Radioactivity measurements at Milano Bicocca

JUNO-Italia meeting
Frascati, March 14-15, 2018

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& Milano-Bicocca group

Liquid Scintillator (LAB) radiopurity assay techniques

^{238}U:	10^{-15}
^{232}Th:	10^{-15}
^{40}K:	10^{-16}
^{210}Pb:	$1.4*10^{-22}$

Purified →

^{238}U:	10^{-17}
^{232}Th:	10^{-17}
^{40}K:	10^{-18}
^{210}Pb:	10^{-24}

→ Extremely demanding requirements!

Two complementary approaches:

LAB Radiopurity test @ Daya Bay:

- exploiting Bi-Po coincidence

LAB Radiopurity measurement @ Milano-Bicocca:

- exploiting NAA and gamma measurements on HPGe
- further step: implementing beta-gamma coincidence to reduce background and improve the sensitivity on the LAB radiopurity.

Neutron Activation Analysis

Chosen radioassay technique for Acrylic: NAA

- Advantages: very high sensitivity, proven feasibility
- Disadvantage: sensitive only to radioactive chain progenitors

$$^{A}Z + n \rightarrow {}^{A+1}Z + \gamma$$

Three key ingredients:
- high neutron flux
- high enough neutron capture cross section
- “convenient” daughter nucleus (γ emission, half-life time)

Sensitivity depends on:
- neutron exposure time
- interferences in the matrix
- background in the region of the gamma emission

care in the sample preparation is extremely important!

Neutron Activation Analysis for ^{40}K, ^{232}Th and ^{238}U

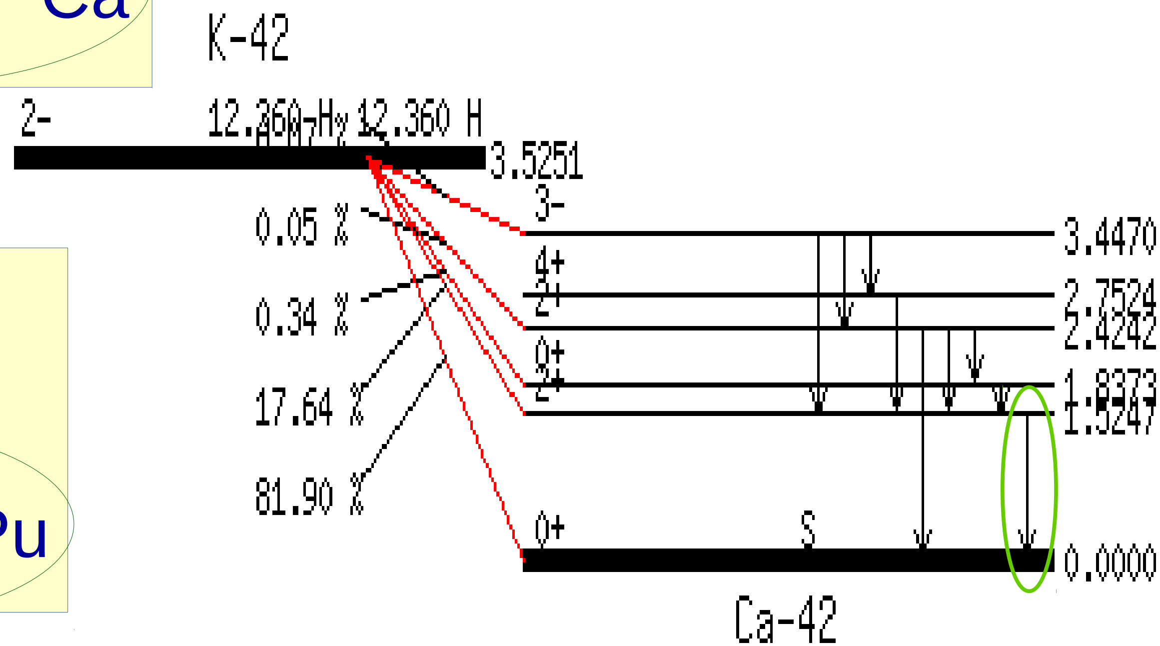
$n + {}^{41}K \rightarrow {}^{42}K \xrightarrow[12.36\ h]{\beta^-} {}^{42}Ca$

$n + {}^{238}U \rightarrow {}^{239}U$

${}^{239}U \xrightarrow[23.5\ m]{\beta^-} {}^{239}Np \xrightarrow[2.36\ d]{\beta^-} {}^{239}Pu$

$n + {}^{232}Th \rightarrow {}^{233}Th$

${}^{233}Th \xrightarrow[22.3\ m]{\beta^-} {}^{233}Pa \xrightarrow[27.0\ d]{\beta^-} {}^{233}U$

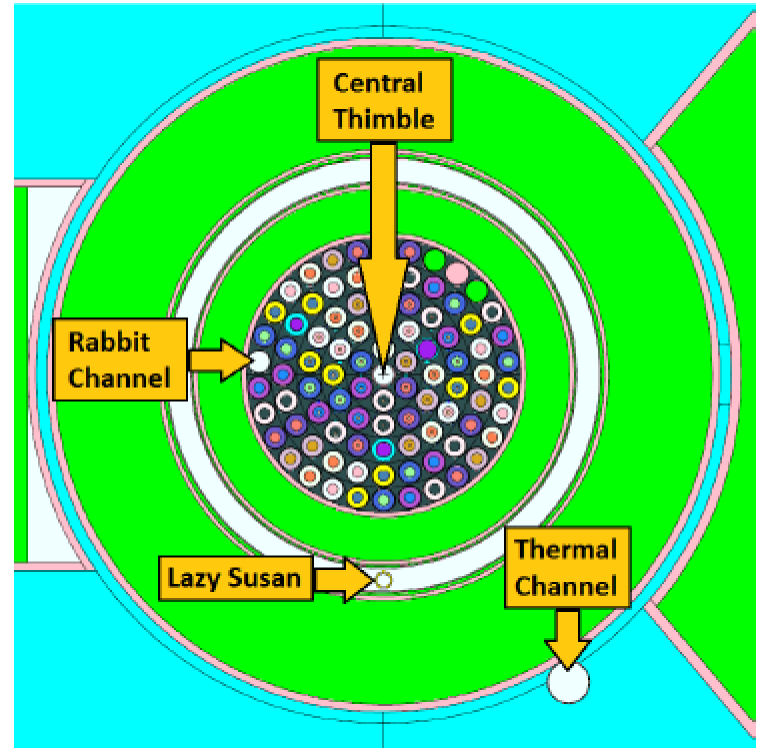


gamma measurements on HPGe

from peak activity
to contaminant concentration
by comparison with NAA standards

NAA irradiation campaigns @ TRIGA reactor

TRIGA Mark II research reactor (250 kW) - Pavia, Italy



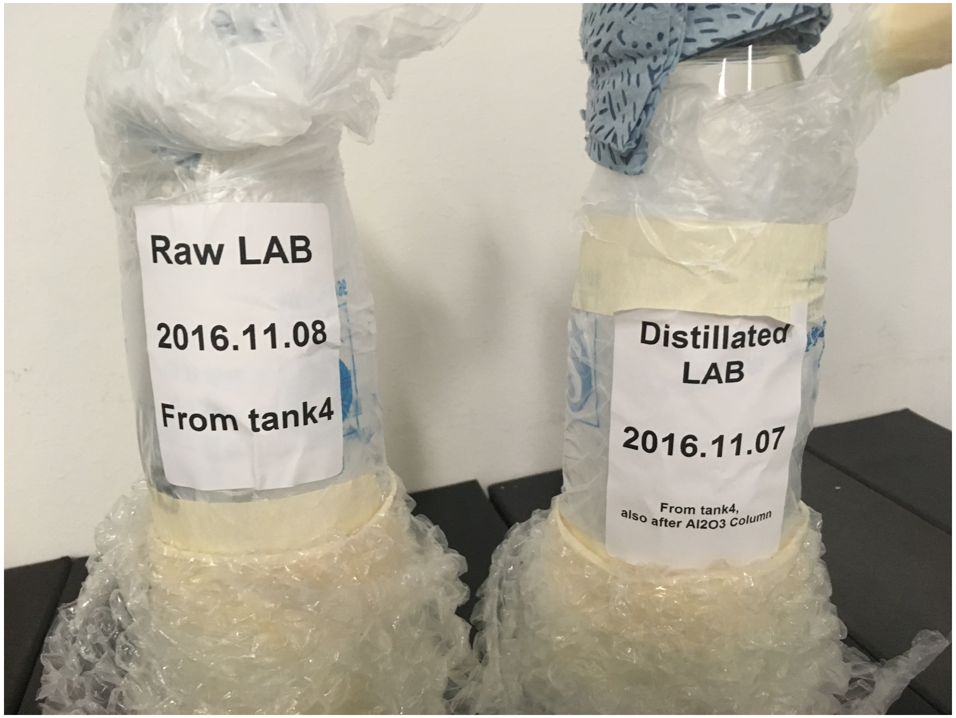
1) First neutron irradiation test on December 18, 2017

One **RAW LAB sample** was irradiated for **3 hours** in the Lazy Susan facility (together with the U, Th, and K standards and the container blank sample).

2) Second neutron irradiation test on January 15, 2018

One **distilled LAB sample (after Al_2O_3 column)** was irradiated for **6 hours** in the Lazy Susan facility (together with the U, Th, and K standards and the container blank sample).

NAA irradiation campaigns @ TRIGA reactor



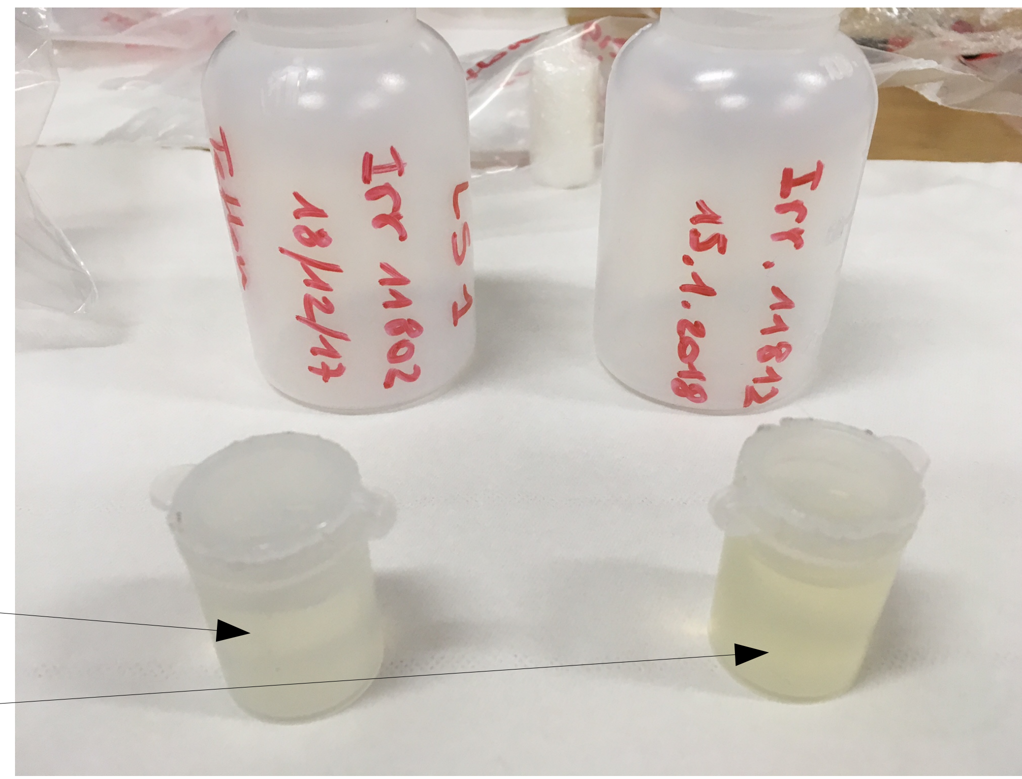
The two LAB samples

- RAW LAB: Irradiation #1
- Distillated LAB (after Al_2O_3 column): Irradiation #2

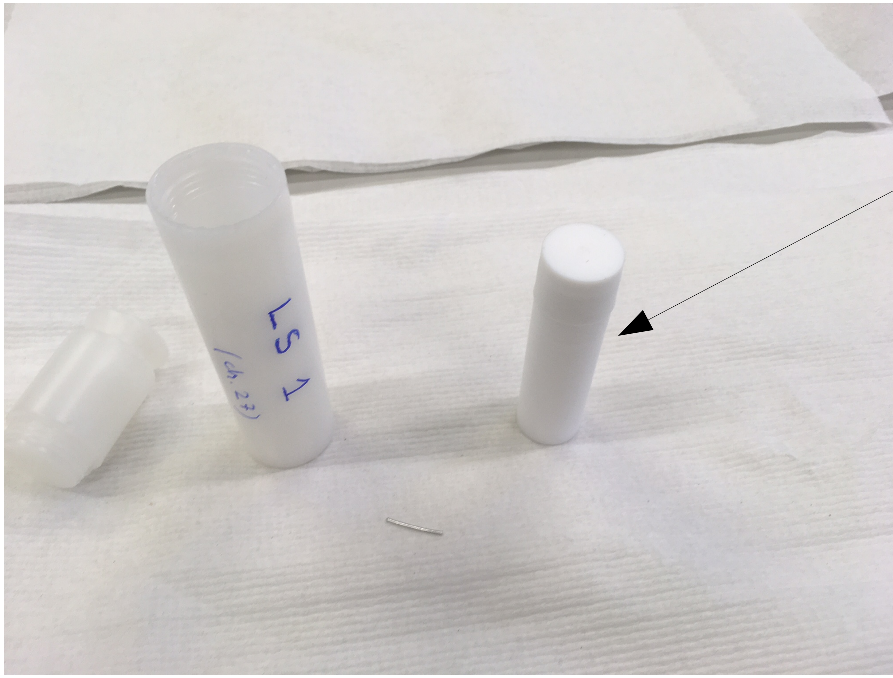
First irradiation tests to study:
- Radiolysis
- Interfering contaminants

