.0741	1.8672	1.7181	2.2220	2.0027	U	0.0255
Mg	1.627494	1.3932	1.0979	1.5270	1.5130	1.6448	1.3626	0.8358	Mo	0.0205
K	1.542515	2.7691	2.0219	1.8048	2.0928	1.8744	2.5875	6.1570	Os	0.0164
C	1.304503	0.9755	1.1254	1.7732	0.5879	2.4771	0.7374	0.2697	Br	0.0090
H	0.635594	0.5785	0.9153	0.9497	0.7751	0.9942	0.5640	0.1268	Se	0.0058
Ti	0.345638	0.2065	0.2606	0.3473	0.3208	0.3111	0.2729	0.2102	Ge	0.0031
S	0.165519	0.2103	0.2218	0.2029	0.1580	0.2991	0.1306	0.1621		
Mn	0.071807	0.0516	0.0599	0.0691	0.0695	0.0755	0.0367	0.0721		
P	0.064210	0.0382	0.0428	0.0645	0.0525	0.0661	0.0539	0.0560		
Ba	0.048562	0.0464	0.0523	0.0476	0.0639	0.0457	0.0802			
N	0.037413	0.0155	0.0182	0.0384	0.0034	0.0154	0.2311			
Sr	0.030093	0.0177	0.0236	0.0288	0.0322	0.0231	0.0340	0.0217		
Zn	0.029954	0.0181	0.0275	0.0300	0.0261	0.0479	0.0320	0.0282		
Zr	0.015448	0.0160	0.0148	0.0152	0.0143	0.0116	0.0164	0.0150		
V	0.007750	0.0032	0.0046	0.0071	0.0055	0.0045	0.0043			
Rb	0.006185	0.0078	0.0072	0.0070	0.0056	0.0063	0.0105	0.0132		
Cr	0.005183	0.0014	0.0022	0.0045	0.0021	0.0026	0.0030	0.0026		
Li	0.004848	0.0031	0.0034	0.0045	0.0027	0.0029	0.0027	0.0020		
Ce	0.004773	0.0052	0.0044	0.0049	0.0039	0.0035	0.0044			
Ni	0.003981	0.0017	0.0027	0.0038	0.0028	0.0038	0.0023	0.0035		
Ga	0.003311	0.0031	0.0033	0.0032	0.0036	0.0027	0.0051	0.0134		
Co	0.003273	0.0013	0.0010	0.0013	0.0009	0.0009	########	0.0437		
Cu	0.002567	0.0013	0.0038	0.0027	0.0049	0.0048	0.0735	0.0430		
Pb	0.001889	0.0014	0.0023	0.0016	0.0011	0.0025	0.0018	0.0829		
Y	0.001383	0.0016	0.0013	0.0011	0.0010	0.0008	0.0007	0.0086		
Sc	0.000961	0.0007	0.0007	0.0009	0.0008	0.0005	########			
As	0.000946	0.0012	0.0007	0.0013	0.0009	0.0011	0.0011			
Nb	0.000902	0.0009	0.0007	0.0009	0.0006	0.0007		0.0108		
Th	0.000809	0.0008	0.0008	0.0007	0.0005	0.0005		0.0296		
Cs	0.000745	0.0006	0.0004	0.0007	0.0002	0.0002	########	0.0002		
Eu	0.000113	9.15E-05	9.36E-05	1.01E-04	9.17E-05	7.00E-05	9.99E-05	6.26E-05		
W							0.1476		W	0.4116
Pd							2.76E-05			
합계	100	100	100	100	100	100	100	100		

[1] A. Lee, N. S. Jung, H. S. Lee, Composition analysis of Ordinary Concrete to Estimate Residual Isotopes in the Decommissioning of Particle Accelerator, Poster presentation, 14th Specialists' workshop on Shielding aspects of Accelerators, Targets, and Irradiation Facilities (SATIF14), Gyeongju, Korea, 2018.

❑ Geometry

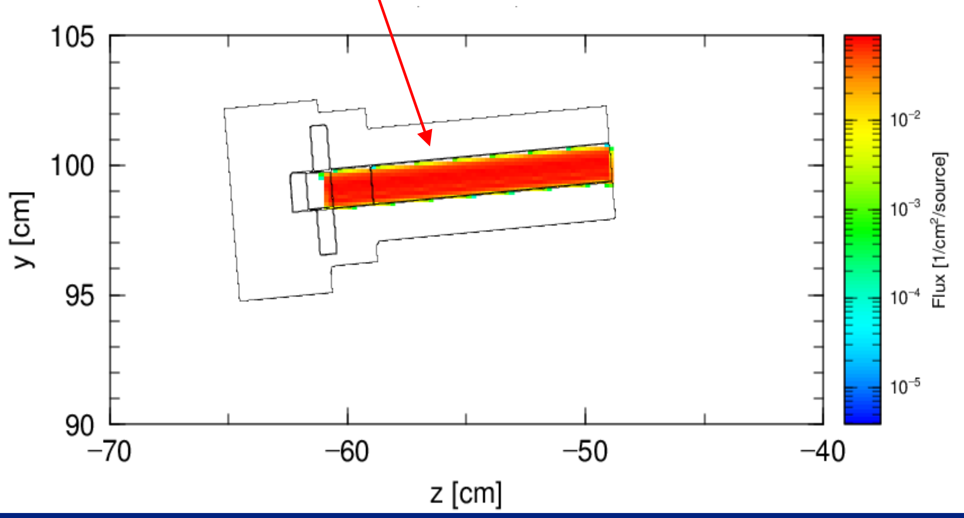
- A simplified geometry of a PETtrace cyclotron





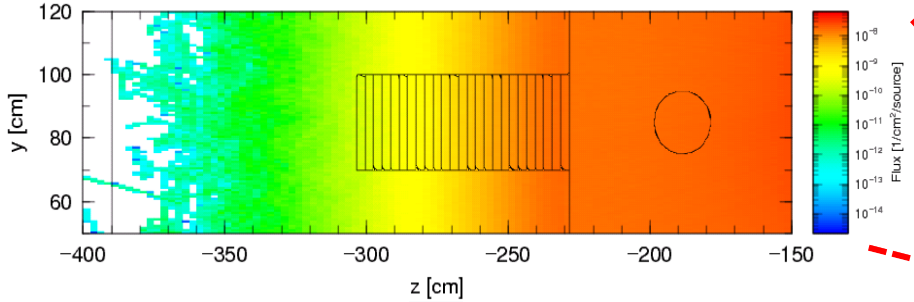
- Proton beam
- 16.5 MeV

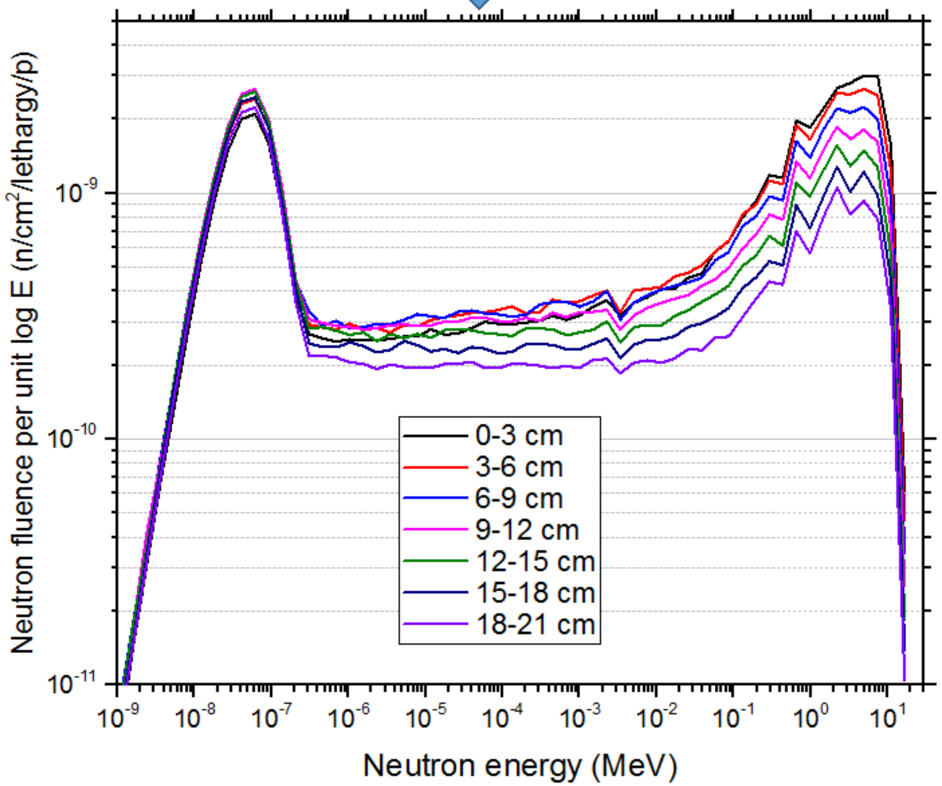
- H_2O is used as the target with enriched O-18 for F-18 production via: $^{18}O(p,n)^{18}F$ reaction

