Multifunctional scintillation materials of the garnet structure for nonhomogeneous detecting cells of electromagnetic calorimeters to operate in a harsh irradiation environment

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Materials of interest : radiation hard oxide scintillation materials

Future detector systems for accelerating facilities based on scintillator materials for high energy physics will operate in intensive radiation fields.
It requires a high resistance level as to electromagnetic as well as to hadronic parts of the radiation environment.

Material	ρ, g/cm^3	Z_{eff}/photo absorp. coeff., 511 keV,cm^{-1}/ X_0,cm	Yield, ph/MeV	τ_{sc}, ns	λ_{Max}, nm
$Gd_3Al_2Ga_3O_{12}$:Ce (GAGG)	6.67	50.6/0.12/1.61	46,000	80/800	520
$(Gd\text{-}Y)_3(Al\text{-}Ga)_5O_{12}$:Ce	5.8	45/0.08/1.94	60,000	100/600	560
$Y_3Al_5O_{12}$:Ce (YAG)	4.55	32.6/0.017/3.28	11 000	70	550
$YAlO_3$:Ce (YAP)	5.35	32/0.019/2.2	16 200	30	347
$(Y_{0.3}\text{-}Lu_{0.7})$ AlO_3:Ce (LuYAG)	7.1	60/0.21/1.3	13 000	18/80/450	375
Lu_2SiO_5:Ce (LSO)	7.4	66/0.28/1.1	27 000	40	420
$(Lu\text{-}Y)_2SiO_5$:Ce (LYSO)	7	60/0.20/1.35	30 000	37	420

Yield per MIP for different materials

Ionization losses per 1 mm of the media for 10 GeV e$^-$ and 50 Gev π^-

Material	Density ρ, g/cm^3	dE/dx @ e$^-$, MeV/mm	dE/dx @ π^-, MeV/mm
Plastic scintillator (vinyltoluene based)	1.032	0.154	0.154
$Y_3Al_5O_{12}$ (YAG)	4.55	0.591	0.589
$Y_3(Al_{0.5}\text{-}Ga_{0.5})_5O_{12}$ (YAGG)	4.80	0.614	0.612
$YAlO_3$ (YAP)	5.50	0.708	0.705
$Gd_3Al_2Ga_3O_{12}$ (GAGG)	6.63	0.808	0.804
Lu_2SiO_5 (LSO)	7.4	0.879	0.873
$(Lu_{0.8}\text{-}Y_{0.2})_2SiO_5$ (LYSO)	7.2	0.85	0.85

Light output per MIP (10 GeV e$^-$) per 1 mm in different scintillation materials

Material	LY, ph/MeV	dE/dx @ e$^-$, MeV/mm	Yield, ph per 1 mm per MIP
Plastic scintillator (vinyltoluene based)	10000	0.154	1540
$Y_3Al_5O_{12}$ (YAG)	11000	0.591	6500
$Y_3(Al_{0.5}\text{-}Ga_{0.5})_5O_{12}$ (YAGG)	30000	0.614	18420
$YAlO_3$ (YAP)	16000	0.708	11350
$Gd_3Al_2Ga_3O_{12}$ (GAGG)	46000	0.808	37200
Lu_2SiO_5 (LSO)	27000	0.879	23700
$(Lu_{0.8}\text{-}Y_{0.2})_2SiO_5$ (LYSO)	30000	0.85	25500

Materials of interest

Material	LY, ph/MeV	dE/dx @ e^-, MeV/mm	Yield, ph per 1 mm per MIP	Radiation hardness to protons
Plastic scintillator (vinyltoluene based)	10000	0.154	1540	-
$Y_3Al_5O_{12}$ (YAG)	11000	0.591	6500	+
$Y(Ga\text{-}Al)_5O_{12}$ (YAGG)	30000	0.614	18420	-
$YAlO_3$ (YAP)	16000	0.708	11350	-
$Gd_3Al_2Ga_3O_{12}$ (GAGG)	46000	0.808	37200	+
Lu_2SiO_5 (LSO)	27000	0.879	23700	+
$(Lu_{0.8}\text{-}Y_{0.2})_2SiO_5$ (LYSO)	30000	0.85	25500	+

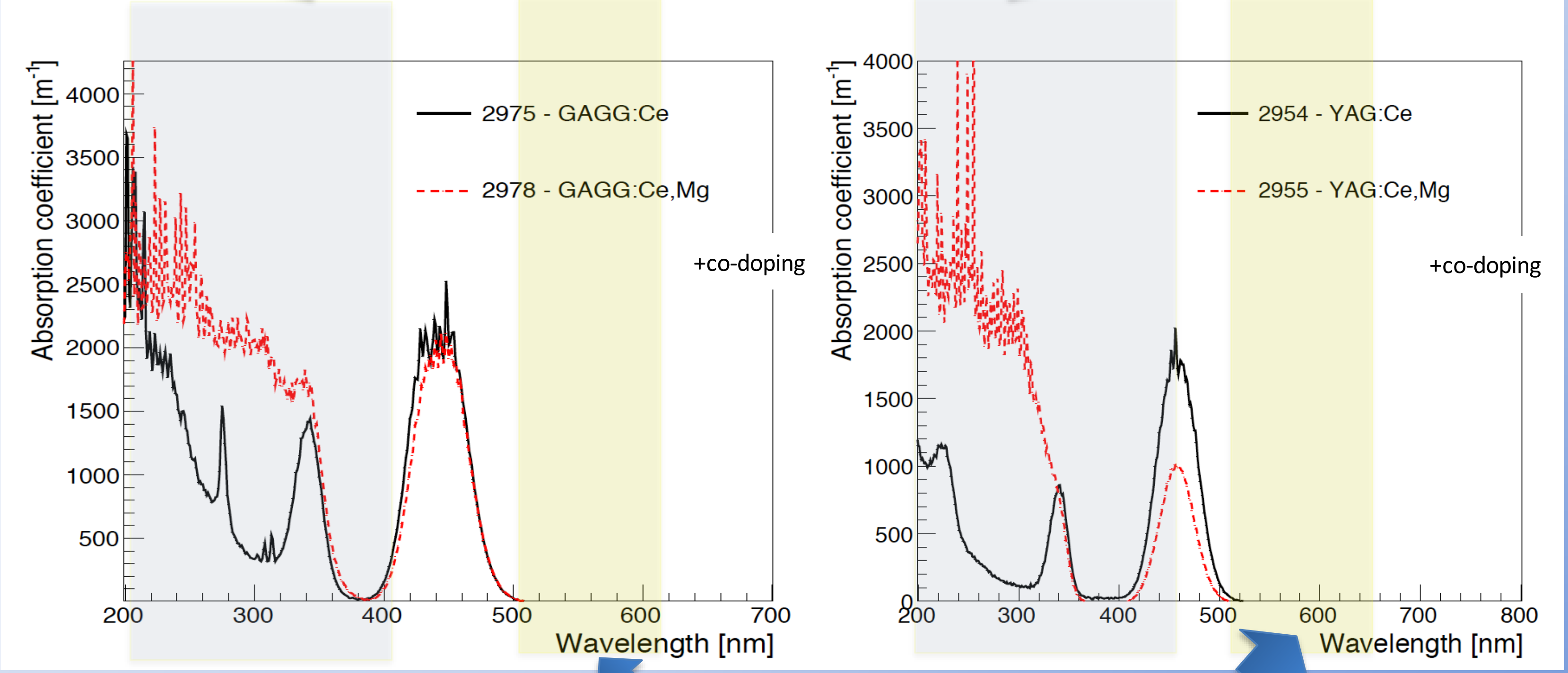
Garnets are more radiation hard than perovskites and oxyorthosilicates