3 hour irradiation (mild color change)

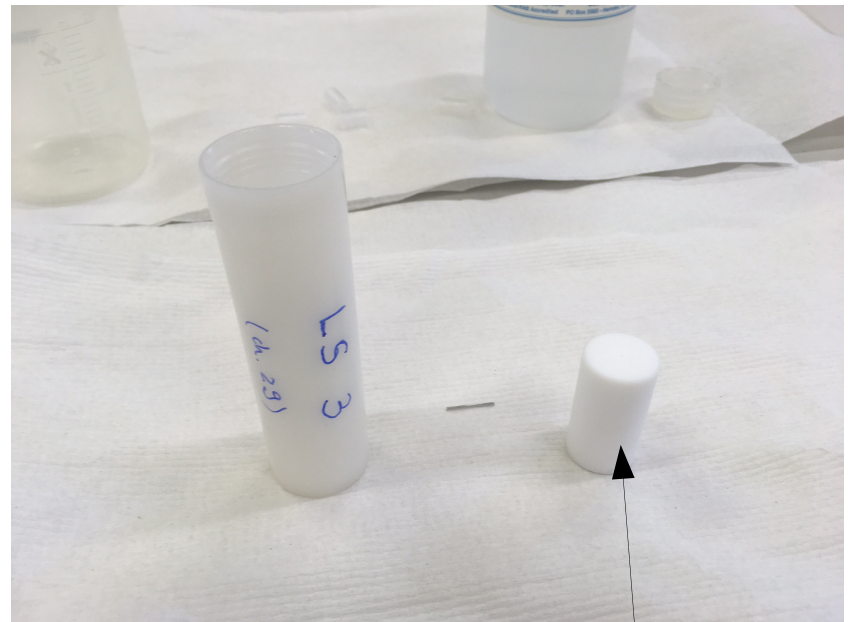
6 hour irradiation (LAB has become yellowish)



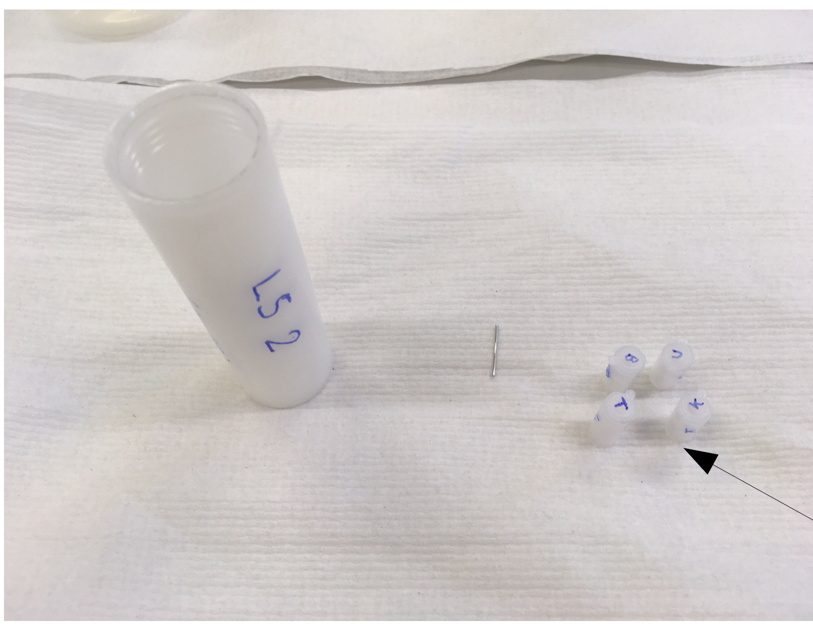
NAA irradiation campaigns @ TRIGA reactor



Teflon container with LAB inside (~9 g)



Teflon blank sample



Irradiation standards (K, Th, U, blank)

NAA irradiations: very preliminary results

LAB bulk contaminations

Analysis still going on,
numbers are not definitive

JUNO requests: ~ 1E-15 g/g	^{40}K [1E-12 g/g]	^{238}U [1E-12 g/g]	^{232}Th [1E-12 g/g]
RAW LAB	0.35 ± 0.09	< 1.2	< 13
Distilled LAB	0.13 ± 0.02	< 0.9	< 6.9

limits @ 90% C.L.

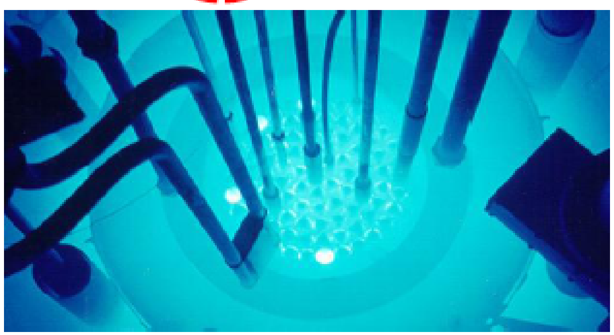
- We have an indication of a small (~10^{-13} g/g) contamination with ^{40}K. May be leaching from the teflon container during neutron irradiation: we will carefully check this with further tests
- We are setting up a new measuring system that exploits beta-gamma coincidences to improve the sensitivity

Coincident detectors for activated samples

A possible way to increase sensitivity…

Neutron Activation on LAB samples

$$^{A}X + n \xrightarrow{\beta^-} {}^{A+1}_{Z+1}Y + \gamma_{cascade}$$



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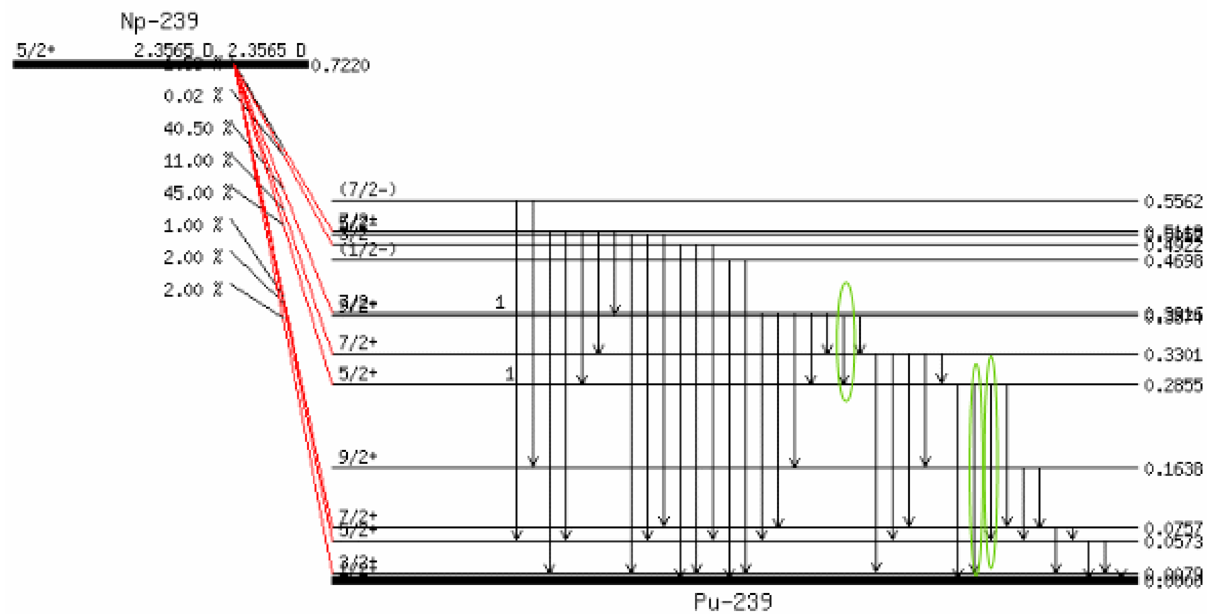
Coincidence measurements on sample activated

γ–γ Ge-Ge detector @MIB

β–γ GeSparK detector @MIB

Strong background reduction



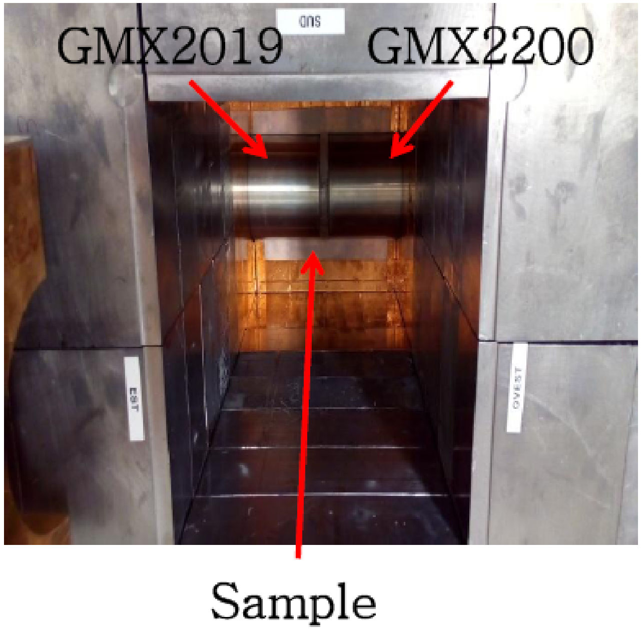
γ – γ coincidence system

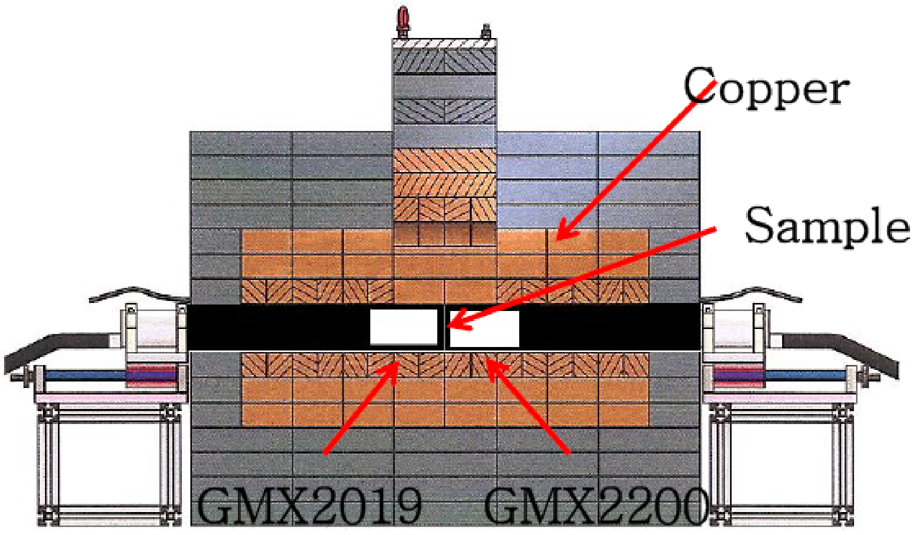
Ge-Ge HPGe: γ-γ detector

2x GMX detector

- Coaxial detector (n-type)
- Relative efficiency: 100%
- Ultra Low Background configuration
- Low Threshold (20keV)