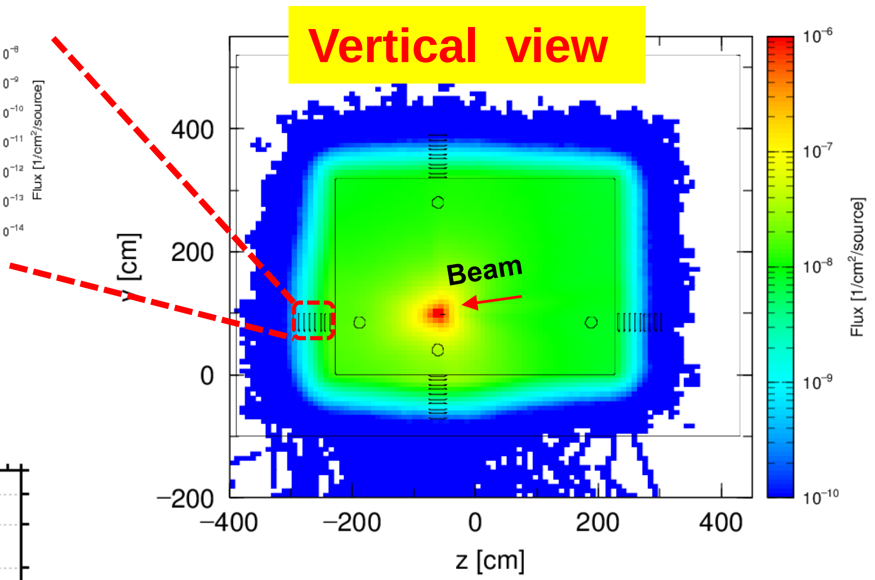


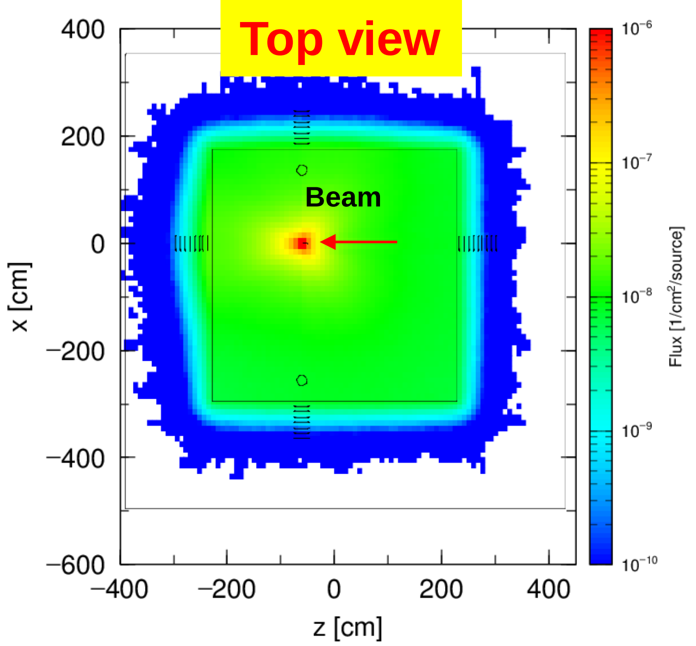
Monte Carlo simulations

- T-Track tally was activated in PHITS to score neutron fluence.
- Detector size: 30*30*3 [cm^3]
- Neutron fluence obtained at different depths in concrete





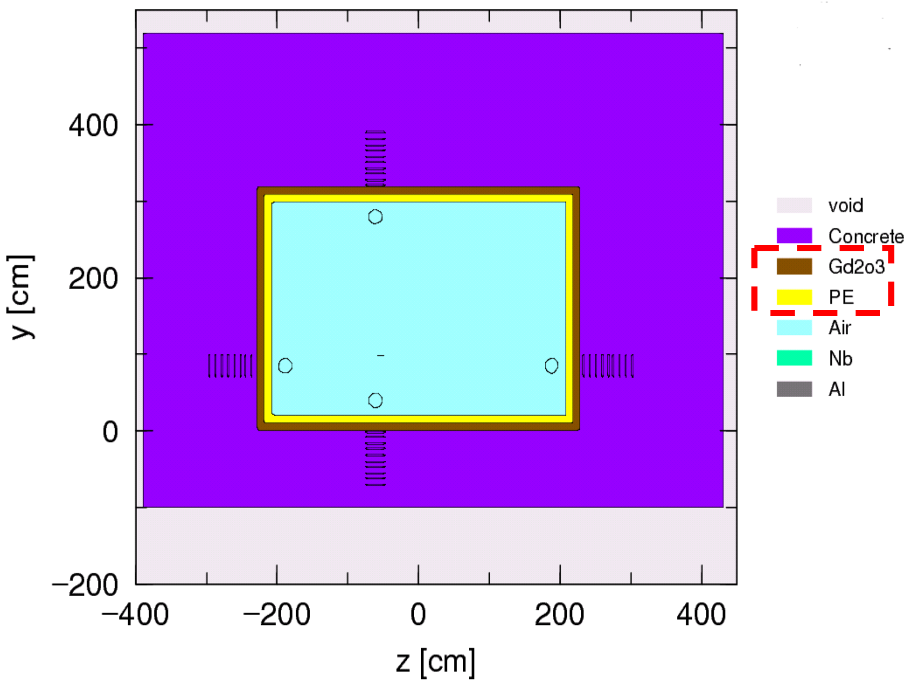


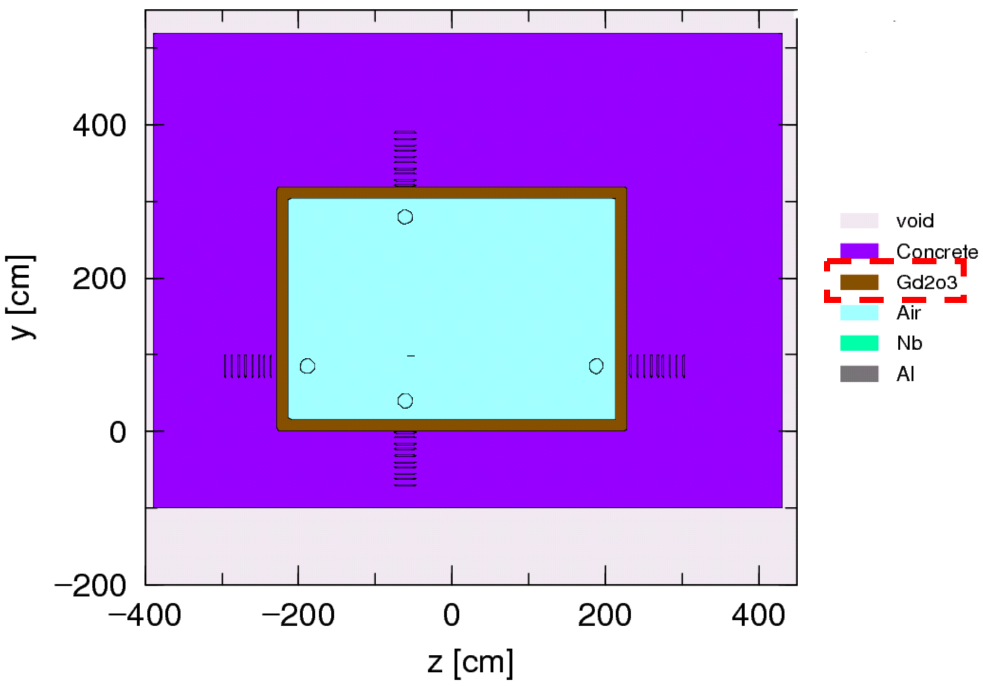


3. Results for neutron absorbing materials

- **Gd, Polyethylene (PE) and B are well-known absorbing materials**
- **All cyclotron vault walls were covered**

Material	Thickness (cm)
Gd_2O_3	5 10 15
B_4C	5 10 15
PE	5 10 15
BPE (5%wt)	5 10 15
PE+ Gd_2O_3	10+10 5+10
PE+ B_4C	10+10



