University of Milano Bicocca - Dep. of Physics
and
INFN of Milano Bicocca

Ultra-trace element determination by neutron activation analysis in Acrylic material

Low Radioactivity Techniques 2017
Ewha Womans University, Seoul, Korea

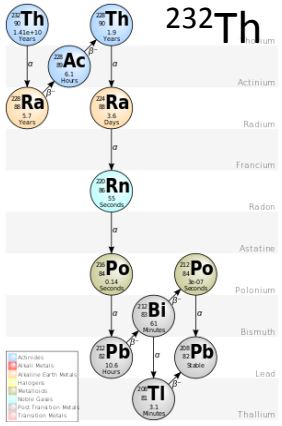
Introduction

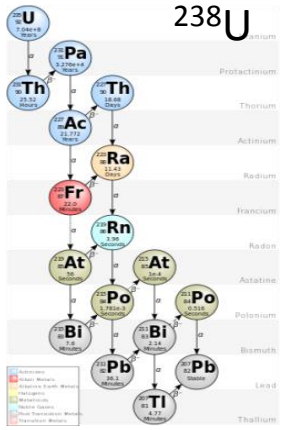
The **radio purity of materials is the essential condition** upon which rely the latest experiments of Rare Events Physics

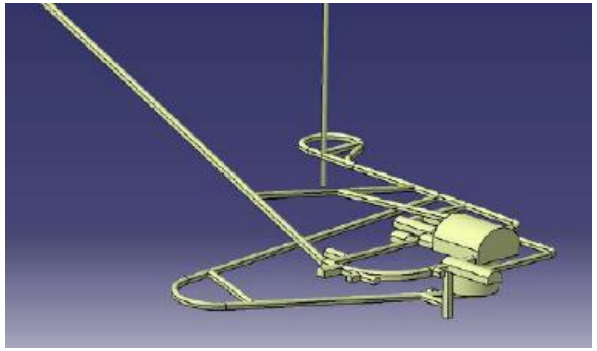
The greatest risk

radioactive background
overlap the observable energy regions of interest

Fundamental a selection of the different components of the detector







Typical requirements for contamination level of detector materials

^{232}Th $< 10^{-12}$ g/g (4 uBq/kg)
^{238}U $< 10^{-12}$ g/g (12 uBq/kg)
$^{40}K < 10^{-12}$ g/g (270 uBq/kg)

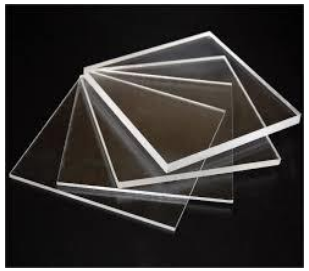
Crucial to have
tools able to achieve that sensitivity

Acrylic material

Acrylic is a widely used material in many experiments in the physics of rare events

optical properties

mechanical properties



Jiangmen Underground Neutrino Observatory

Inverse Beta DecayReaction

$\bar{\nu} + p \longrightarrow e^+ + n$

Delayed signal (~ 200μs) **2.2MeV ϒ-ray**

JUNO Central Detector

~20 kton LS detector

Acrilic Vessel

12cm thickness

Diameter: 35.5

It is crucial to carefully select acrylic material at ~ppt level of contamination (^{238}U,^{232}Th, ^{40}K) in order to reduce the accidental rate in the active volume of the detector

Acrylic samples

Samples received from China from **3 different companies**

Each company has produces the samples from sheets of 2 cm or 12 cm thickness

Company A
2 cm sheet production:
7 samples 248×248×20 mm3

Company B1
from 12 cm sheet production (panel surface):7 samples 248×248×20 mm3

Company B2
from 12 cm sheet production (panel centre):
7 samples 248×248×20 mm3

Company C
from 12 cm sheet production (panel centre):
7 samples 248×248×20 mm3

Available radioassay techniques @INFN-MIB

HPGe Spectroscopy*

Sensitivity on Th and U: ~ 10 mBq/kg

Bulk contaminations

γ emitting nuclides

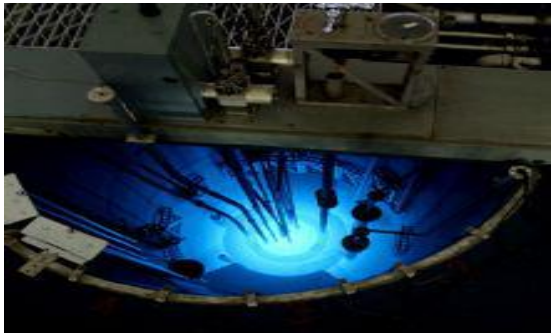
Large amount of materials: few tens of kg