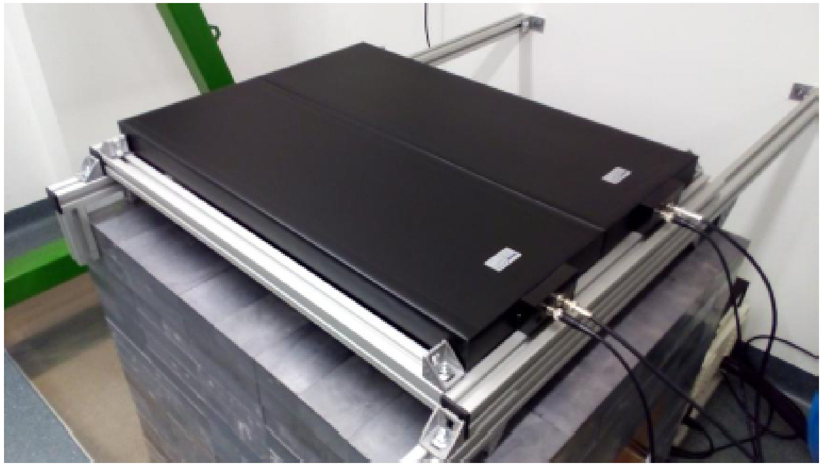
Shielding:
15cm copper
20cm lead





γ – γ coincidence system

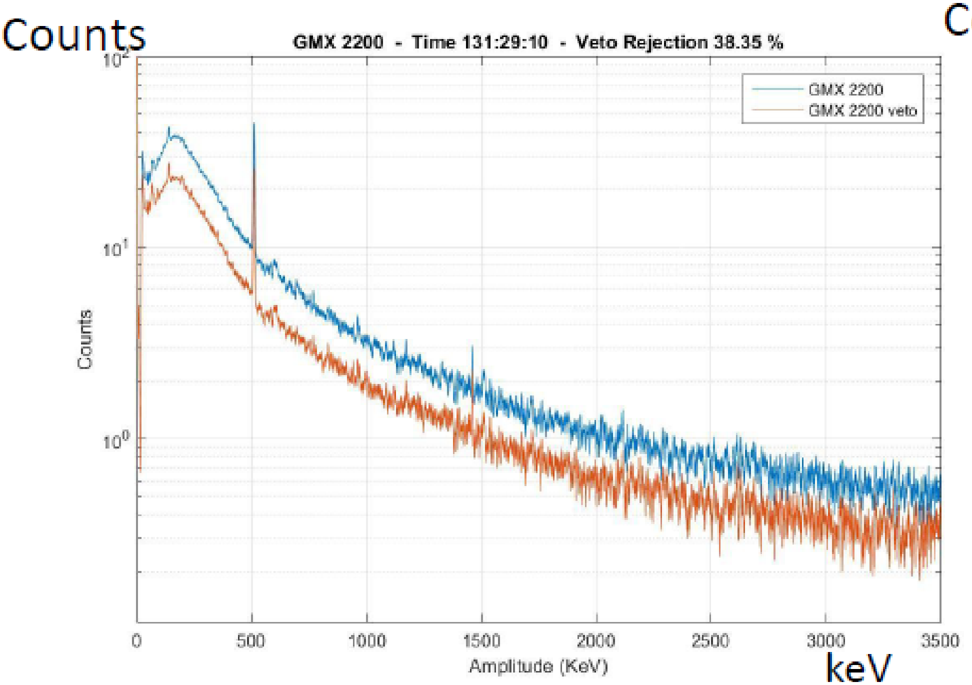
Ge-Ge HPGe: Veto System

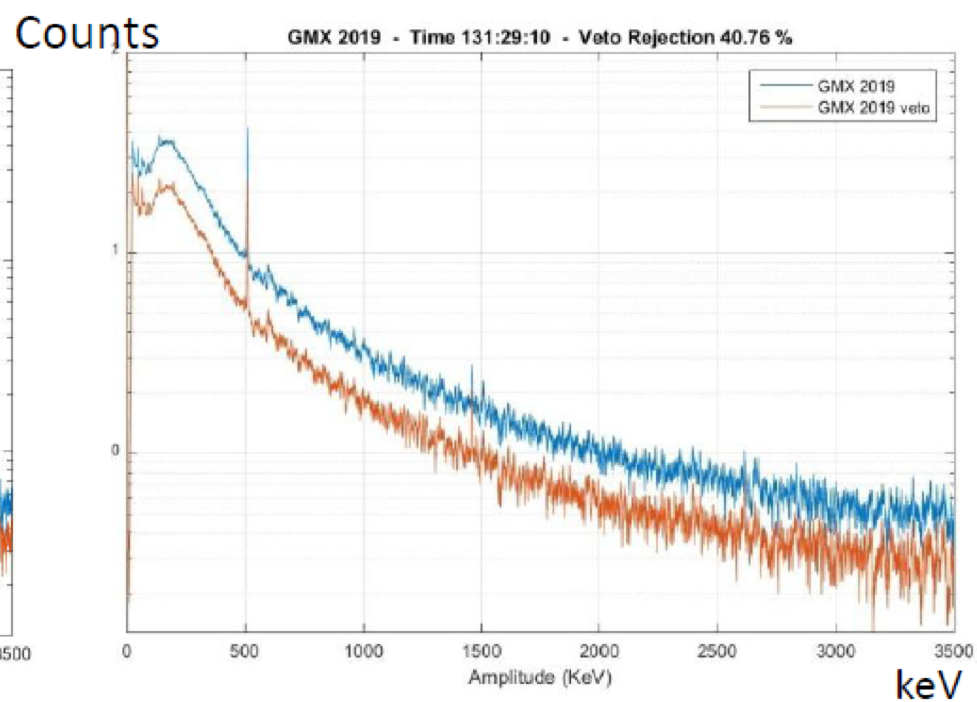


Anticoincidence technique

Plastic scintillator detector

Background suppression **~40%**



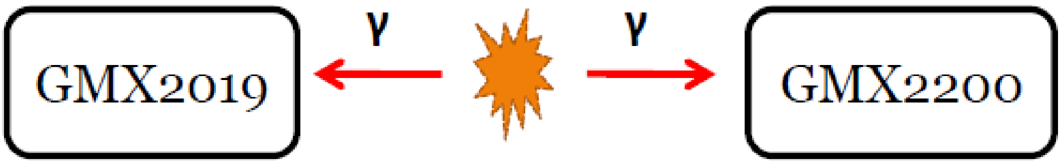


γ – γ coincidence system

Ge-Ge HPGe: γ-γ coincidence

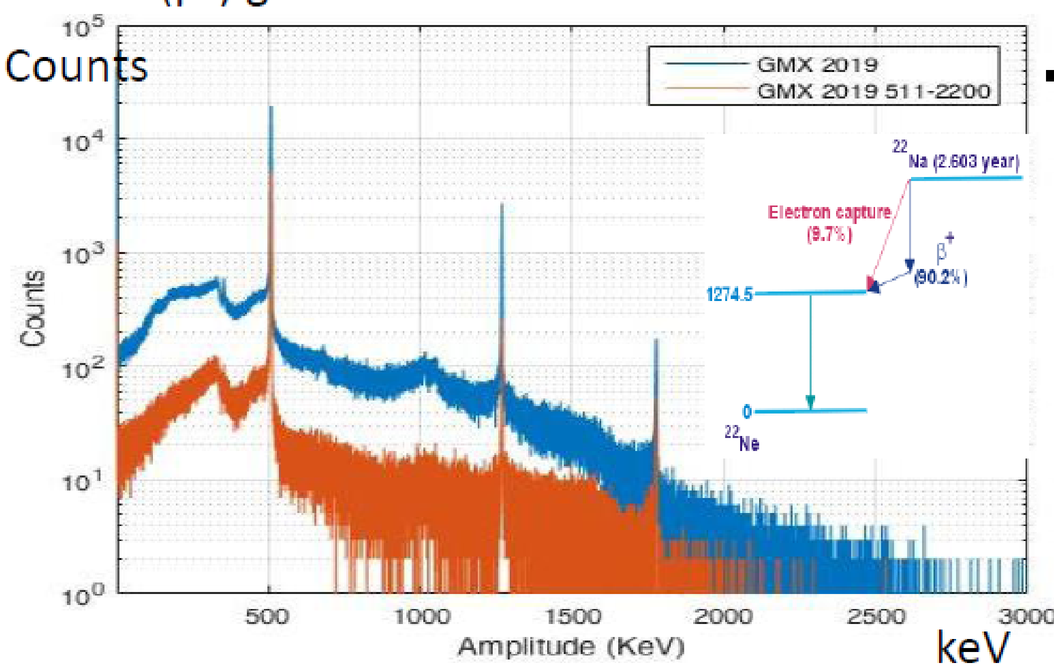
Dedicated acquisition system
allow to detect signals registered in coincidence
considering a very sharp time windows

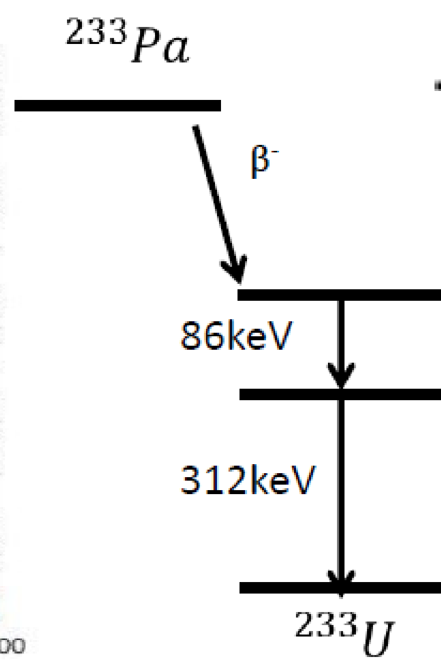
Background Reduction

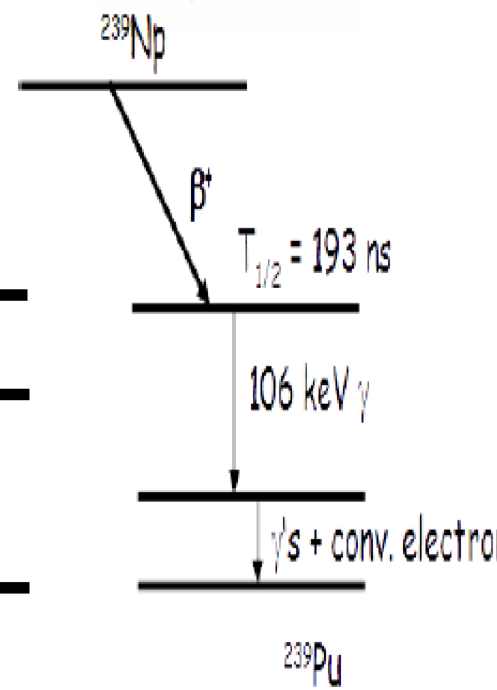


The system is also suitable for **γ cascade measurements** in which the events are not spatially correlated

Coincidence analysis: Source measurements
$^{22}Na(\beta^{+})$ gamma 511keV back to back







β – γ coincidence system

Ge-SparK: β$^-$ - γ detector

Work in progress…

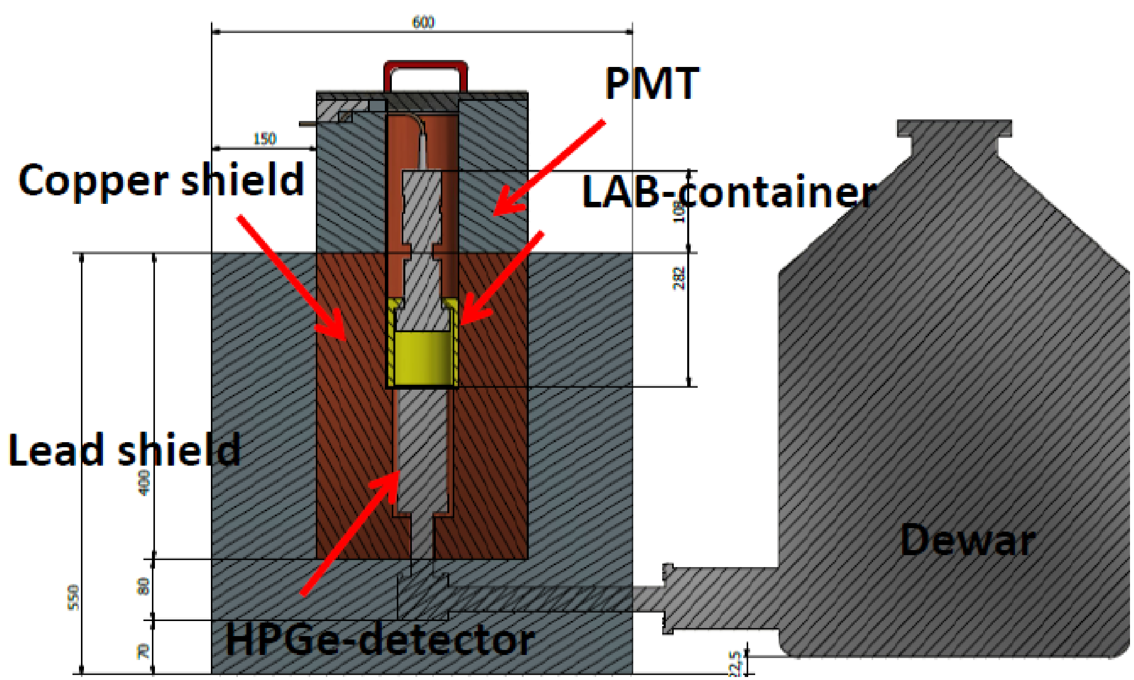
LAB samples will be **exposed** to a neutron flux

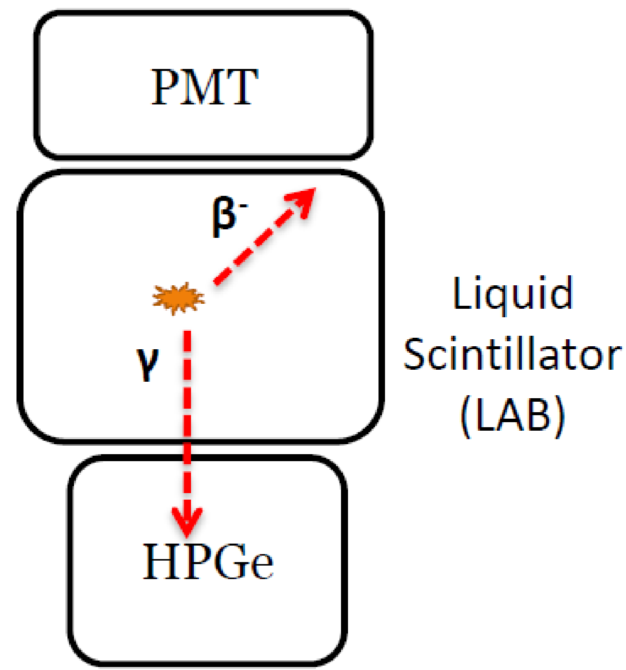
$$^{A}X + n \xrightarrow{\beta^-} {}^{A+1}_{Z+1}Y + \gamma$$