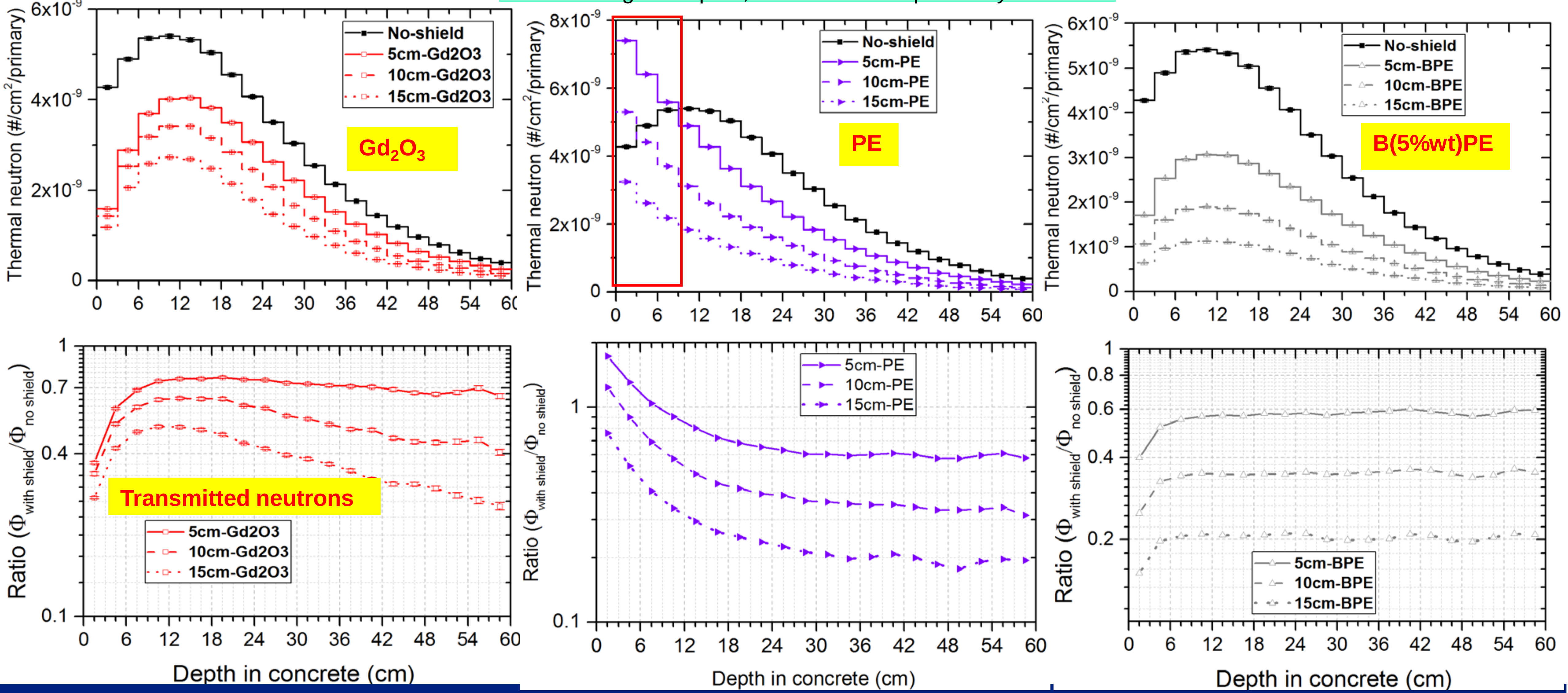


Results for neutron absorbing materials

- Reduction of thermal neutrons using Gd

- PE thermalizes fast neutrons
- At concrete surface, thermal neutrons increase
- In the higher depths, neutrons are captured by concrete

- Neutrons thermalized in PE are captured by ^{10}B

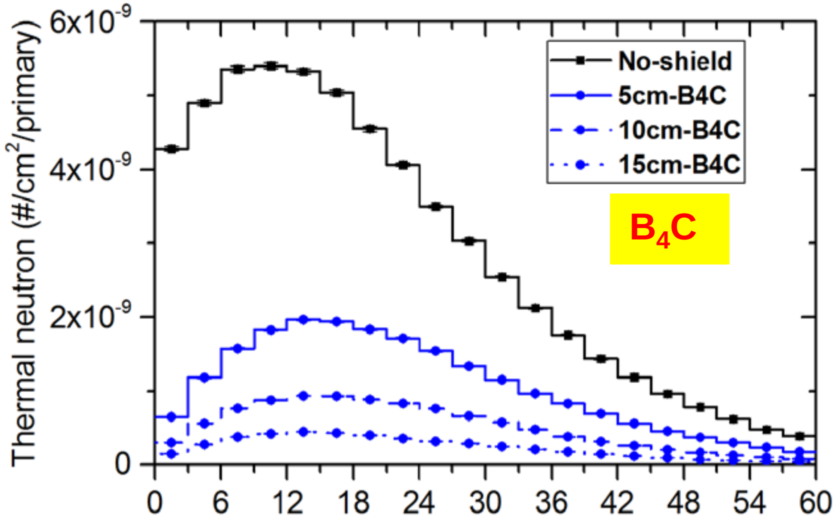


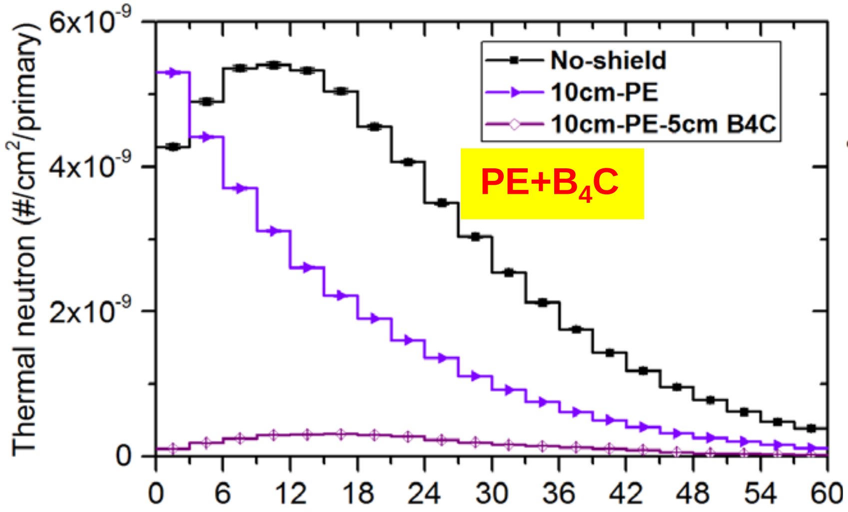
Results for neutron absorbing materials

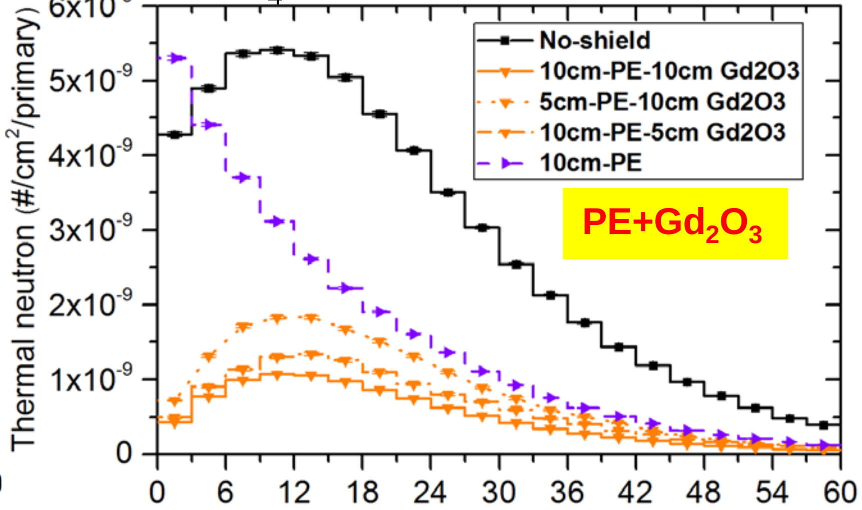
- B_4C can reduce thermal neurons dramatically.