Alpha spectroscopy*

Sensitivity: 10^{-6}Bq/cm^2

α emitting nuclides

Surface contaminations



Neutron Activation Analysis

Sensitivity on Th and U : ~ **uBq/kg**

Bulk contaminations

Primordial parents

Sample of few tens of g

*needs counting times from several weeks to months

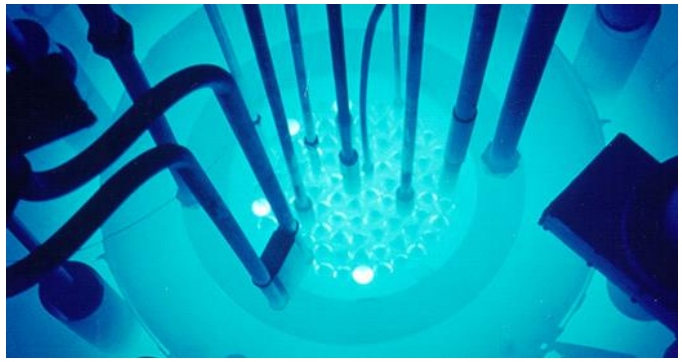
Neutron Activation Analysis (NAA)

The neutron activation process consists in the production of unstable isotopes through neutrons absorption by the nuclei present in the sample

$$ {}^{A}X + \mathrm{n} \rightarrow {}^{A+1}X \xrightarrow[\beta^-]{} {}^{A+1}_{Z+1}Y^{*} \rightarrow {}^{A+1}_{Z+1}Y + \gamma $$

The NAA technique consists of several steps:

Sample **exposure** to a neutron flux



Extraction of the irradiated sample and **measurement** of induced ϒ radioactivity (HPGe- detector)

Calculation of the **quantity of precursor** element (${}^{A}X$)

NAA key parameters:

Reaction cross section

Efficiency HPGe-detector

Neutron Flux

Irradiation Time

γ emission and BR

Neutron Activation Analysis

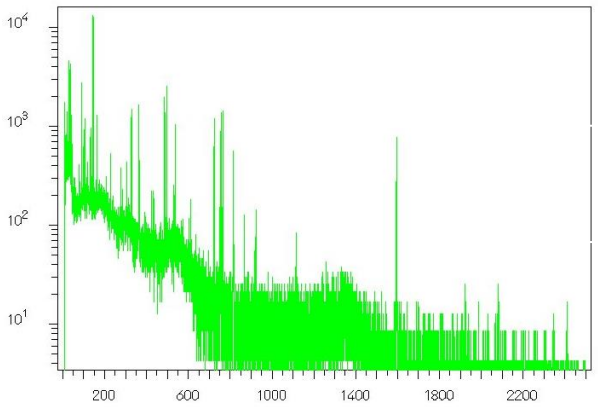
Comparative method

Sample and a reference Standard are prepared and irradiated → STD containing a known amount of the element to be measured

- Time of irradiation is the same → $T_{irr-Sample} = T_{irr-STD}$
- Same position in the irradiation channel → Neutron Flux $\phi_{Sample} = \phi_{STD}$
- $Geometry_{Sample} = Geometry_{STD}$ → **Gamma spectroscopy - HPGe**

Geometry and size must be similar

$Efficiency_{Sample} = Efficiency_{STD}$



The activities of samples and standards are compared → **Sample trace analysis**

Neutron Activation Analysis

Three key ingredients:

high neutron flux $\phi \approx 10^{12} \div 10^{13} cm^{-2} s^{-1}$

high enough neutron capture cross section $^{A}X + \mathrm{n} \longrightarrow {}^{A+1}_{Z+1}Y + \gamma$

"convenient" daughter nucleus (γ emission, half-life time)

$$n + {}^{41}K \longrightarrow {}^{42}K \xrightarrow[12{,}36\,h]{\beta^-} {}^{42}Ca \quad +\gamma(1524keV\ -BR\ 17\%)$$

$$n + {}^{238}U \longrightarrow {}^{239}U \xrightarrow[23{,}5\,m]{\beta^-} {}^{239}Np \xrightarrow[2{,}36\,d]{\beta^-} {}^{239}Pu + \gamma(106keV\ -BR26\%)$$

$$n + {}^{232}Th \longrightarrow {}^{233}Th \xrightarrow[22{,}3\,m]{\beta^-} {}^{233}Pa \xrightarrow[27{,}0\,d]{\beta^-} {}^{233}U + \gamma(311keV - BR38\%)$$