GAGG shows the best yield per MIP

GAGG also shows 170 ps coincidence time resolution with ^{22}Na which is close to one of LSO or LYSO

Why are garnets irradiation tolerant?

Spectral range of the colour centers



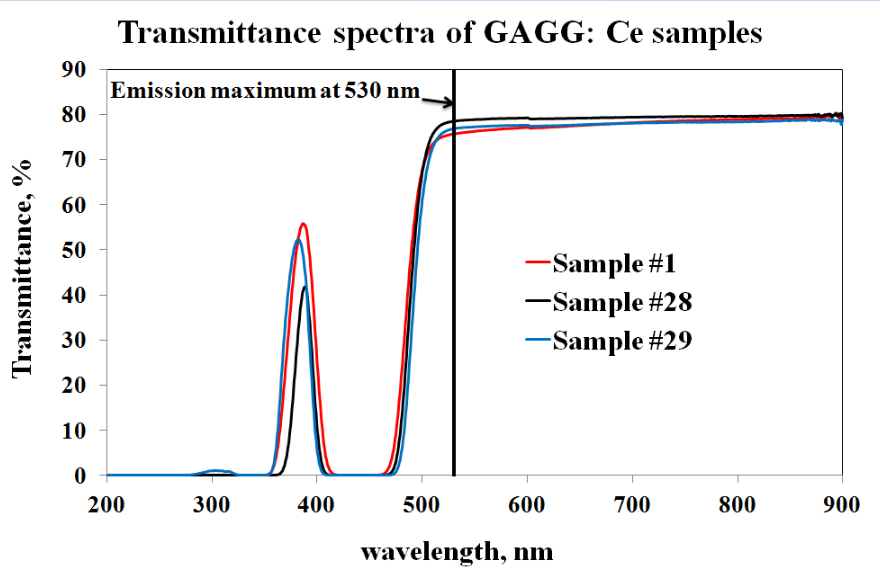
Spectral range of the luminescence

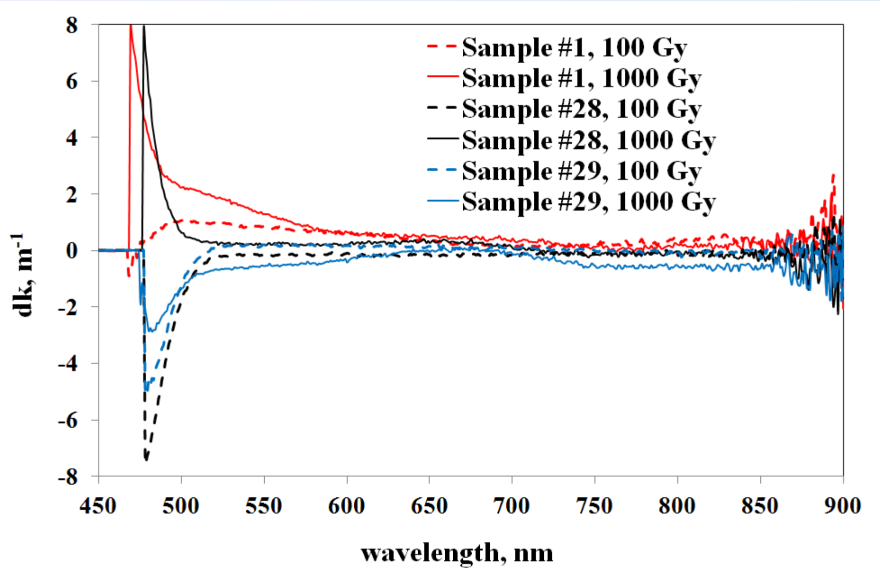
Induced absorption of GAGG: Ce and YAG: Ce, also co-doped crystals are out of luminescence range.

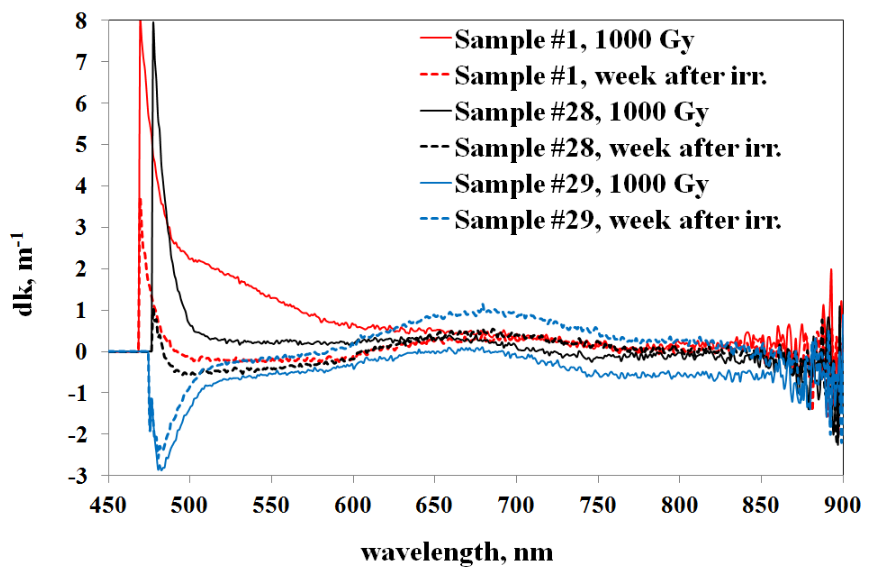
GAGG : Ce, Mg, Ti.
Induced absorption after irradiation with gamma-quanta (^{60}Co)