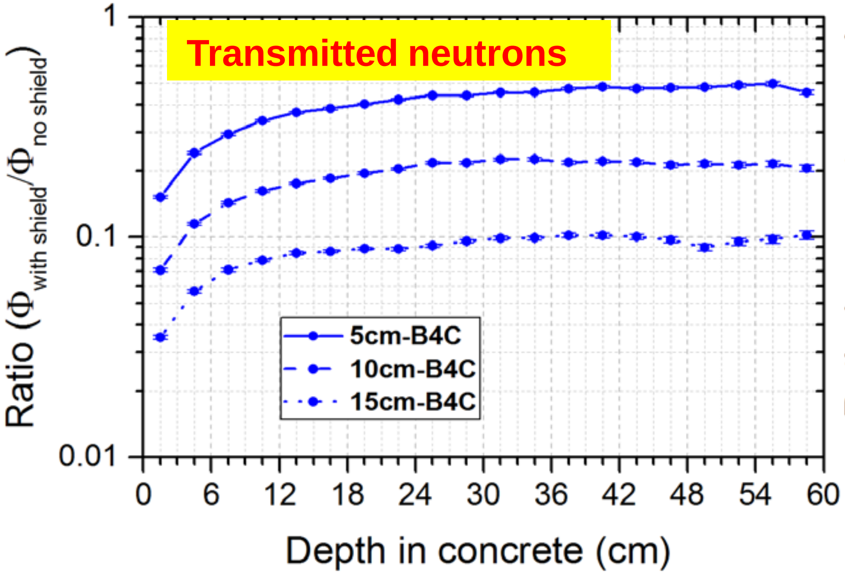
- Placing PE before B_4C can help to remove the majority of thermal neutrons

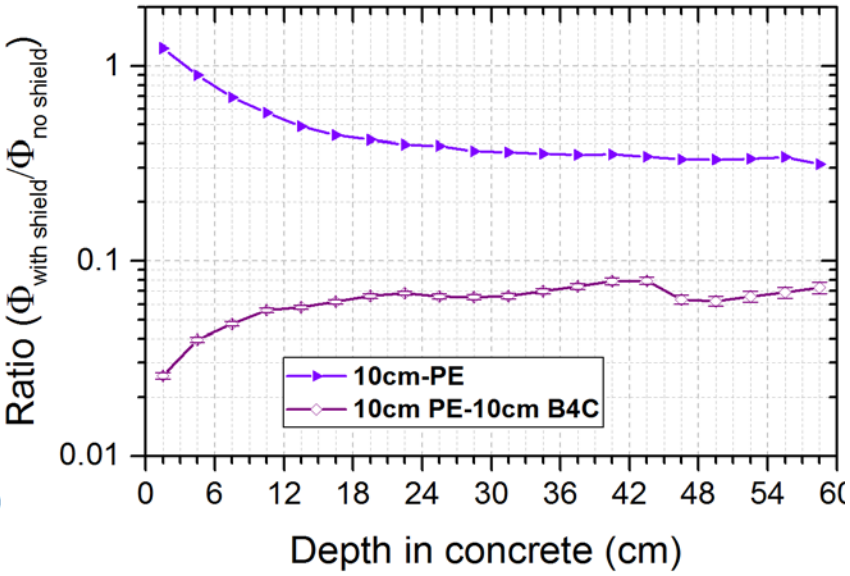
- Placing PE before Gd_2O_3 can help to remove thermal neutrons but less than B_4C