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!**

Advantage:

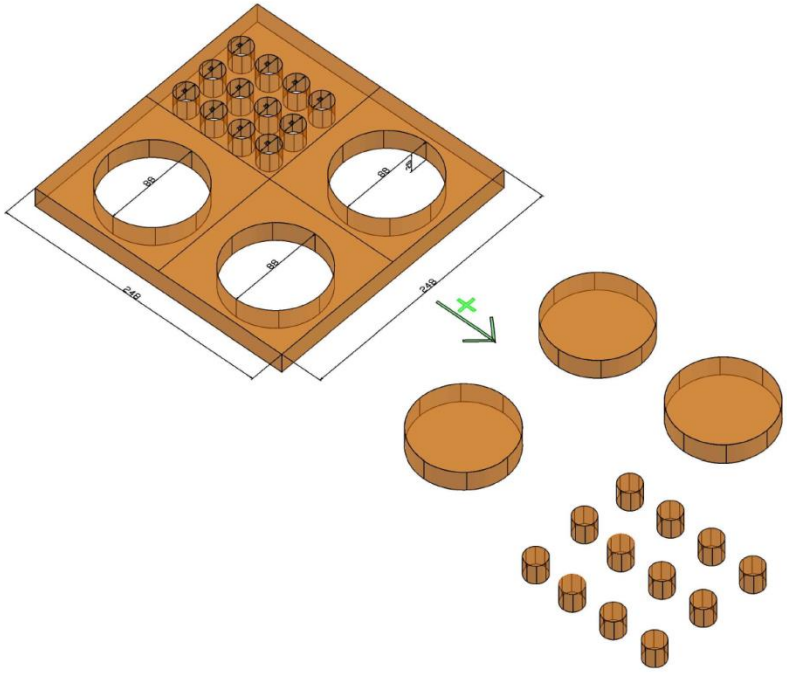
very high sensitivity :~ **1ppt**

Disadvantage:

sensitive only to radioactive chain progenitors

Sample preparation: Laser cut

To avoid the risk of contaminating the samples, we used the laser cutting technique

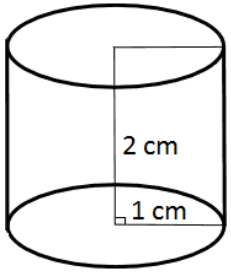


Extreme care was put to avoid contact of the samples with dirty surfaces

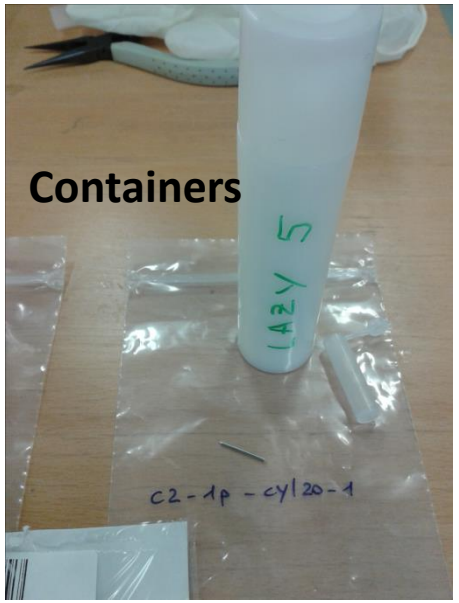
Sample preparation in clean room

Acrylic samples washed several times in US bath (30°C) with MilliQ water in clean room 1000 atmosphere

Acrilyc sample



Irradiation containers cleaned with ultra pure nitric acid solution (1%) in clean room 1000 atmosphere



STD preparation

Liquid Standard (1mg/ml)

3 Vials for each STD (0.02ml)

3 Vials blank with water

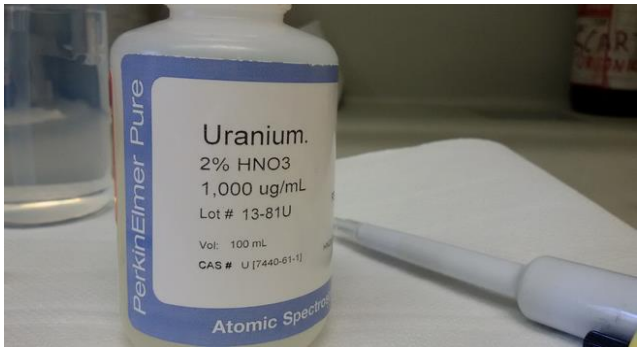
Liquid standards prepared in separate containers from samples