3 samples of GAGG were tested.
GAGG #1: Ce, Mg , Ti;
GAGG #28: Ce, Mg, Ti;
GAGG# 29: Ce

$$\Delta k = \ln\left(\frac{T_{bef}}{T_{after}}\right)\cdot\frac{1}{d}$$

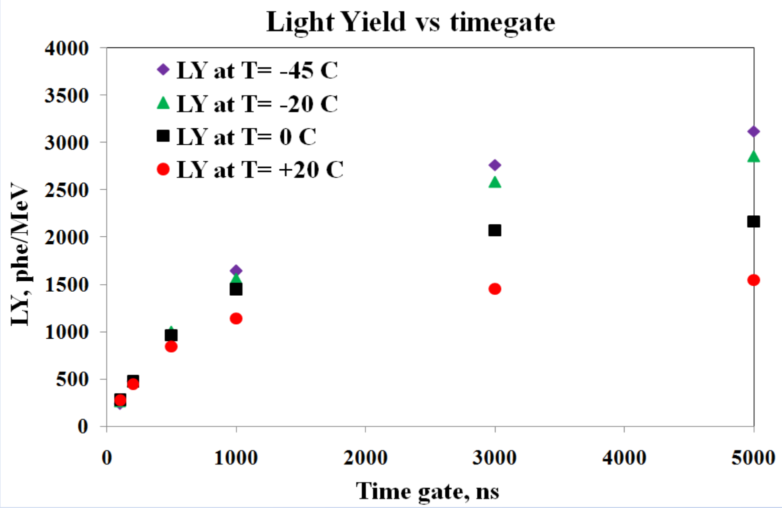

Transmittance spectra of GAGG: Ce samples

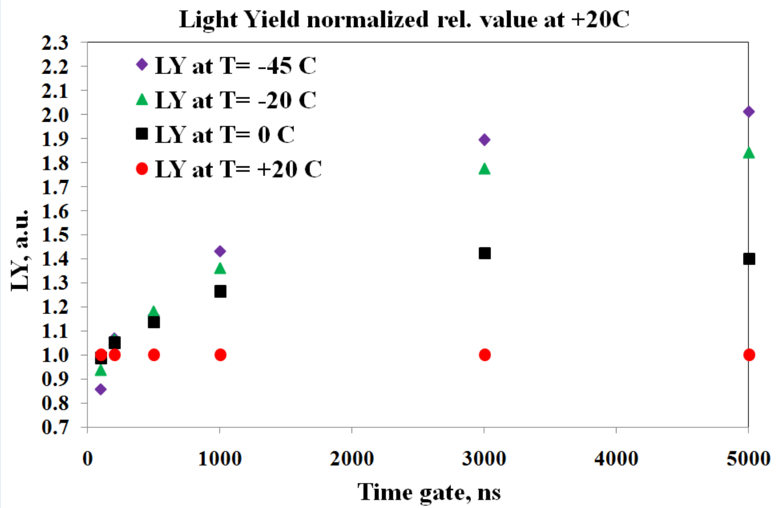




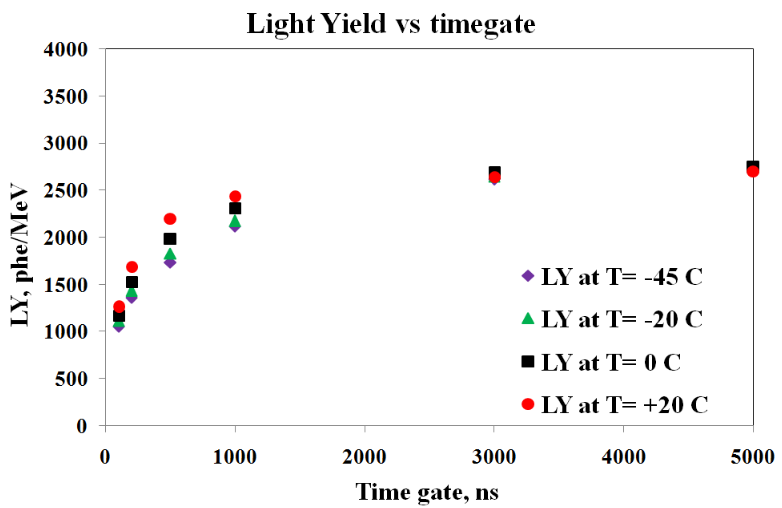
Lighy Yield of garnets vs operational temperature.