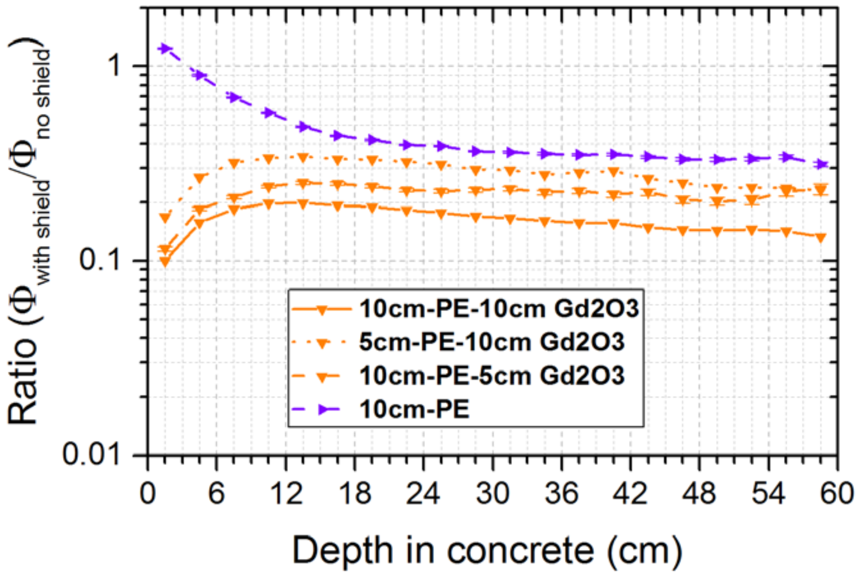








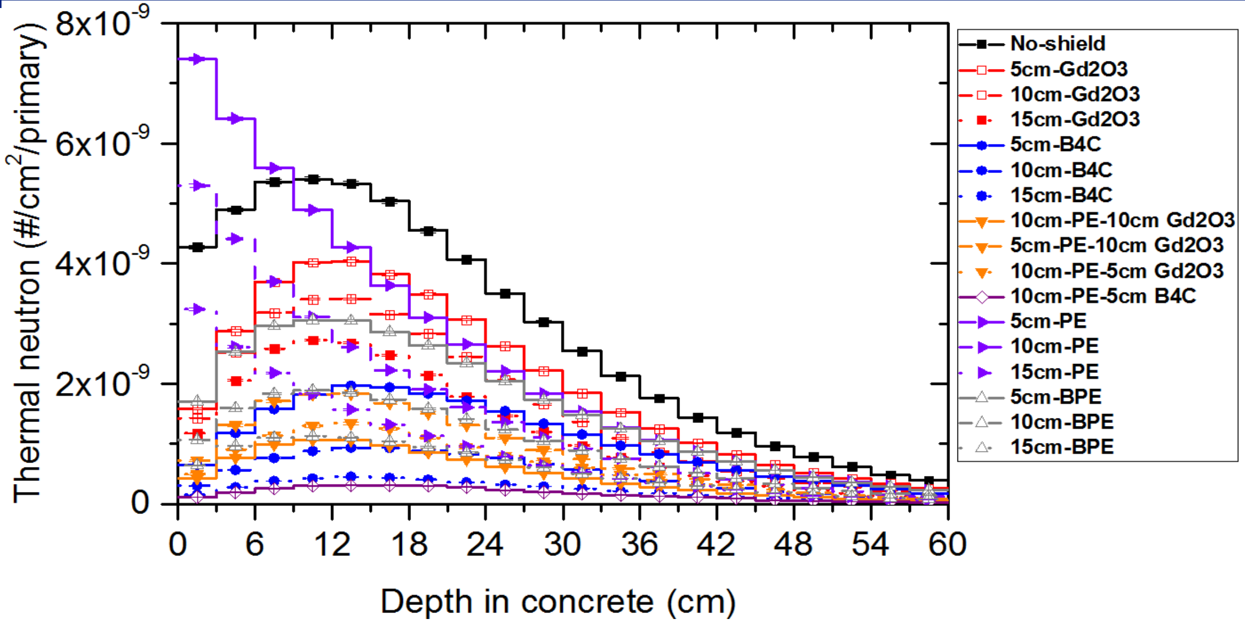


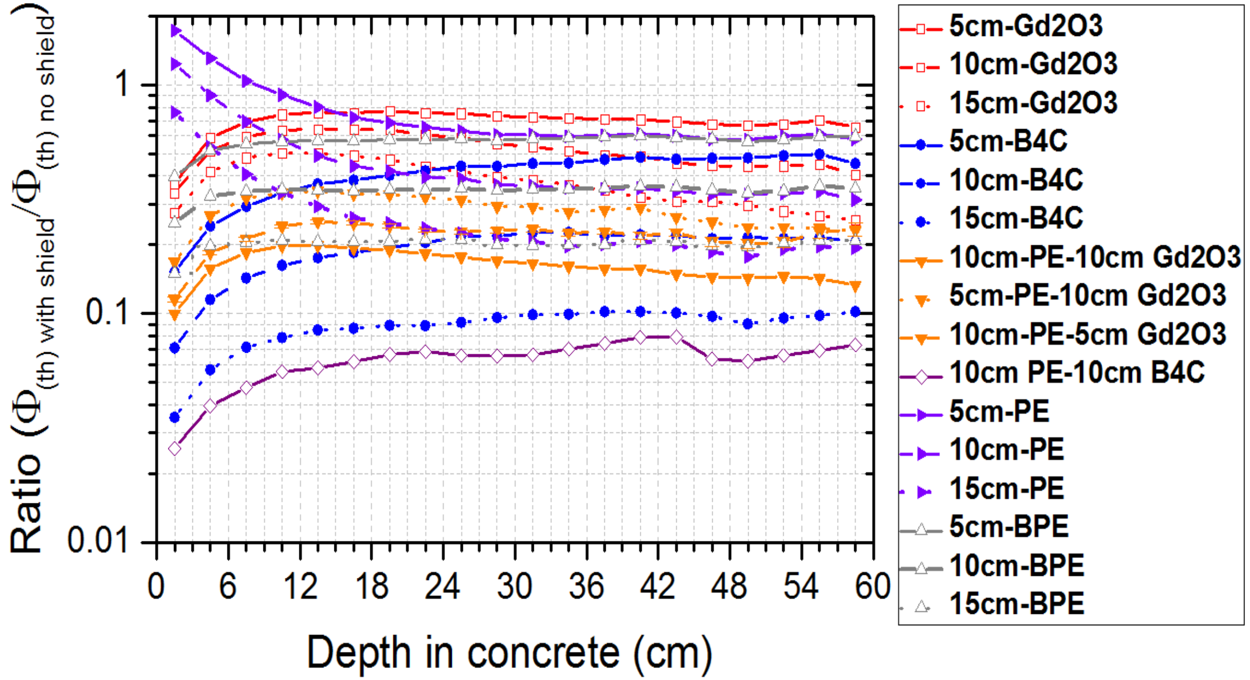


Results for neutron absorbing materials

- B_4C seems to reduce the thermal neutrons better than Gd_2O_3.
- By placing 10 cm Polyethylene, it thermalizes fast neutrons and then can be captured by Gd or B isotopes dramatically.

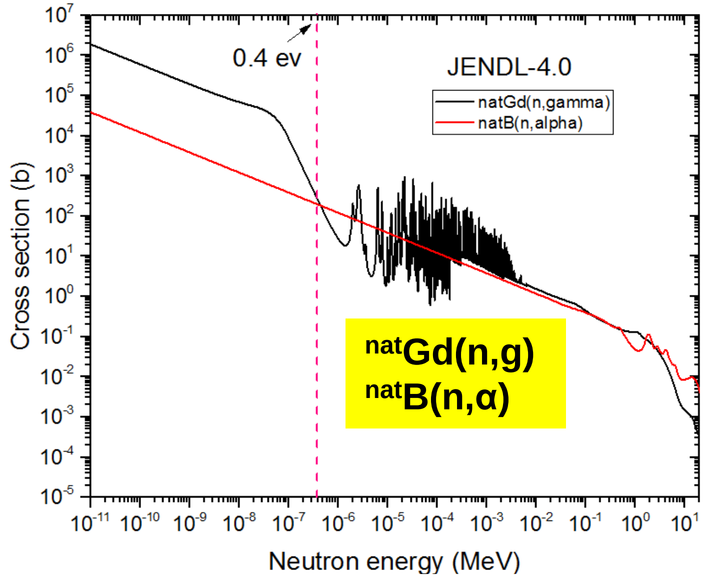
Material	Thickness (cm)	Transmitted neutrons (%)
Gd_2O_3	5 10 15	74 63 **52**
B_4C	5 10 15	34 20 **10**
PE	5 10 15	68 42 **25**
BPE (5%wt)	5 10 15	58 35 **20**
PE+ Gd_2O_3	10+10 5+10	20 23
PE+ B_4C	10+10	**7**

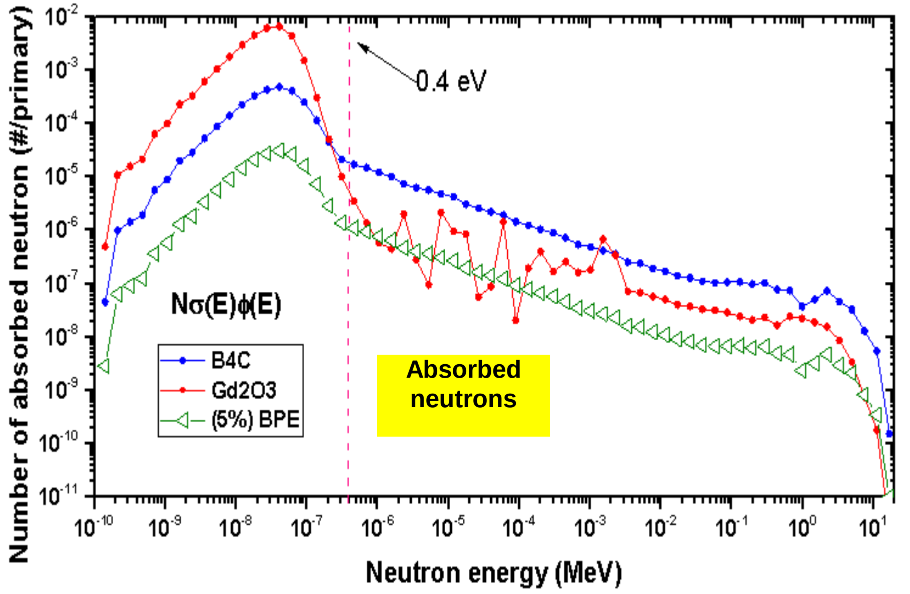




Results for neutron absorbing materials

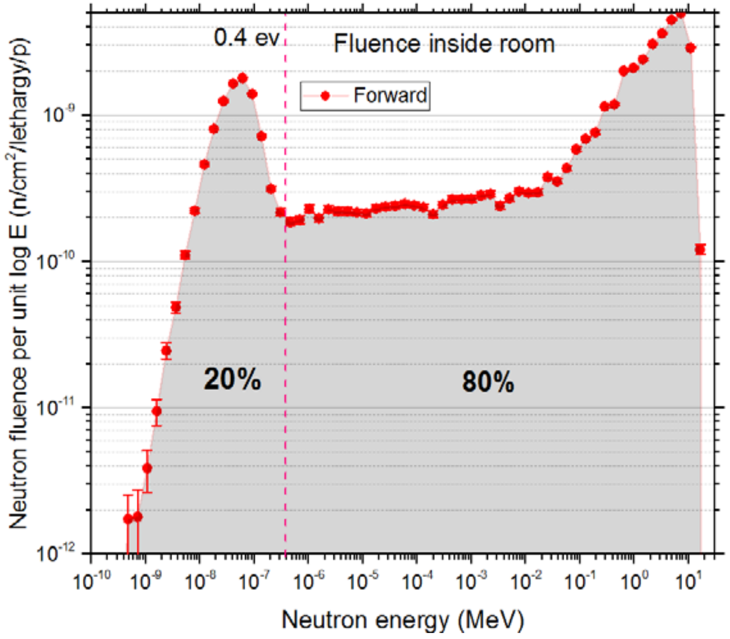
- Gd has high thermal neutron CX below 0.4 eV
- Many resonances above 0.4 eV in natGd CX
- B has no resonances in the CX
- Average CXs above 0.4 eV for ^{nat}Gd and ^{nat}B are approximately equal
- Number density of B in B_4C is higher than Gd in Gd_2O_3 by factor of 4.5

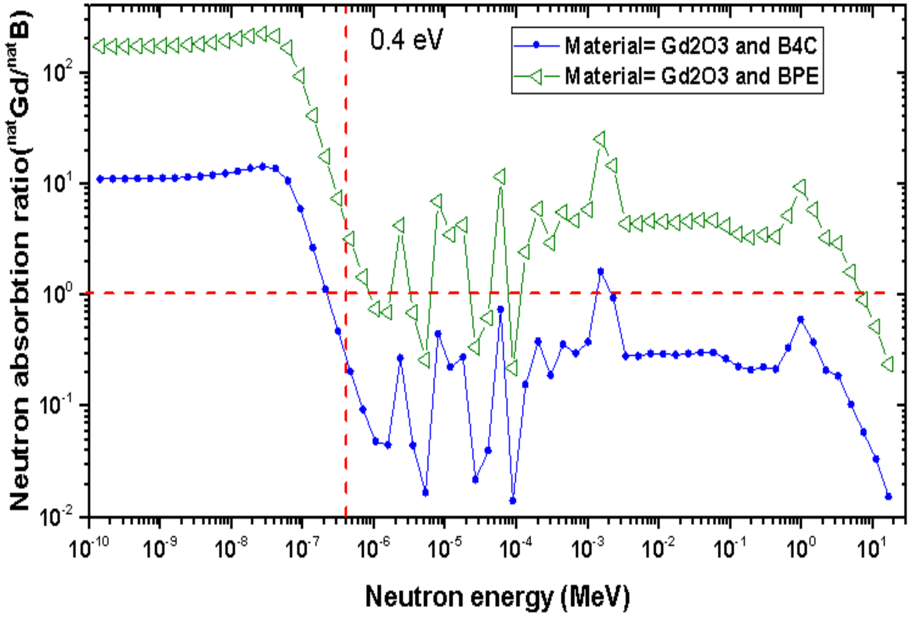




- 80% of neutrons are in the epithermal and fast areas
- B can block more neutrons in these areas than Gd