Will be perform **coincidence** measurements on sample activated by **β – γ detector**

We are developing a new detector suitable for this purpose

Preliminary design

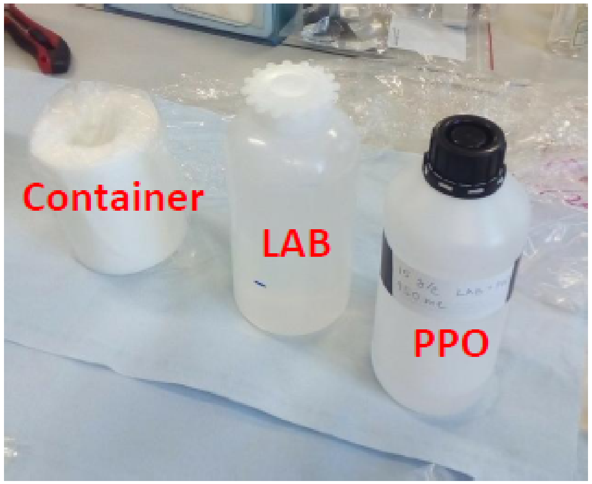


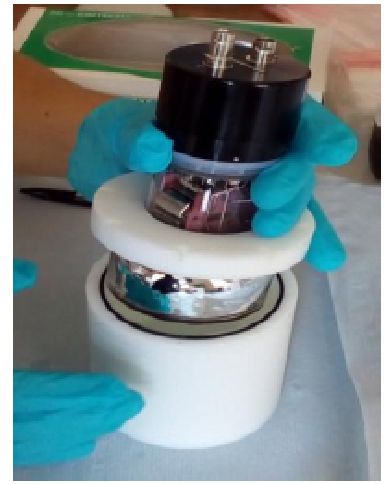


β – γ coincidence system

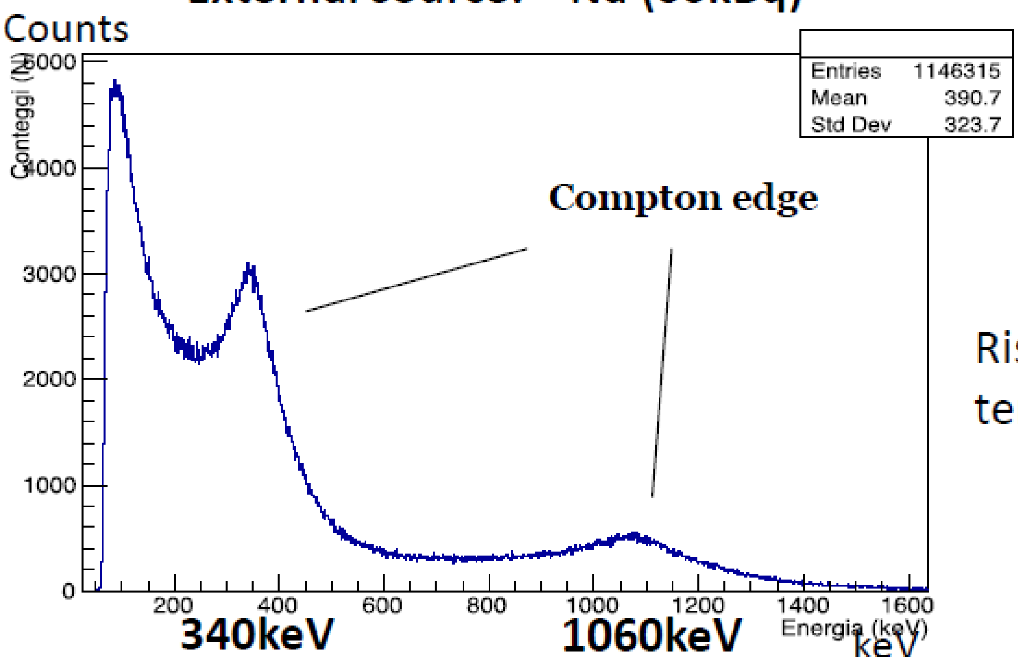
Liquid Scintillator: LAB (Linear Alkyl Benzene)

Hamamatsu R12669



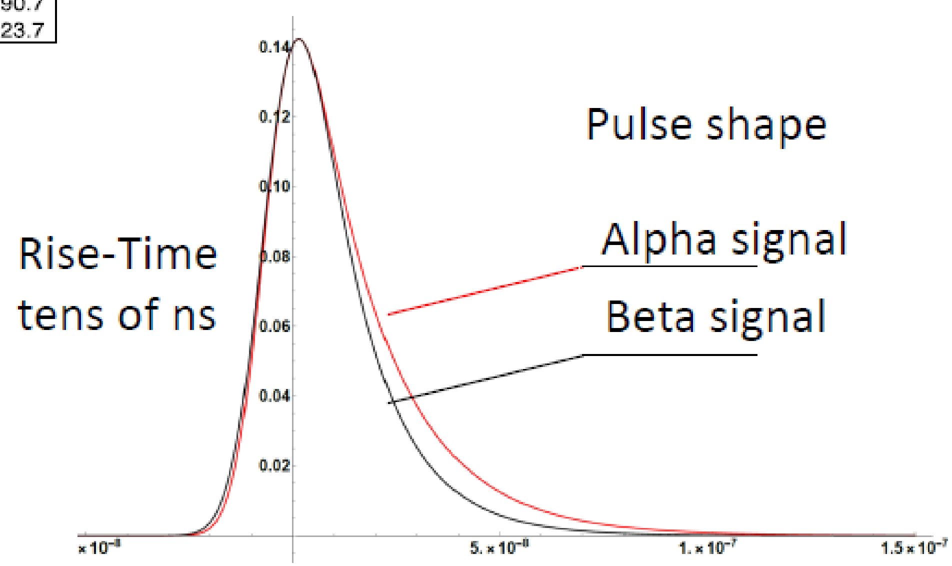


External source: ^{22}Na (60kBq)



Low Density (D=0.86 Kg/cm^3)

Low Atomic Number Z



β – γ coincidence system

HPGe detector setup

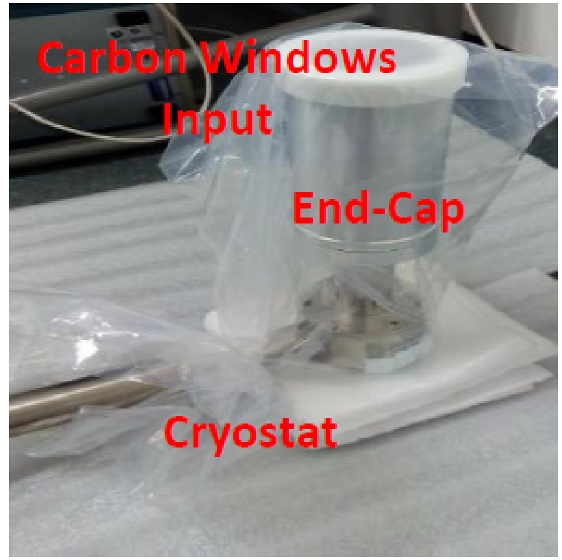
HPGe P-Type

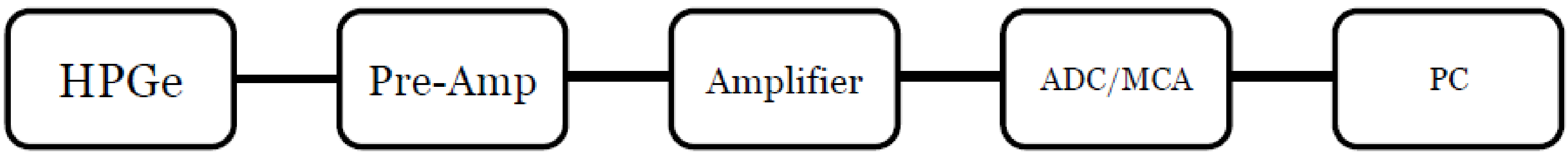
Relative Efficiency: 30%

Cryostat: L configuration

Carbon Window Input

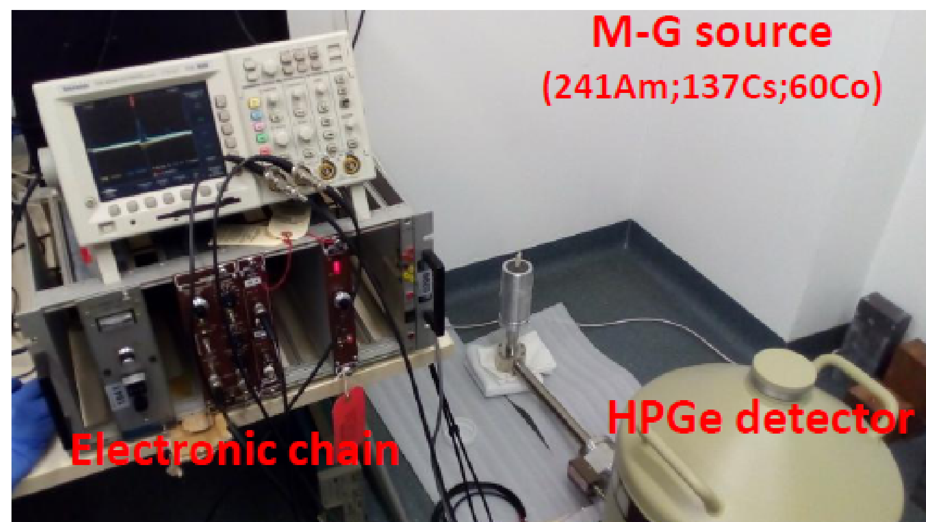
Low Background configuration





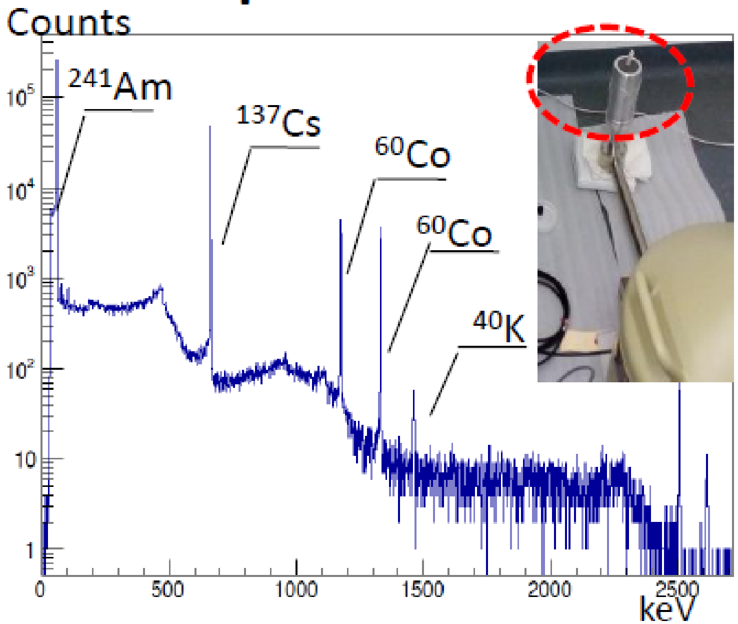
Electronic Chain :

- Amplifier: ORTEC model 672
- H.V.: ORTEC model 659
- ADC/MCA: National Instruments Board 2Gs/s; 2Channel; 64MB/ch