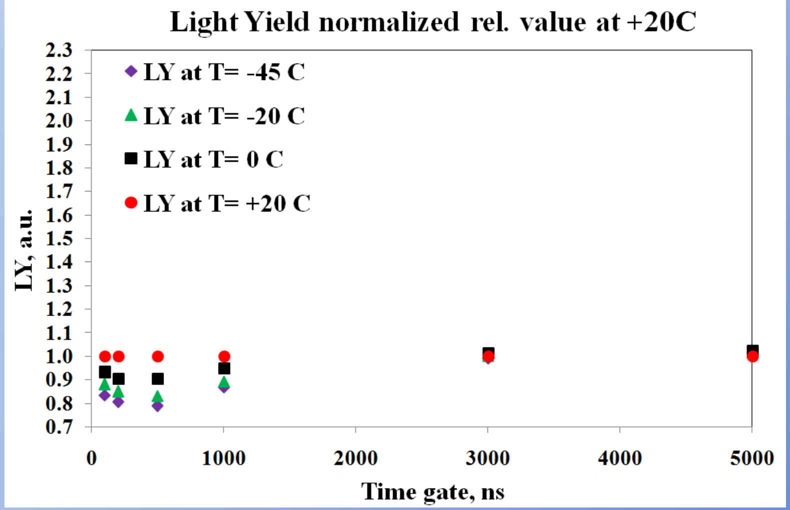
YAG: Ce





YAG: Ce + Mg

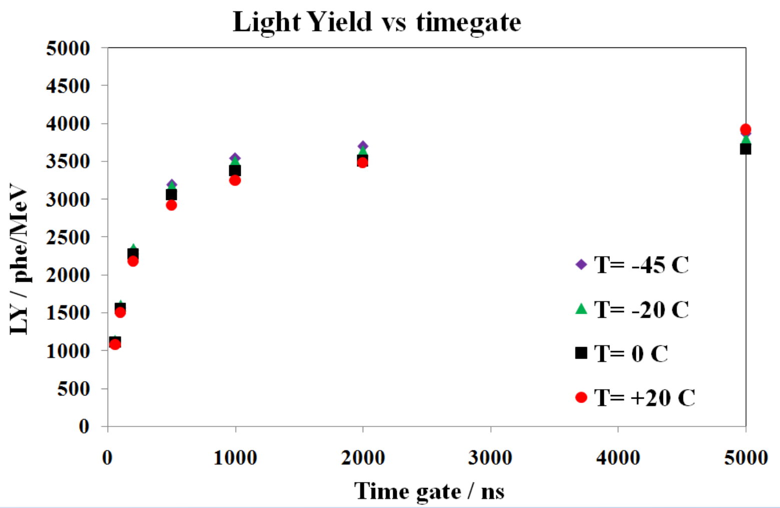


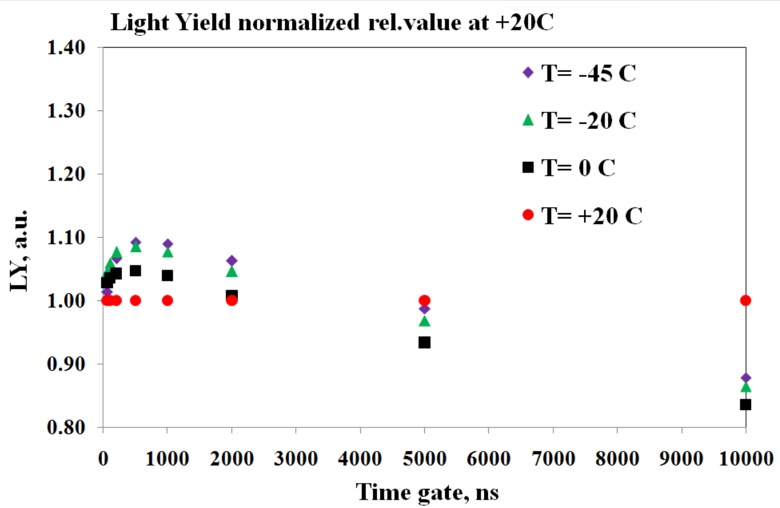


Light Yield vs integration time was measured (Hamamatsu PMT XP 2059, ^{241}Am)

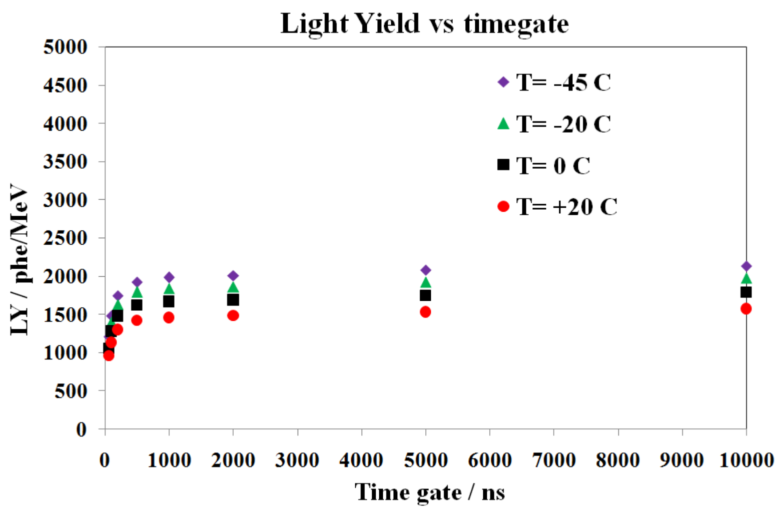
Lighy Yield of garnets vs operational temperature.

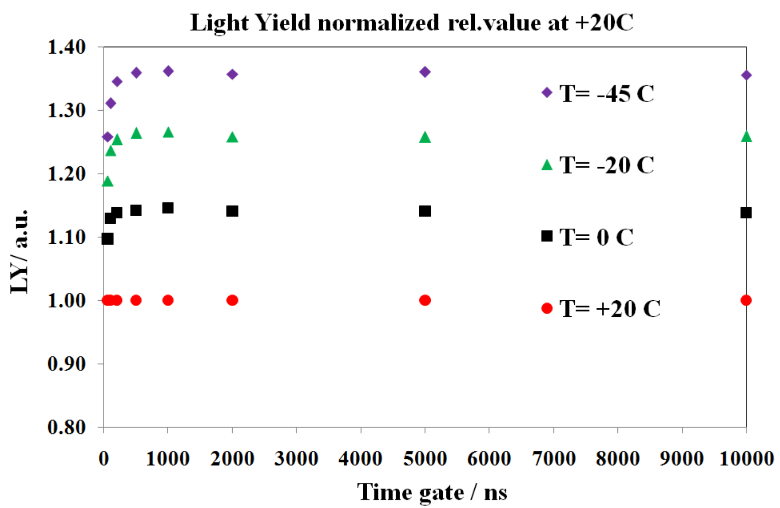
GAGG: Ce





GAGG:Ce + Mg, Ti





Light Yield vs integration time was measured (Hamamatsu PMT XP 2059, ^{241}Am)

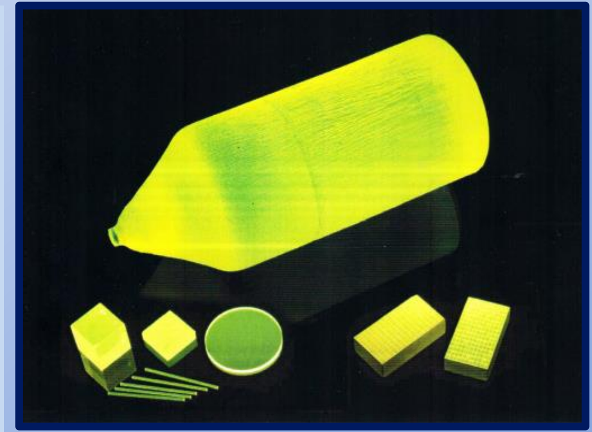
GAGG-gadolinium aluminum gallium garnet $Gd_3Al_2Ga_3O_{12}$