Therefore, B_4C seems to reduce the thermal neutrons better than Gd_2O_3





❑ Estimating production yield of (Eu-152 & Co-60)

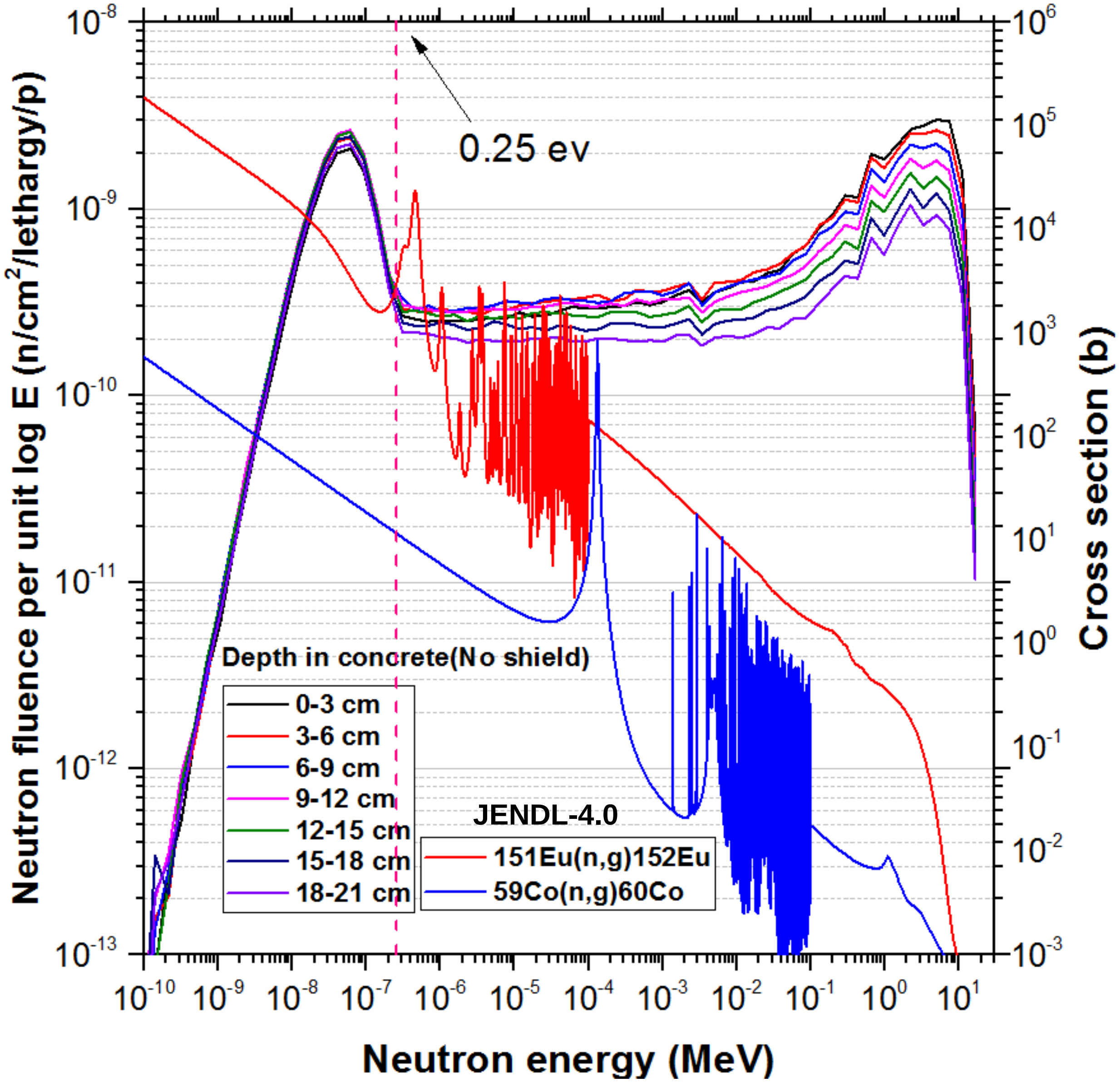
I. Calculating the thermal neutron flux using PHITS

II. Applying JENDL-4.0 library

III. Folding thermal neutron flux and the cross section (E_n< 0.25 eV)

$$R = \sum N\sigma(E)\Phi(E)\Delta E$$

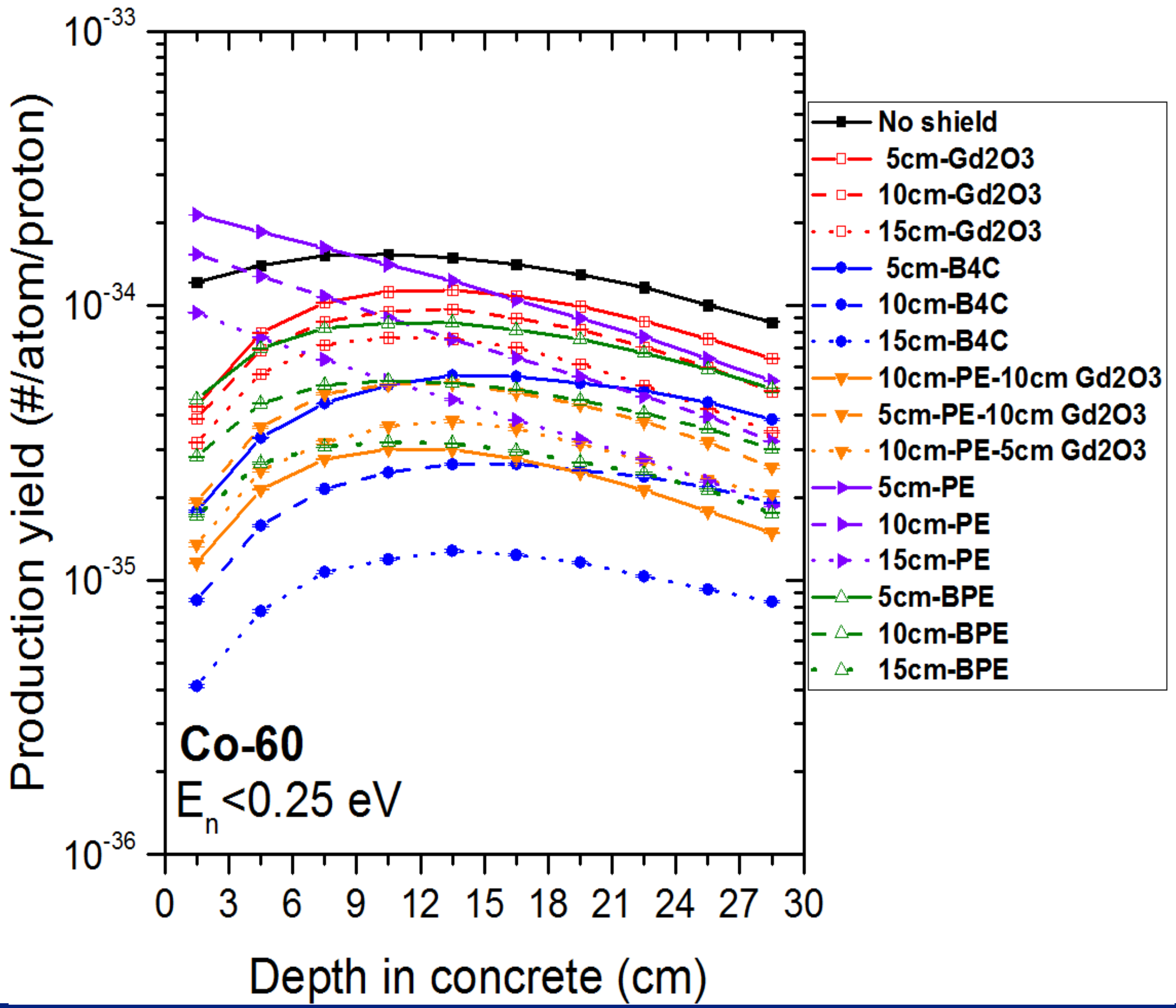
- R: production yield
- N: number of atoms of the target nuclei
- σ: production cross section of the interested nuclei [cm^2]
- Φ: Thermal neutron flux [n/cm^2/MeV/source]
- ΔE: Energy bin (MeV)