The vials were sealed and washed

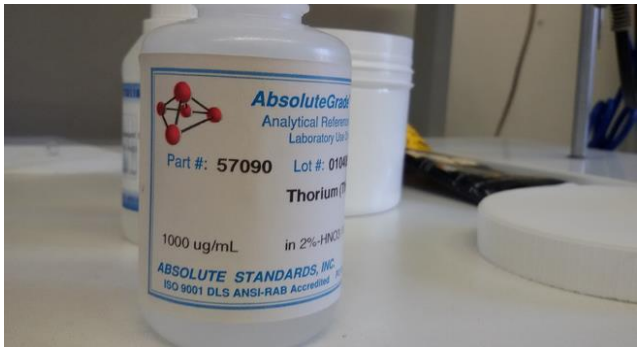
Solid Standard:

Al-Co (Co 0.5%wt) wire standards put in every irradiation channel as a cross-check of the results

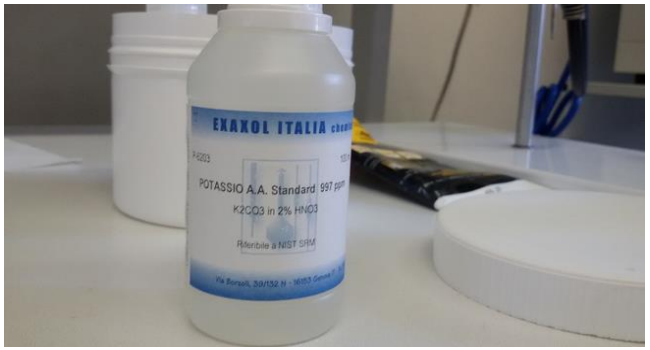
Uranium



Thorium

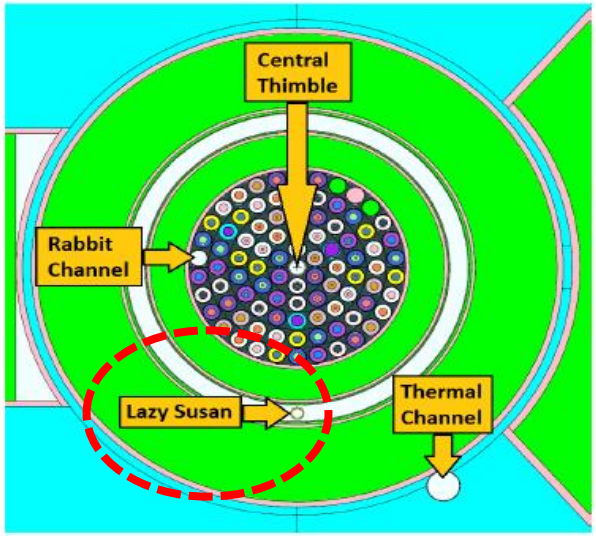


Potassium



NAA irradiation campaigns

TRIGA Mark II



Research reactor (250 kW) - Pavia, Italy

LAZY SUSAN facility:
Flux of neutrons: $\approx 10^{12} cm^{-2} s^{-1}$

Irradiation Time: 12 hours

LAZY SUSAN facility is a rotary specimen rack in a circular well within the radial reflector

40 irradiation channels available around the reactor core, 7 adjacent channels were used

Irradiation Channel	LS1	LS2	LS3	LS4	LS5	LS6	LS7
Sample/STD liquid	A	STD2	B1	STD4	B2	STD6	C
STD solid	SS1	SS2	SS3	SS4	SS5	SS6	SS7

STD solid and samples were in the same channel

STD liquid and samples were in different channels to avoid contamination

Gamma spectroscopy

HPGe GePV

Beryllium Window

Relative efficiency: 25%

Coaxial geometry

Three detectors in Low background configuration

HPGe GeGEM

Coaxial geometry

Relative efficiency: 30%

HPGe GeKan

Coaxial geometry

Relative efficiency: 70%

HPGe BeGE

Planar geometry

Relative efficiency: 50%

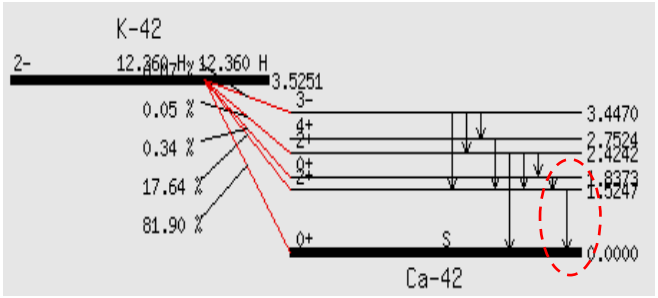
Carbon Window

Broad Energy detector

NAA results: ^{40}K

$$n + {}^{41}K \longrightarrow {}^{42}K \xrightarrow{\beta^-} {}^{42}Ca$$

$\tau = 12{,}36\,h$



IsotopicAbbundace$K$:
$K^{39} \rightarrow \sim 93\%$
$K^{40} \rightarrow \sim 0{,}01\%$
$K^{41} \rightarrow \sim 7\%$