$n + {}^{155}Gd \rightarrow {}^{156}Gd + \gamma$
(a set of soft gammas with total energy ~8.5 MeV)

$n + {}^{157}Gd \rightarrow {}^{158}Gd + \gamma$
(a set of soft gammas with total energy ~8 MeV)

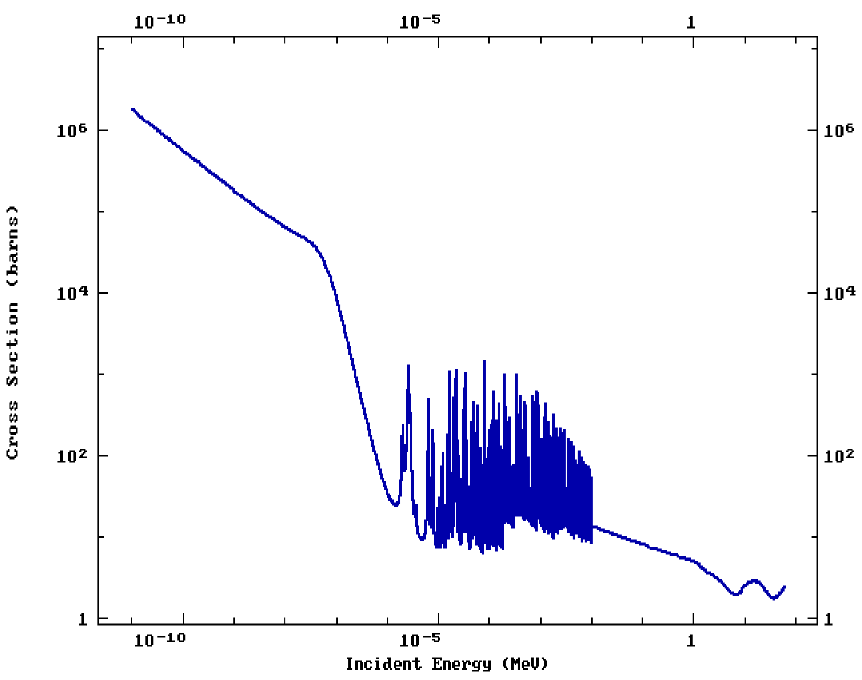
GAGG is the aluminum gallium garnet doped with Ce, Mg, Ti is perfect scintillation material designed to overcome drawbacks of Ce solely doped and Mg co-doped crystals.

GAGG has high energy resolution, perfect time resolution (CTR)
fast scintillation kinetics, high effective charge Z_{eff} of compound, perfect radiation hardness to gamma and hadron irradiation.



Density, g/cm^3	Z_{eff}/photo absorp. coeff., 511 keV,cm^{-1}	Emission maximum, nm	Light yield, ph/MeV	Decay kinetics, ns(%)	Energy resolution, %	Time resolution (CTR), ps
6.68	51/0.12	520	38000(RT) 46000(-45°C)	30 (25%), 80 (60%), 100-200 ns (15%)	6.2%(511keV,-20°C) SiPM 3.6%(1270keV, -20°C) SiPM	170 (-20 to 20°C)

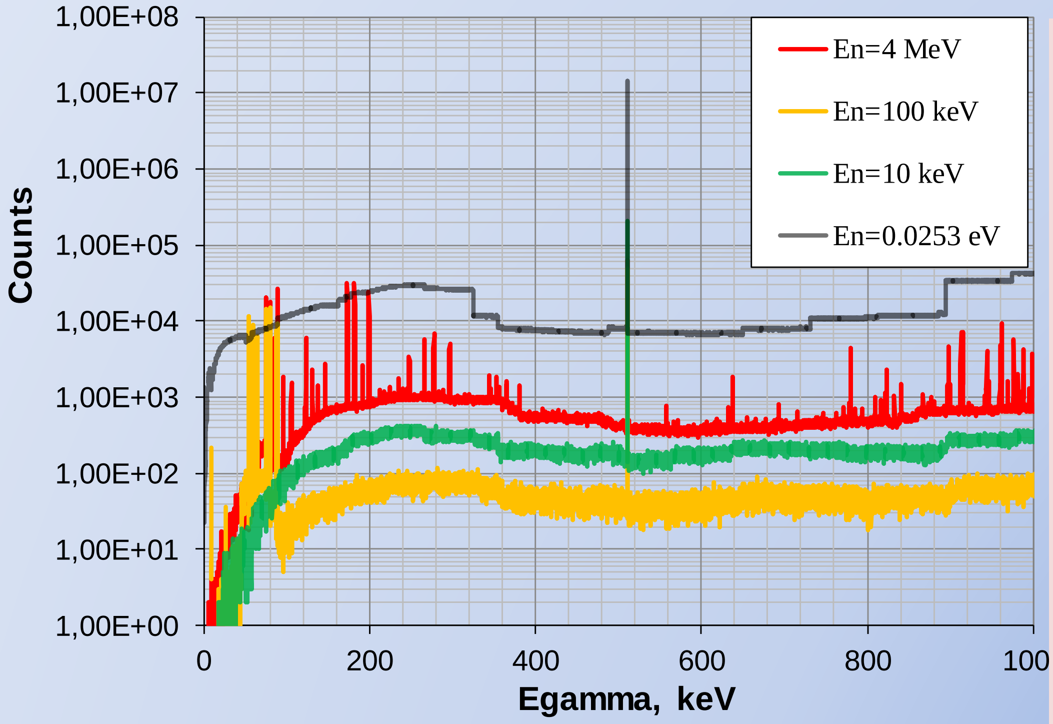
Gd isotopes natural mixture is sensitive to neutrons



Neutron total cross sections of natural mixture of Gd isotopes
Nuclear Data File (ENDF), https://www-nds.iaea.org/exfor/endf.htm

- **Characteristic for them broad zone of resonances increases the neutron absorption efficiency for neutron energies from 1.0 eV up to 10 keV;**
- **Starting from ~ 55 keV threshold of the neutron energy, the process of the neutron inelastic scattering is accompanied by the gamma-quanta emission, forming multiple soft lines in the resulting gamma-quanta spectrum**