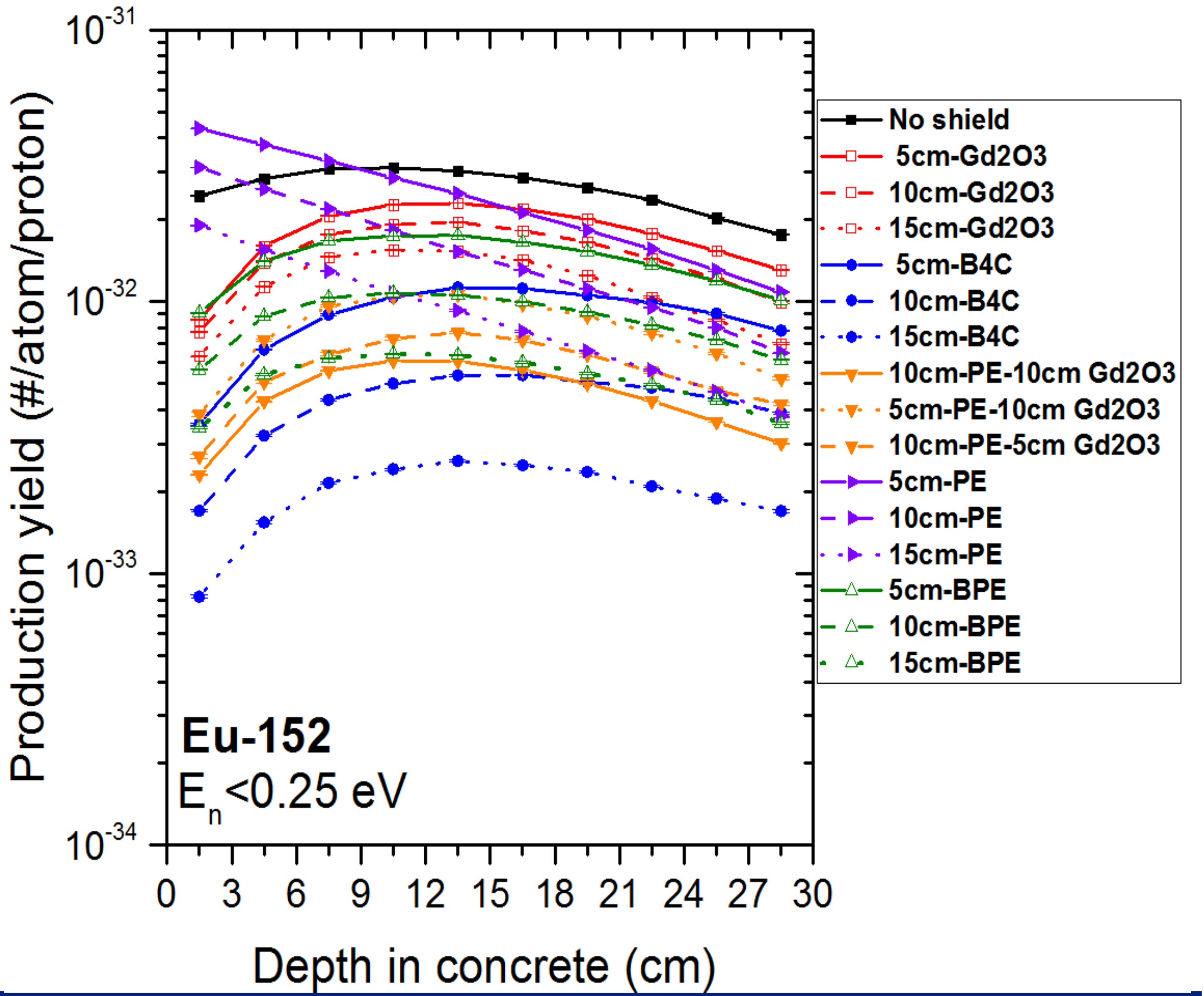


Results for neutron absorbing materials

- Using different neutron absorbing materials:

→ decreasing thermal neutron flux→ Reducing of concrete activation





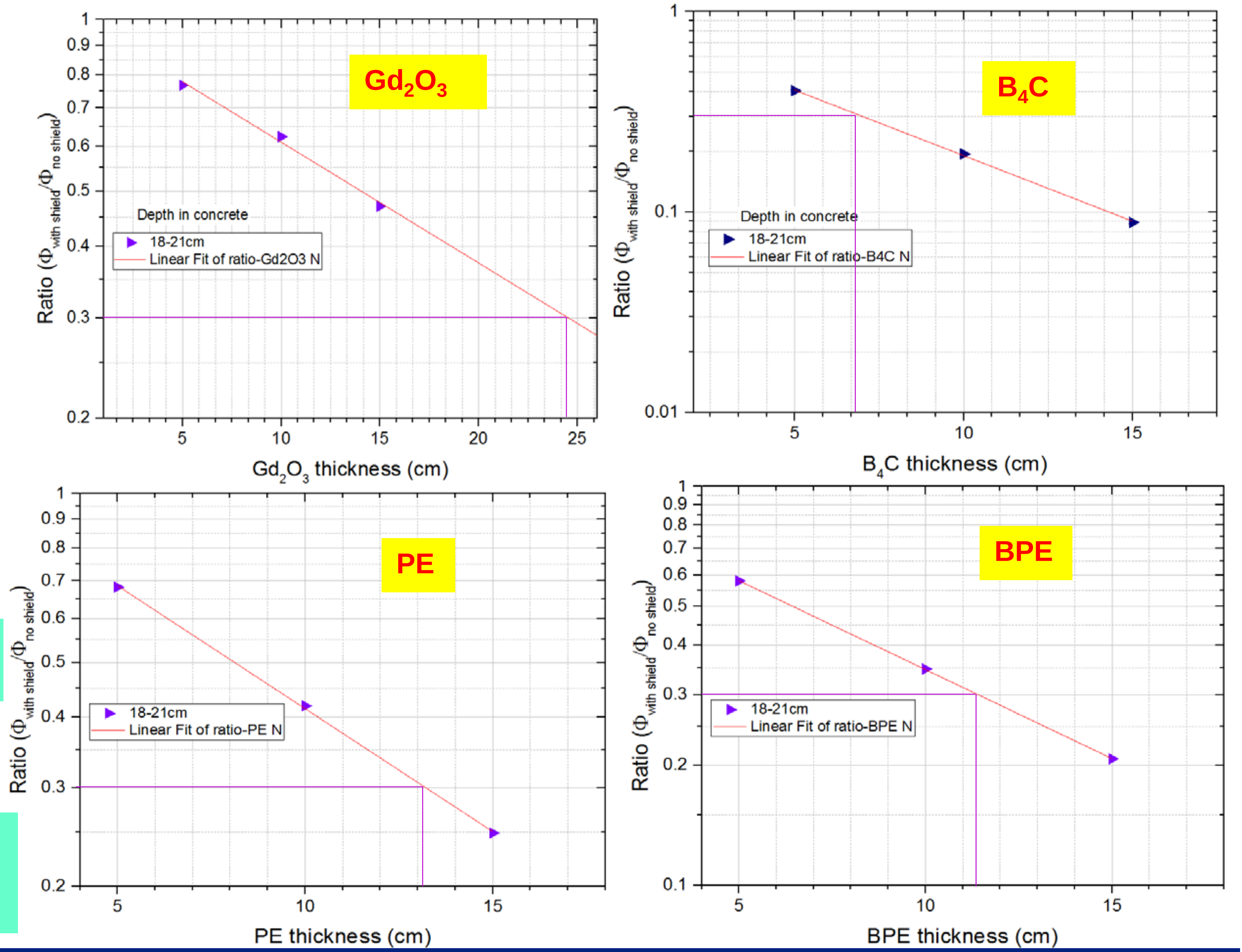
Results for neutron absorbing materials

❑ Attenuation of thermal neutrons

- **Required thickness to reduce thermal neutrons by a factor of 3 @ concrete depth of 20 cm:**
  - ~ 24.5 cm of Gd_2O_3
  - ~ 7 cm of B_4C
  - ~ 14 cm of PE
  - ~12 cm B(5%wt)PE