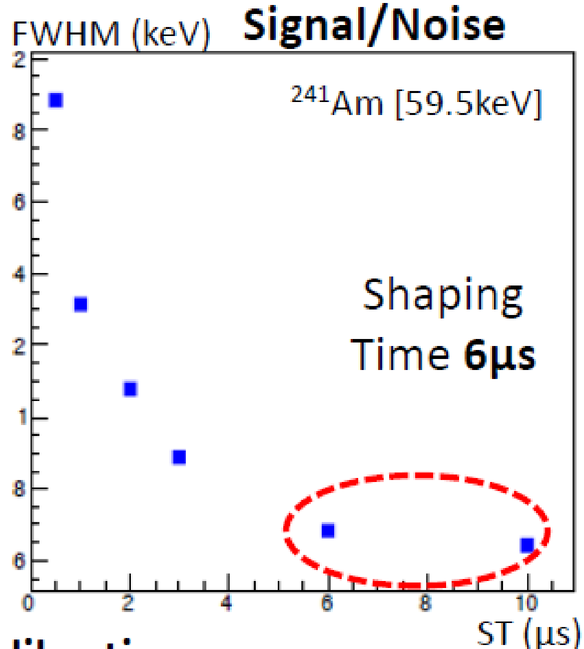


β – γ coincidence system

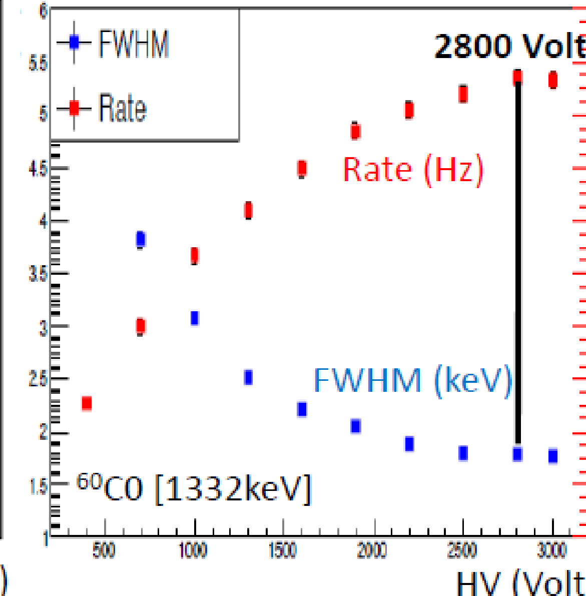
HPGe Optimization



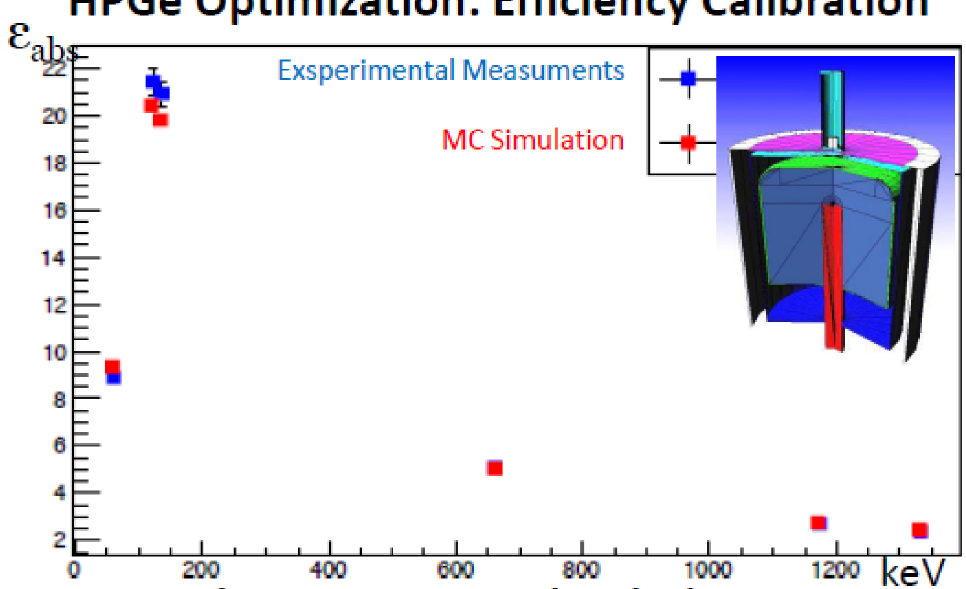
Signal/Noise



Active Volume



HPGe Optimization: Efficiency Calibration

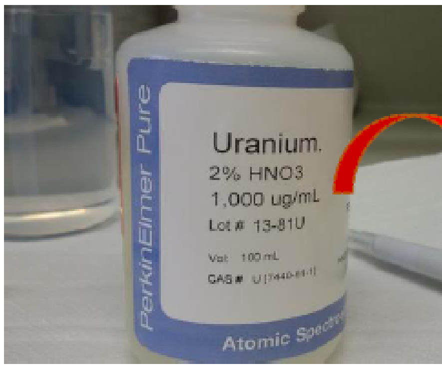


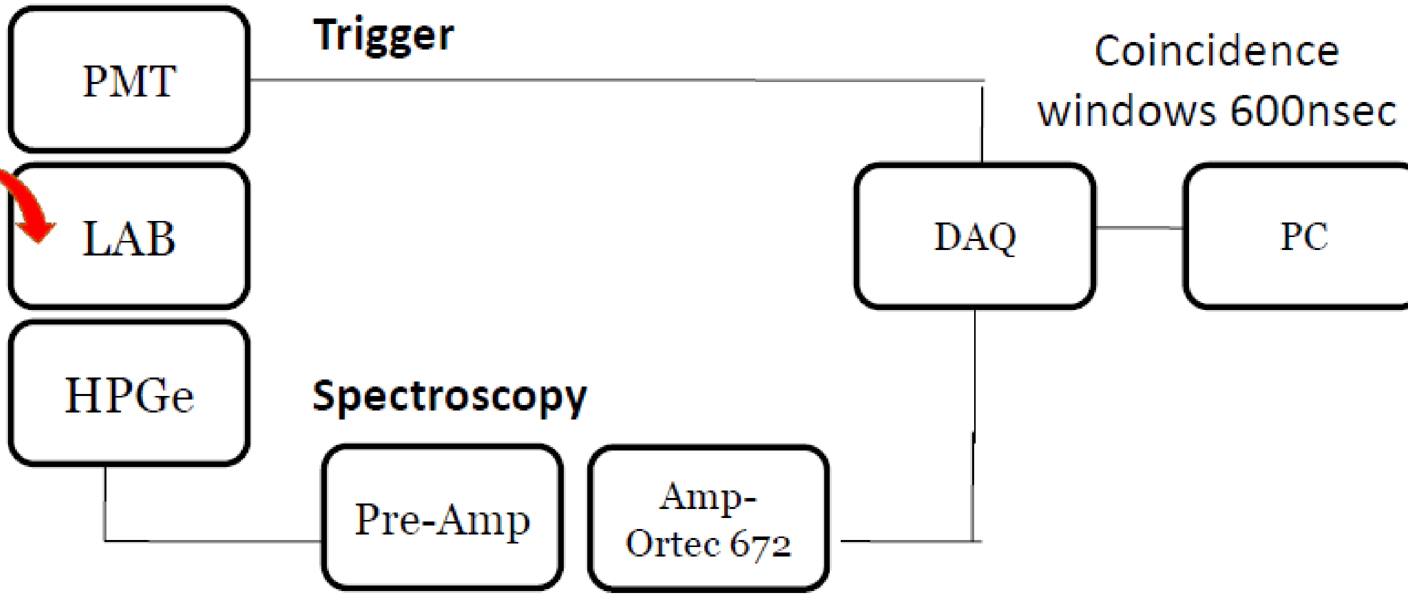
Discrepancy MC simulation < 5%

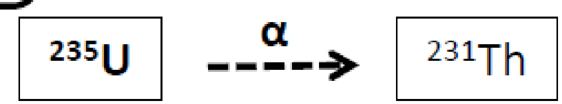
Nuclide	E[keV]	FWHM [keV]	Rate [Hz]
^{241}Am	59.5	0.701 ± 0.002	60.2 ± 0.2
^{57}Co	122	0.760 ± 0.002	107.8 ± 0.3
^{57}Co	136	0.800 ± 0.003	13.4 ± 0.1
^{137}Cs	661	1.28 ± 0.01	17.5 ± 0.1
^{60}C0	1173	1.66 ± 0.02	1.92 ± 0.04
^{60}Co	1332	1.79± 0.02	1.74 ± 0.03

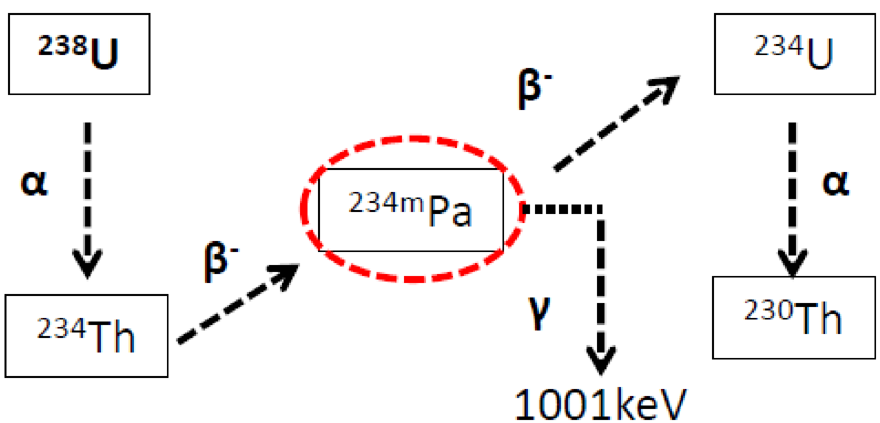
β – γ coincidence system

Coincidence measurement: STD Uranium







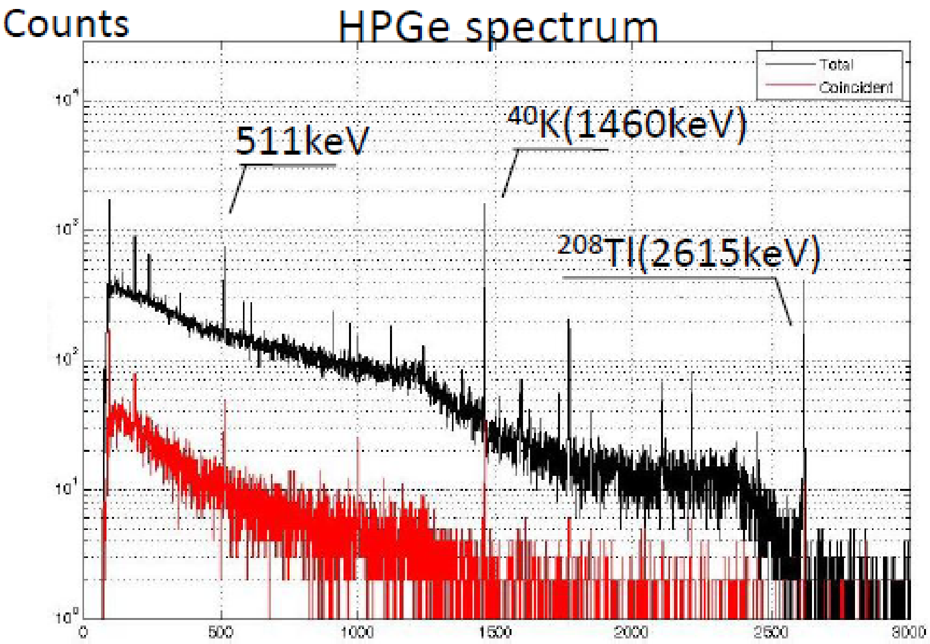


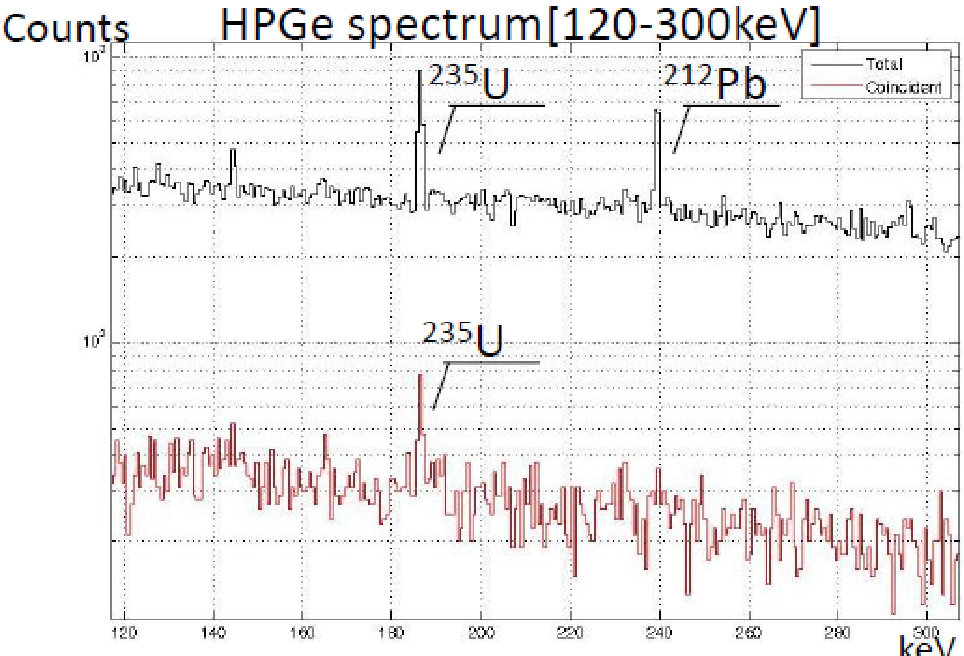
^{235}U-gamma	B.R.(%)
109 keV	1.54
143keV	10.96
163keV	5.08
185keV	57.2
205keV	5.01