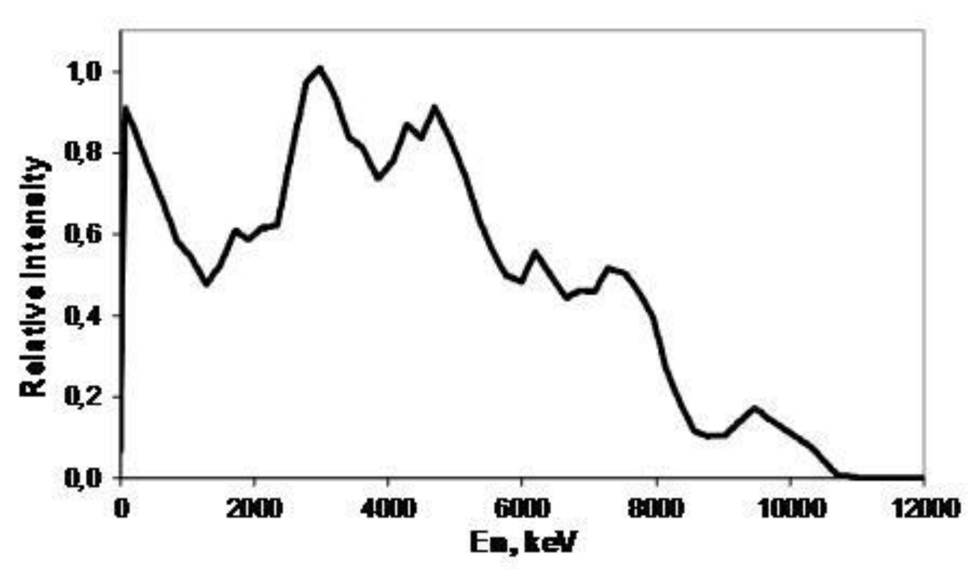
Production of γ-quanta in Gd(n, γ)

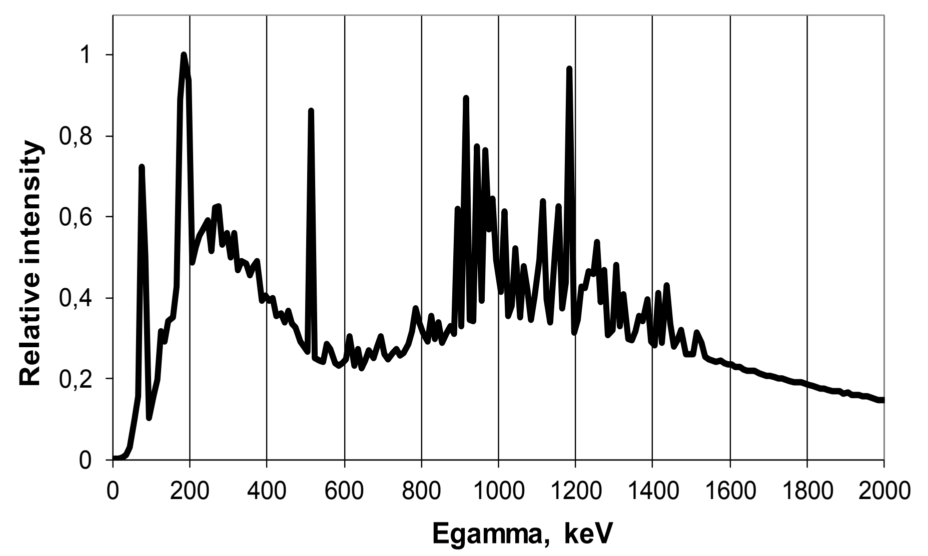


- The 511 keV gamma-line presents across all gamma spectra;
- There are no prominent soft γ-lines in the spectra for thermal neutrons (E_n = 0.0253 eV) and for neutrons with E_n = 10 keV;
- The γ-lines with energies <100 keV
- arise in spectrum for E_n = 100 keV
- resulting from inelastic neutron
- scattering having a threshold
- ~E_n = 55 keV.
- Numerous gamma-lines present
- across all the gamma-spectrum
- for E_n = 4 MeV.

Spectra of the individual energies of the emitted gamma quanta in metallic gadolinium with 2 mm thickness, irradiated with monochromatic neutrons, simulated with GEANT4

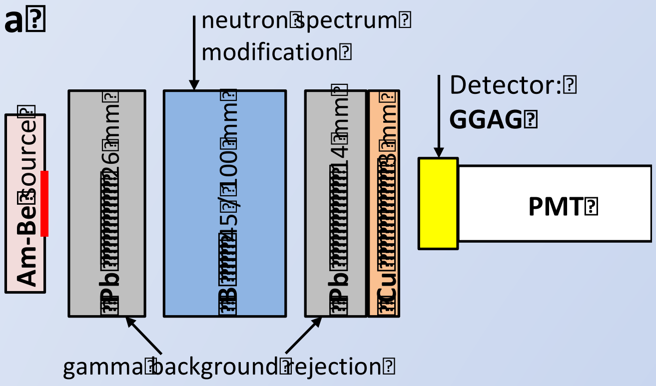
GEANT4 modeled spectrum of γ-quanta born in 2 mm Gd metal plate, irradiated with neutrons from Am-Be source

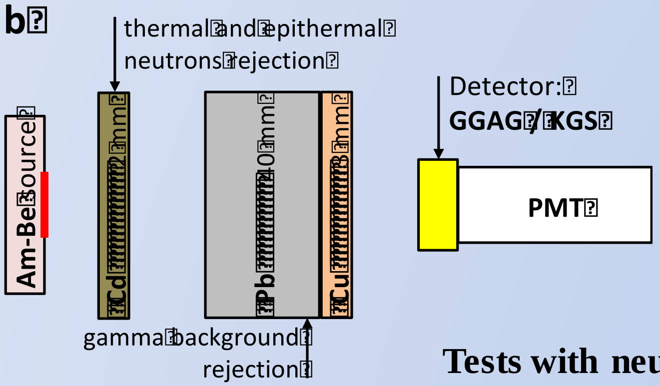




Spectrum of neutron energies from Am-Be neutron source, in accordance with ISO 8529-1:2001(E)