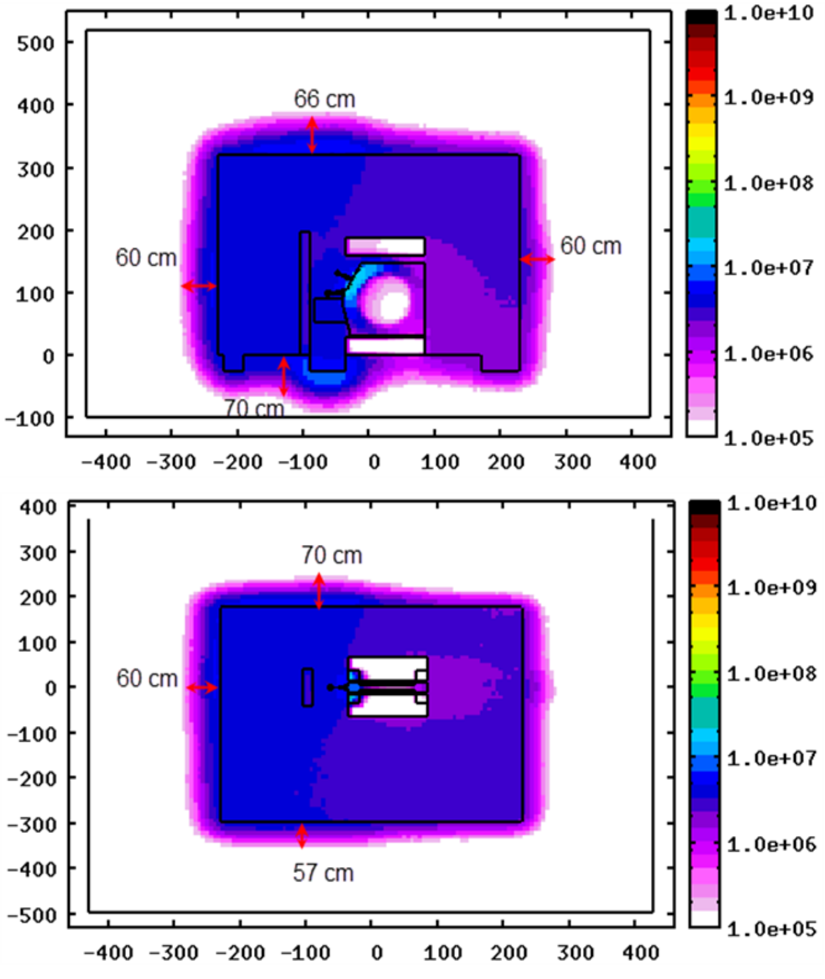
Less expensive than Gd_2O_3 and B_4C

Applied in a real geometry in FLUKA to re-evaluate the neutron distribution

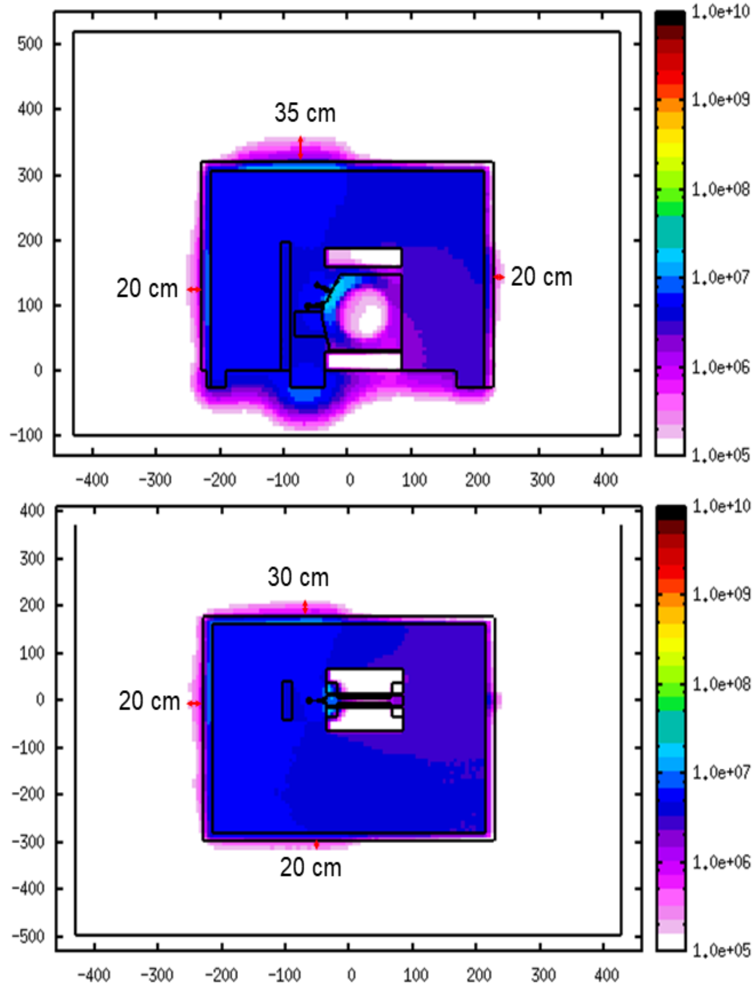


Results for neutron absorbing materials

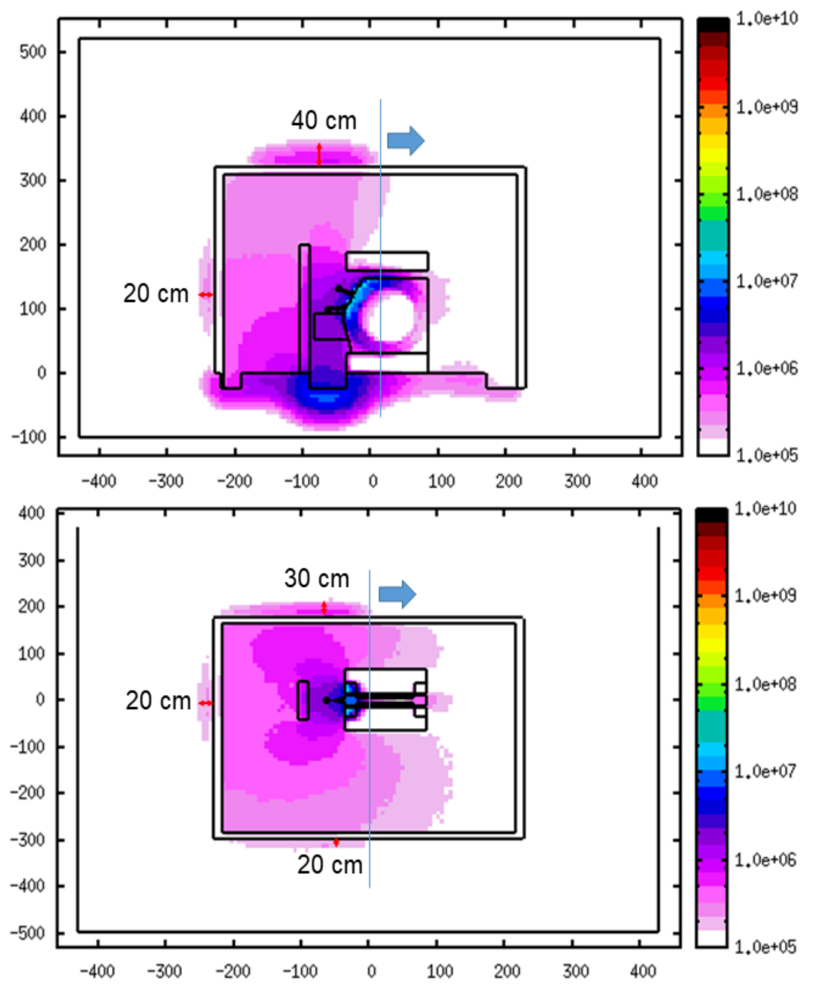
- Expected concrete thickness to be radioactive waste in the PETtrace, without shielding



- Reduced concrete thickness to be radioactive waste in the PETtrace, with 14 cm PE shielding
- Thermal neutrons inside the room is not reduced



- Reduced concrete thickness to be radioactive waste in the PETtrace with 12 cm BPE shielding
- Thermal neutrons inside the room are reduced as well



Conclusions and remarks

- Radioactive waste management is a crucial issue
- There are totally 41 cyclotrons in Korea of which 31 cyclotrons are in operation, generating huge amount of radioactive waste

→ Practical measures must be taken to reduce the waste

- Different cyclotrons were simulated in details using FLULA
- A simple geometry of a PETtrace was simulated in PHITS

→ ~24.5 cm of Gd_2O_3
~7 cm of B_4C
~14 cm of PE
~12 cm of BPE
are needed to reduce the neutrons by a factor of 3 @ 20 cm depth

- Materials with high neutron absorption cross sections are suitable to reduce the activated concrete (Gd_2O_3, B_4C, PE, and BPE)

→ PE and BPE are proved to be acceptable according to the price and neutron absorption capability

→ BPE can reduce the concrete activation and also the thermal neutrons inside the room

Thank you for your attention!