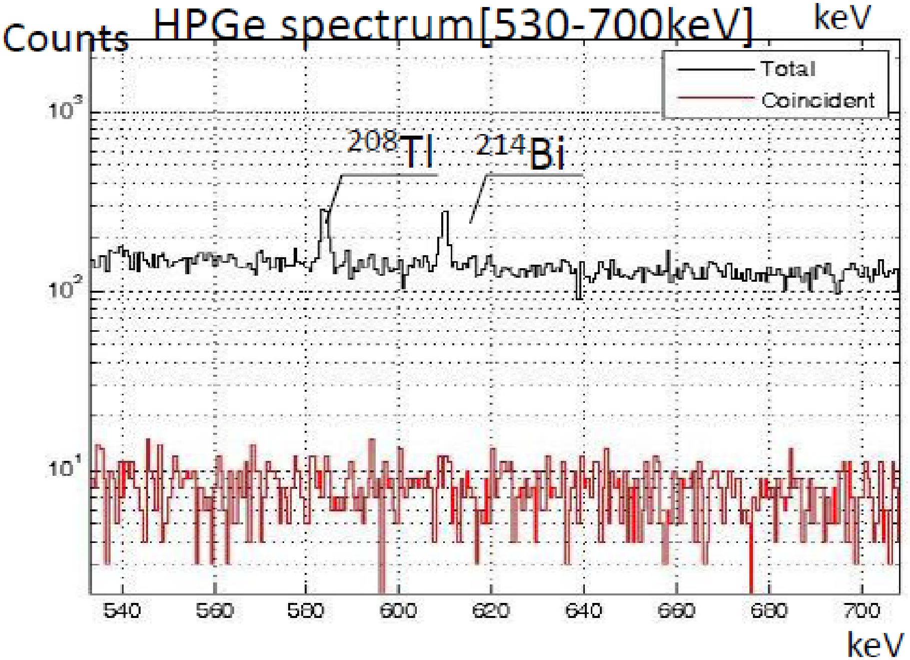
β – γ coincidence system

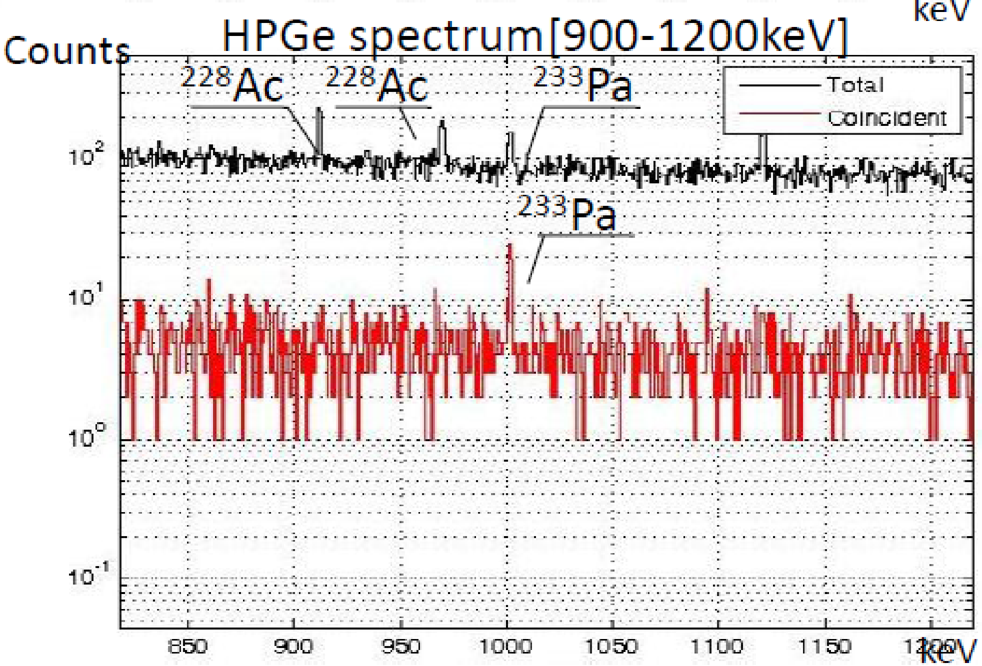
Preliminary results...

Coincidence measurement



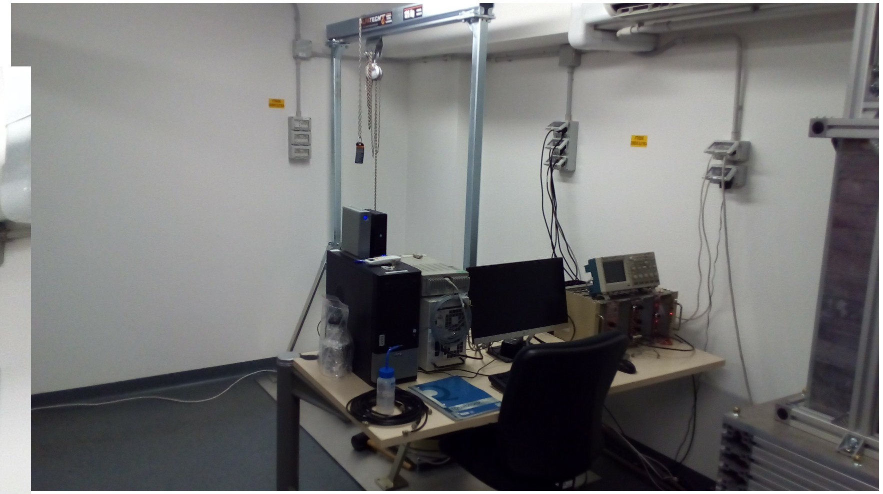


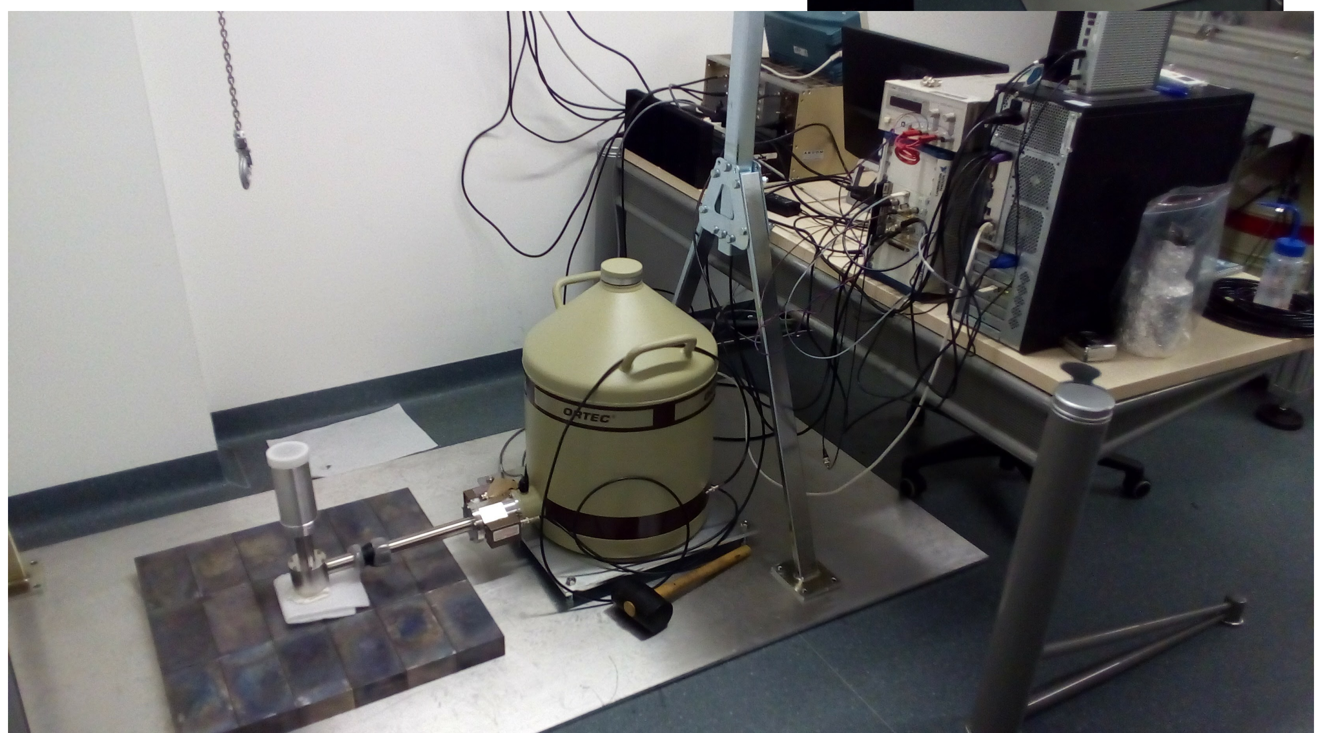




β – γ coincidence system

Mainly hardware work in the last month





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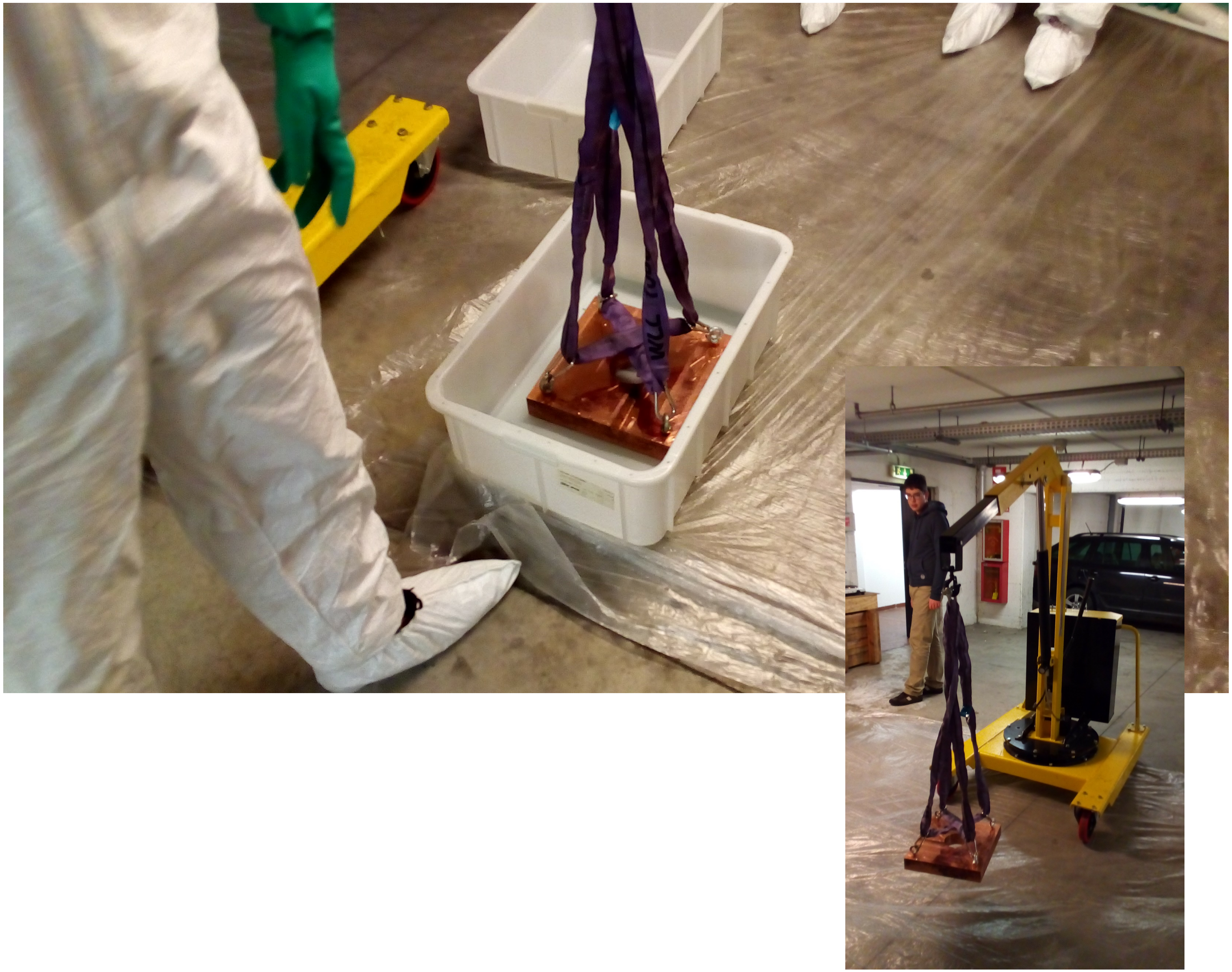
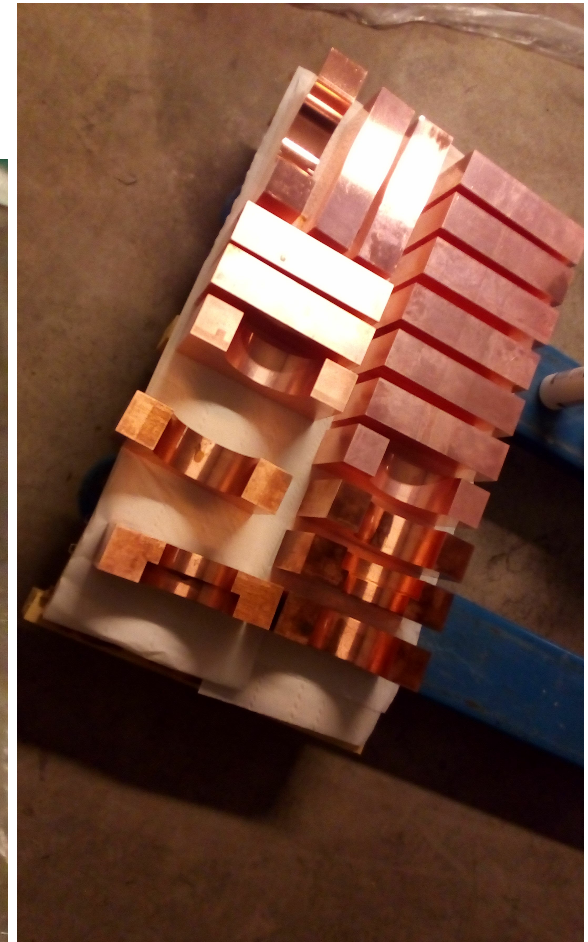