Layout of the measurement scheme



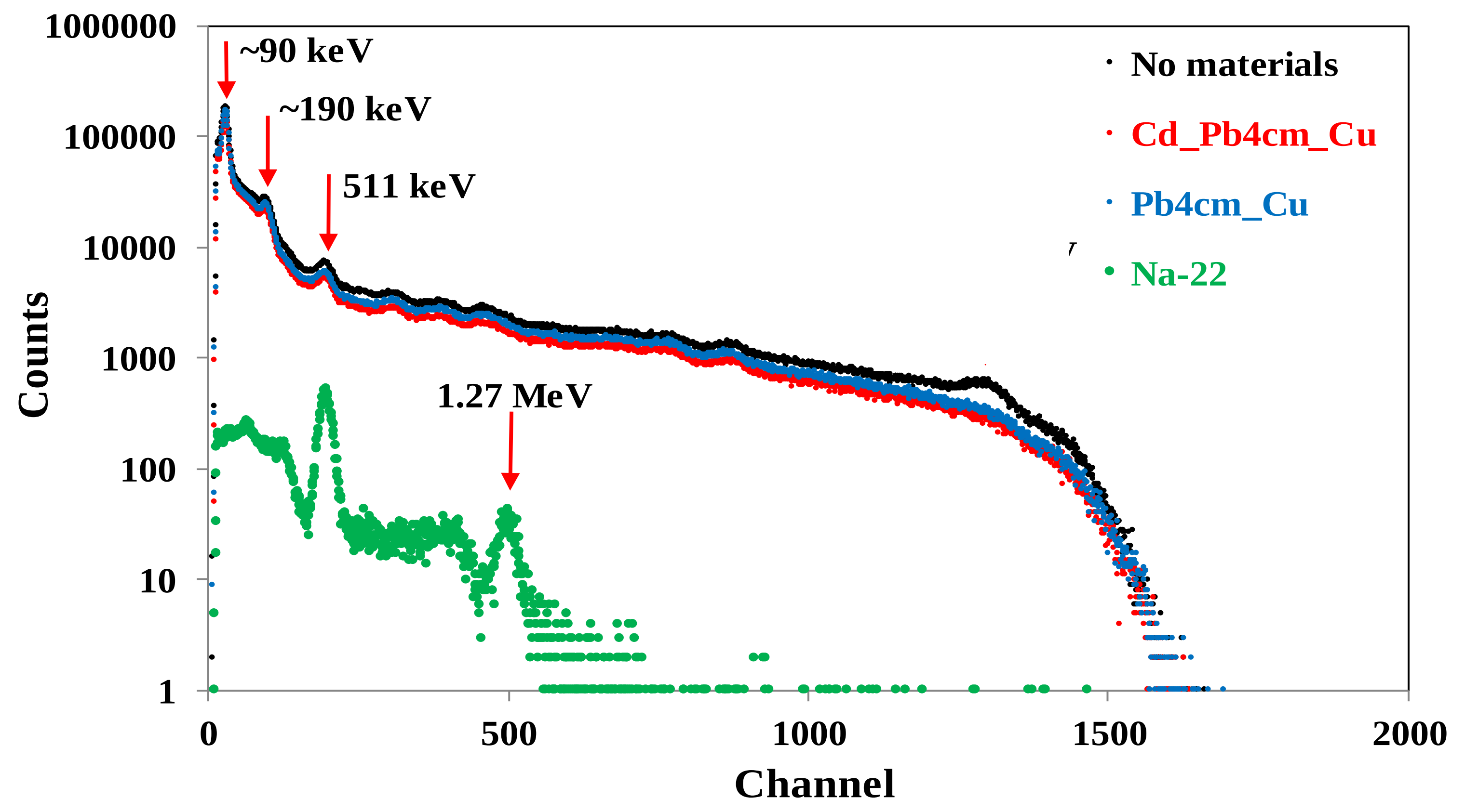


GAGG:Ce samples with following dimensions were tested:

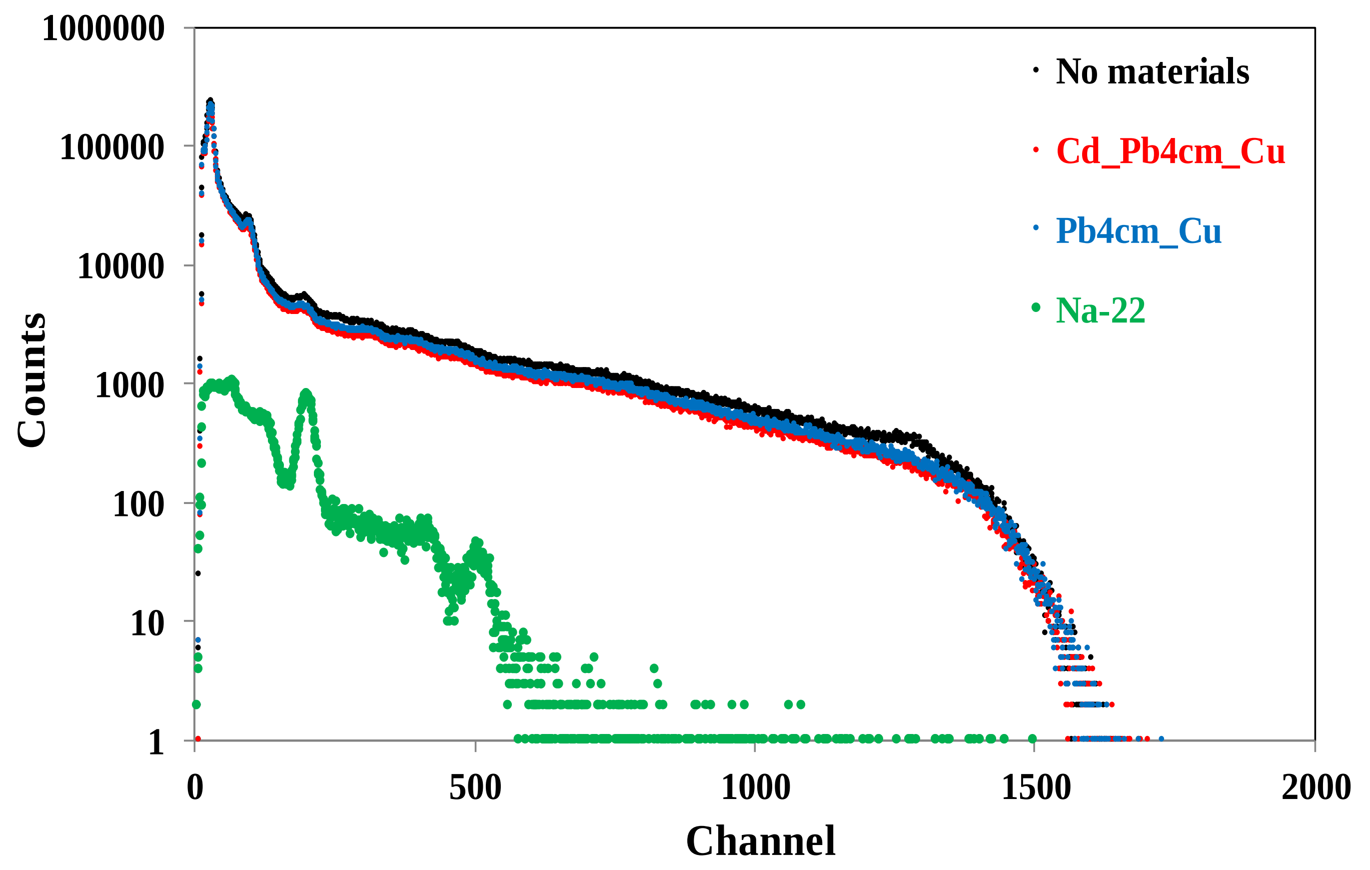
1) **15×18×7 mm size, surface area faced to neutron source 2.70 cm^2, volume 1.89 cm^3;**
2) **Ø30×2 mm, surface area 7.01 cm^2, volume 1.41 cm^3**

Tests with neutrons of an Am-Be source (<En> = 4.2 MeV, En max = 11 MeV) have been performed. 241Am source activity is 220 GBq,
with estimated neutron yield of ≥1.3×10^7 neutron/s.
Each sample was wrapped in multiple layers of Teflon® tape and attached to a Hamamatsu R2059 PMT.
The optical connection between samples and the PMT was performed with “Baysilone® M 300.000” optical grease.

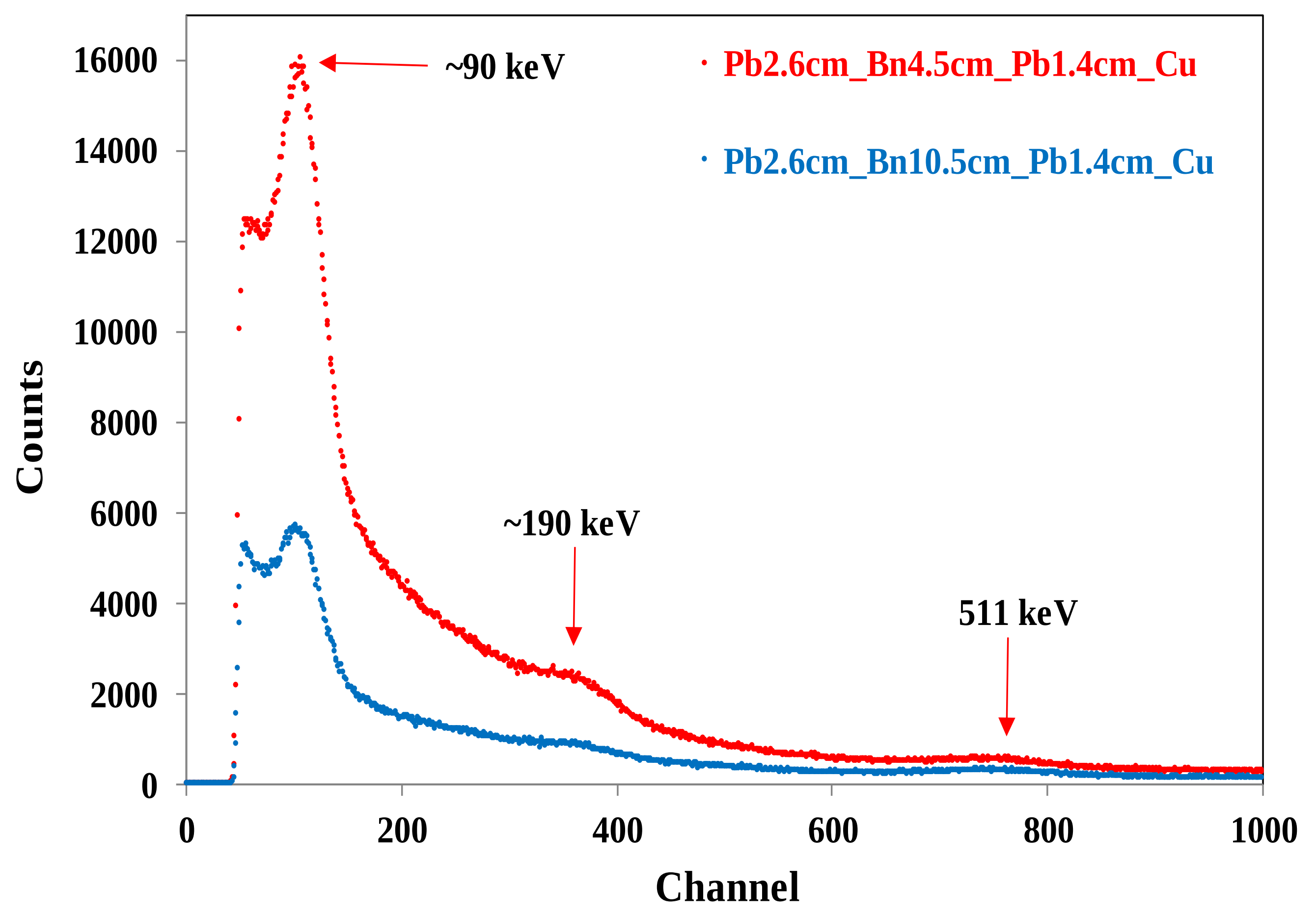
The γ-spectra measured with GAGG 15×18×7 mm sample irradiated by neutrons from Am-Be source.



The γ-spectra measured with GAGG and Ø30×2 mm^3 sample irradiated by neutrons from Am-Be source



Spectra of Am-Be source measured with GAGG 15×18×7 mm at different thicknesses of boron acid absorber



Summary

- Our modeling and experimental evaluations have shown that gamma-lines acquired with GAGG scintillation detector under neutron irradiation are located in the energy range up to 4 MeV. Main gamma lines are located in the energy range shorter than 0.6 MeV.

- Spectrum of MIPs with GAGG crystal plate of the thickness 2 mm already will have signal near 1.5 MeV, so overlapping with gamma quanta, generated with neutrons will be small.

- Gamma-quanta, generated by neutrons in GAGG will appear under detector threshold at the registration of high energy particles while “shashlyk” or “spaghetti” type detectors will be used.

- GAGG has a good potential to be applied for nonhomogeneous detecting cells of electromagnetic calorimeters to operate in a harsh irradiation environment