Radon implantation measurements

Sealed Plexiglass box saturated with Rn gas

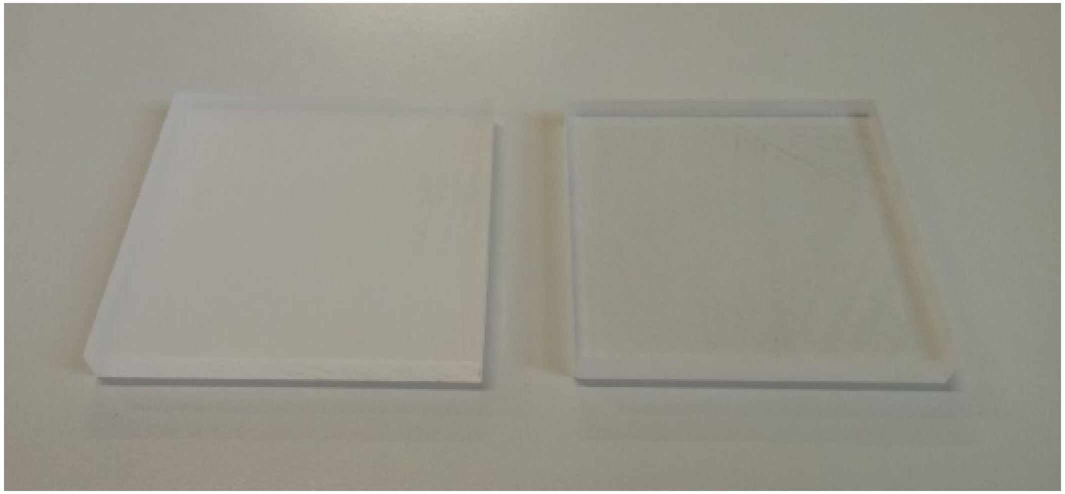
to study different surface protection methods to avoid Rn implantation (in particular on Acrylic)

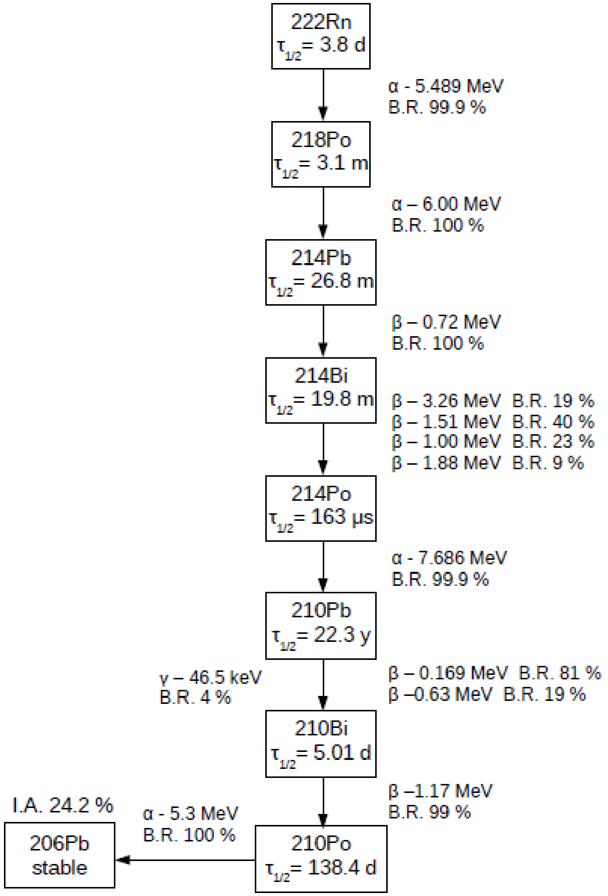
1 Si-barrier surface detector dedicated to these studies

$$A_{^{222}Rn} = (250 \pm 5)\ kBq/m^3$$

Radon implantation measurements

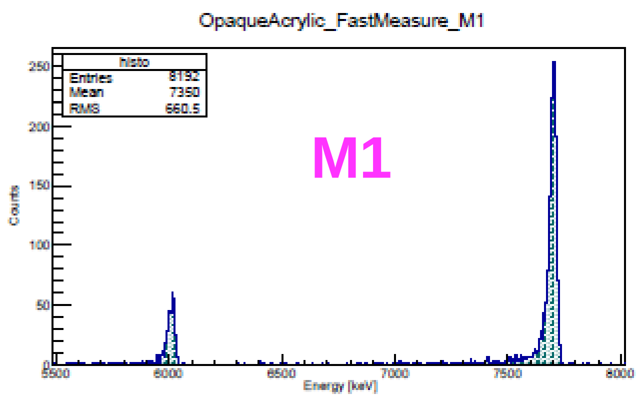
Sample	Exposure time [days]
Acrylic Opaque 1	29
Acrylic Smooth 1	86
Acrylic Opaque 2	72
Acrylic Opaque 3	90
Acrylic Opaque 4	39

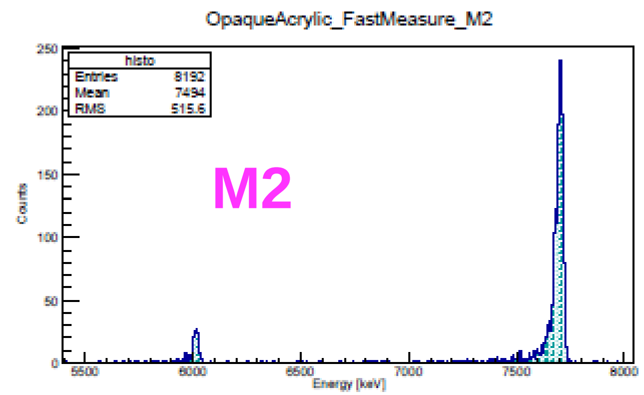


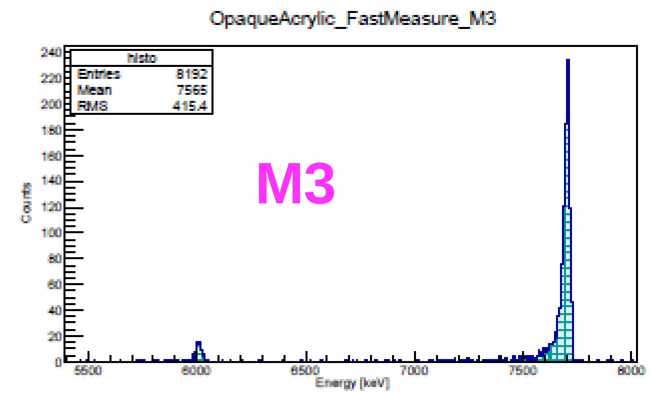


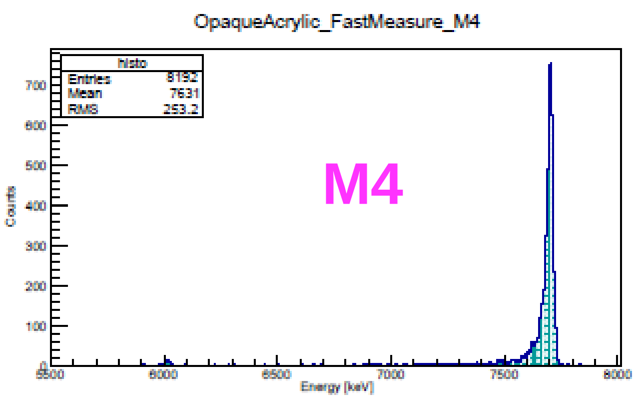
	Duration [min]	Observed isotopes
Measurement 1 (M1)	3	^{218}Po, ^{214}Po
Measurement 2 (M2)	3	^{218}Po, ^{214}Po
Measurement 3 (M3)	3	^{218}Po, ^{214}Po
Measurement 4 (M4)	10	^{214}Po
Measurement 5 (M5)	10	^{214}Po
Measurement 6 (M6)	20	^{214}Po
Measurement 7 (M7)	20	^{214}Po
Measurement 8 (M8)	20	^{214}Po
Measurement 9 (M9)	40	^{214}Po
Measurement 10 (M10)	40	^{214}Po
Measurement 11 (M11)	40	^{214}Po
Long Measurement (LM)	see Table 2.3	^{210}Po

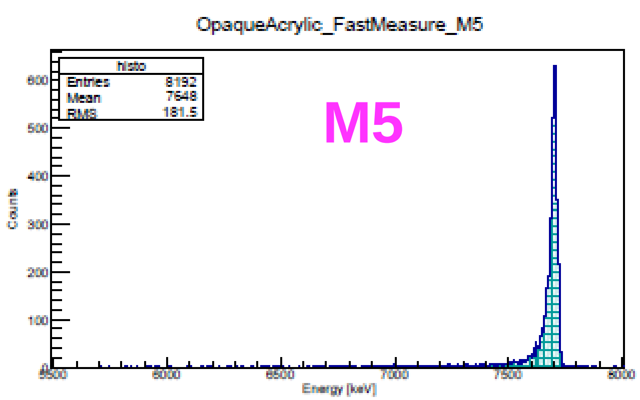
Radon implantation measurements











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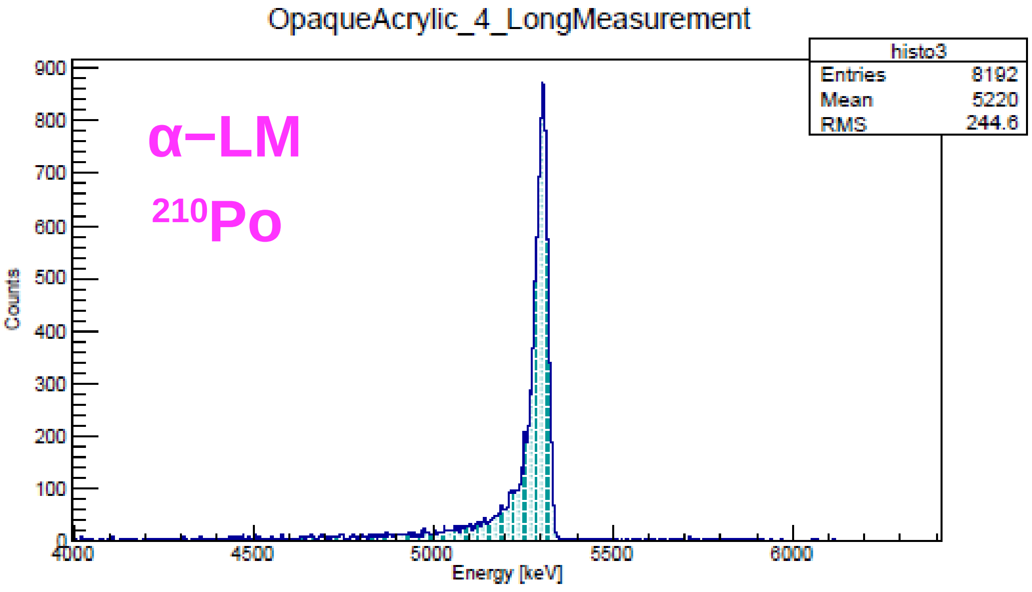
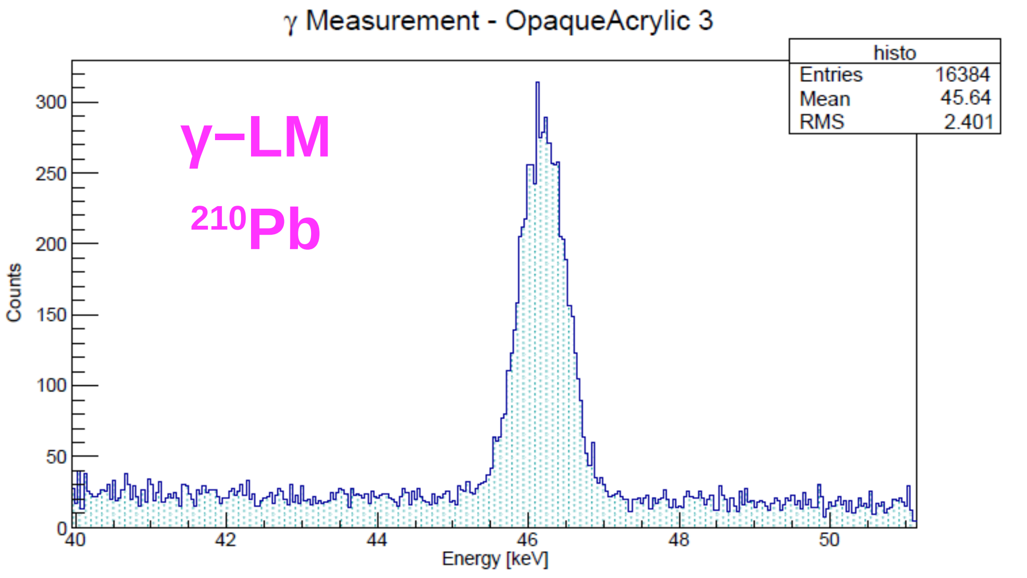
Measurement 5 (M5)	10	^{214}Po
Measurement 6 (M6)	20	^{214}Po
Measurement 7 (M7)	20	^{214}Po
Measurement 8 (M8)	20	^{214}Po
Measurement 9 (M9)	40	^{214}Po
Measurement 10 (M10)	40	^{214}Po
Measurement 11 (M11)	40	^{214}Po
Long Measurement (LM)	see Table 2.3	^{210}Po

Radon implantation measurements





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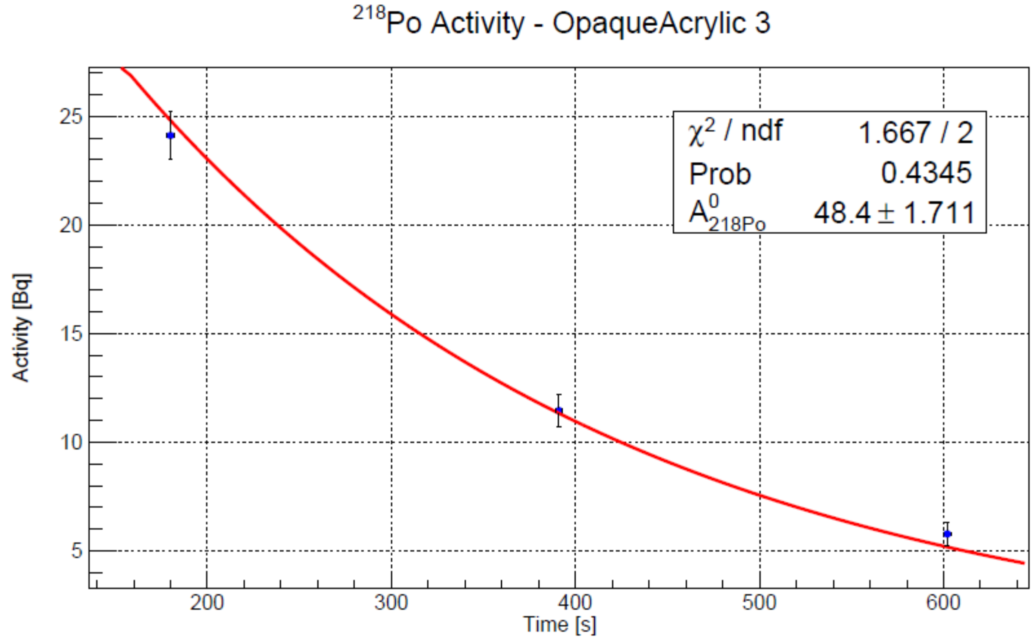
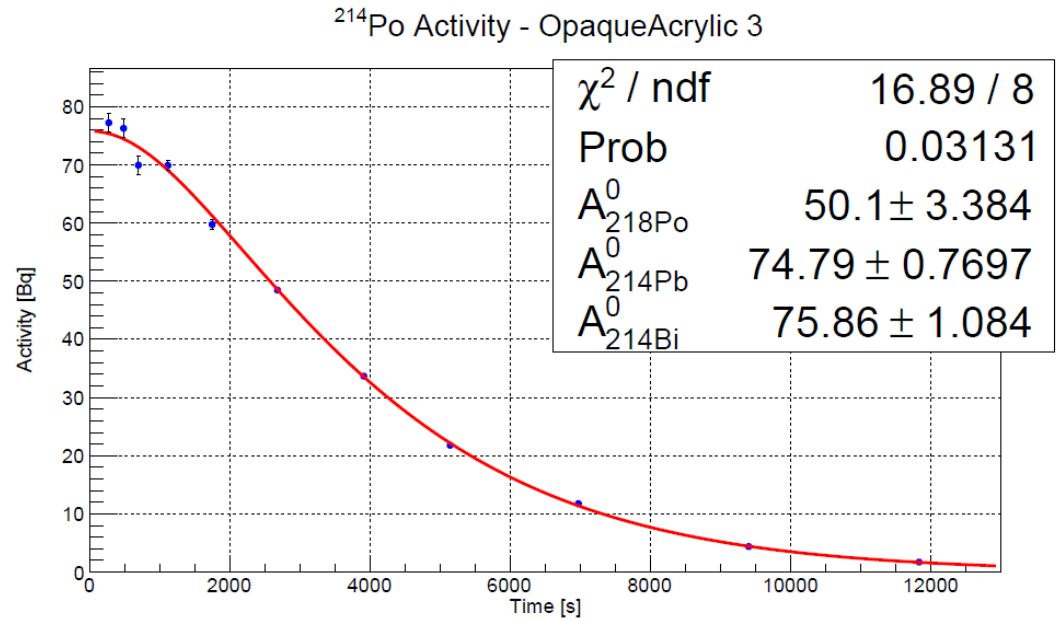
Measurement 5 (M5)	10	^{214}Po
Measurement 6 (M6)	20	^{214}Po
Measurement 7 (M7)	20	^{214}Po
Measurement 8 (M8)	20	^{214}Po
Measurement 9 (M9)	40	^{214}Po
Measurement 10 (M10)	40	^{214}Po
Measurement 11 (M11)	40	^{214}Po
Long Measurement (LM)	see Table 2.3	^{210}Po

Radon implantation measurements





Sample	A^0_{218Po} [Bq]	$\mathrm{Err}_{A^0_{218Po}}$ [Bq]
Opaque Acrylic 1	26.9	1.4
Smooth Acrylic	46.8	2.0
Opaque Acrylic 2	37.2	1.8
Opaque Acrylic 3	48.4	2.0
Opaque Acrylic 4	42.9	1.9

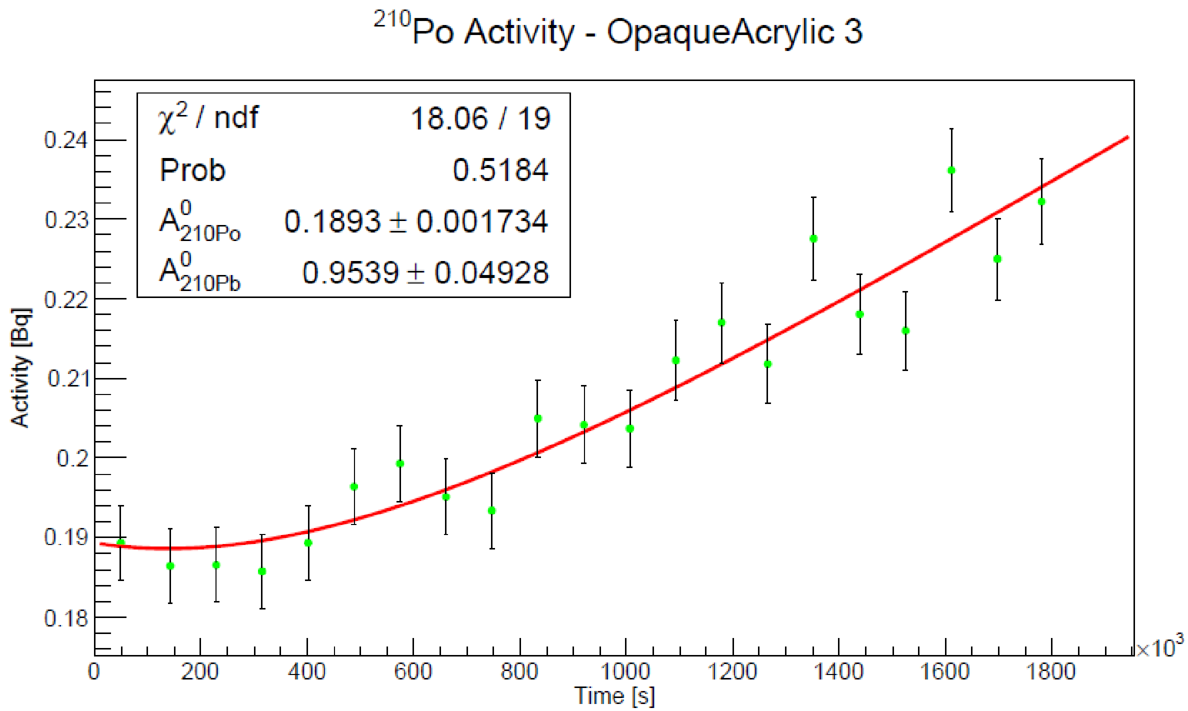
from ^{218}Po data

from ^{214}Po data

Sample	A^0_{218Po} [Bq]	A^0_{214Pb} [Bq]	A^0_{214Bi} [Bq]
Smooth Acrylic	48 ± 3	79.8 ± 0.8	80 ± 1
Opaque Acrylic 2	38 ± 1	54.3 ± 0.7	55.7 ± 0.9
Opaque Acrylic 3	50 ± 3	74.8 ± 0.8	76 ± 1
Opaque Acrylic 4	44 ± 3	65.7 ± 0.7	68 ± 1

Radon implantation measurements

^{210}Po data



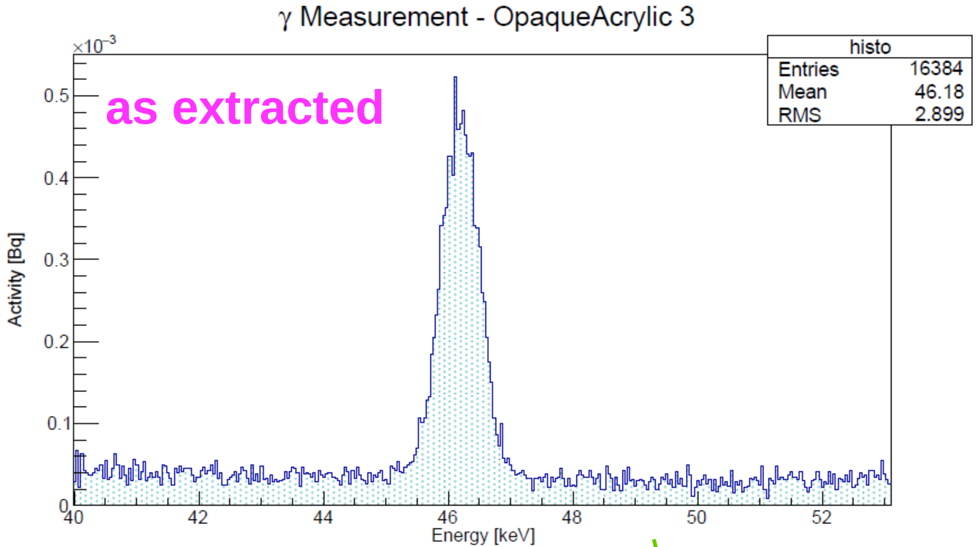
$$TPR_{210Po} = \frac{\text{N. counts }(5.0 - 5.2\text{MeV})}{\text{N. counts }(5.2 - 5.4\text{MeV})} \cdot 100$$

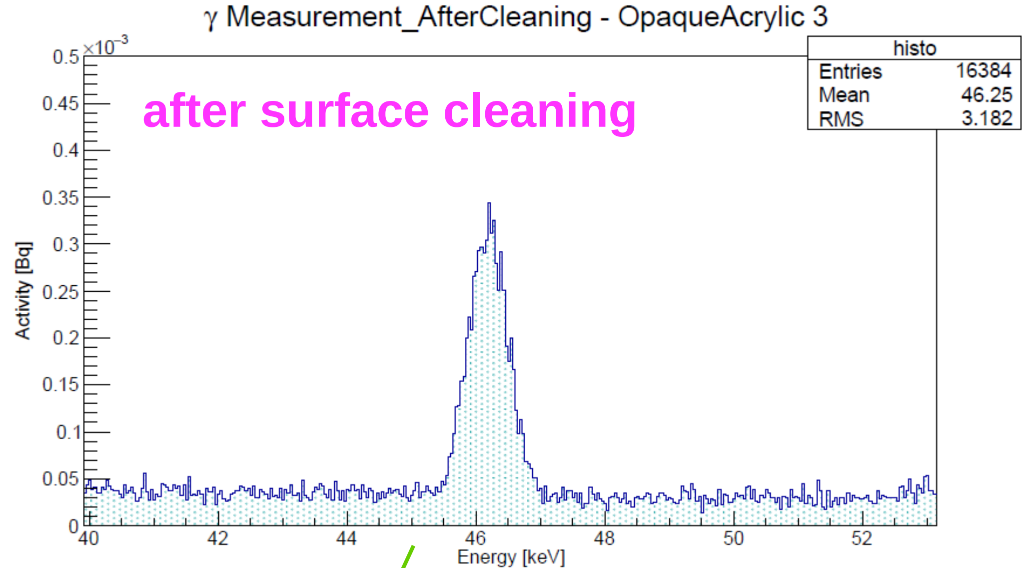
Tail-to-peak ratio for the 3 α peaks: ^{210}Po implantation is deeper

Sample	TPR_{210Po} [%]	TPR_{214Po} [%]	TPR_{218Po} [%]
Smooth Acrylic	3.9 ± 0.1	3.40 ± 0.09	2.0 ± 0.5
Opaque Acrylic 2	13.7 ± 0.4	7.9 ± 0.2	6.6 ± 1.1
Opaque Acrylic 3	11.4 ± 0.2	6.7 ± 0.1	7 ± 1
Opaque Acrylic 4	14.3 ± 0.5	8.4 ± 0.2	7 ± 1

Radon implantation measurements

^{210}Pb γ–measurements





S.F.=surface fraction

Sample	Total activity [Bq]	Deep activity [Bq]	S.F. [%]	D.F. [%]
Smooth Acrylic	0.61 ± 0.03	0.40 ± 0.02	35 ± 7	65 ± 5
Opaque Acrylic 2	0.68 ± 0.04	0.49 ± 0.03	27 ± 7	73 ± 5
Opaque Acrylic 3	0.89 ± 0.05	0.56 ± 0.03	37 ± 7	63 ± 5
Opaque Acrylic 4	0.38 ± 0.02	0.25 ± 0.02	34 ± 7	66 ± 6

D.F.=deep fraction

Conclusions

- By means of the coincident measuring systems we should approach the zero-background condition

In this way we hope to reach the 10^{-14}–10^{-15} g/g sensitivity requested for the LAB radiopurity

- Radon implantation measurements show that dust control during acrylic sphere mounting is crucial to avoid ^{210}Pb of the LAB

➡ Also leaching from acrylic to LAB may be a problem?