ϒ-ray(keV)	BR(%)
312.6	0.34
1524.6	17.64

Untreated samples

Sample	^{40}K(ppt)
A	0.7±0.3
B1	0.7±0.2
B2	0.7±0.2
C	0.9±0.2

limits @ 90% C.L.

We checked the hypothesis that contamination is mainly located on surfaces of the samples.

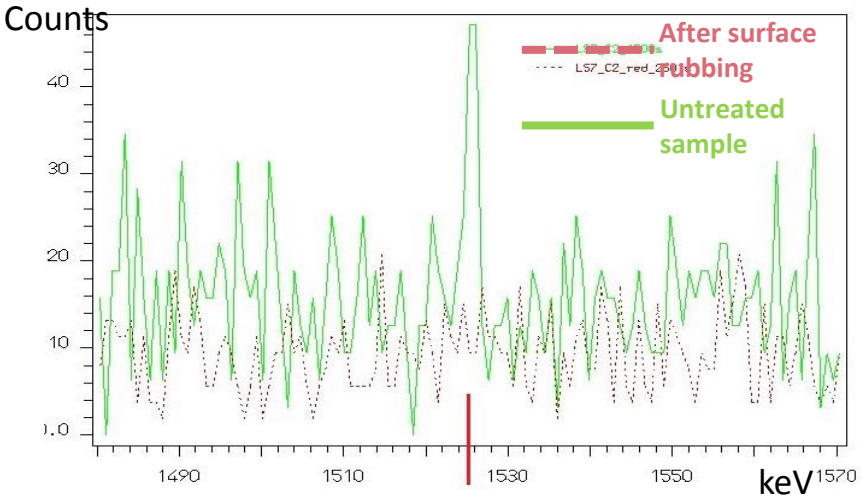
We decide to remove a thin surface layer

NAA results: ^{40}K

Problem of surface handling!

Sample	Mass(g)	Mass(g)
A	6.1786	5.754
B1	6.973	5.6672
B2	6.9494	6.1678
C	7.2181	6.441

Sample	Untreated samples ^{40}K(ppt)	After surface rubbing ^{40}K(ppt)
A	0.7±0.3	**<0.1**
B1	0.7±0.2	**<0.5**
B2	0.7±0.2	**<0.4**
C	0.9±0.2	**<0.1**



Sandpaper

The contamination was concentrated on the sample surface!

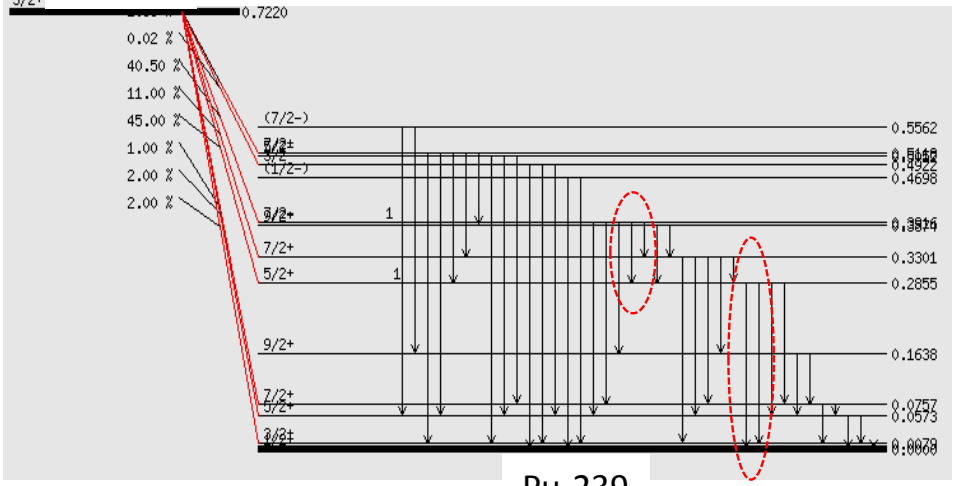
The reported limit on the presence of ^{40}K in acrylic are among the best ever achieved

NAA results: ^{238}U

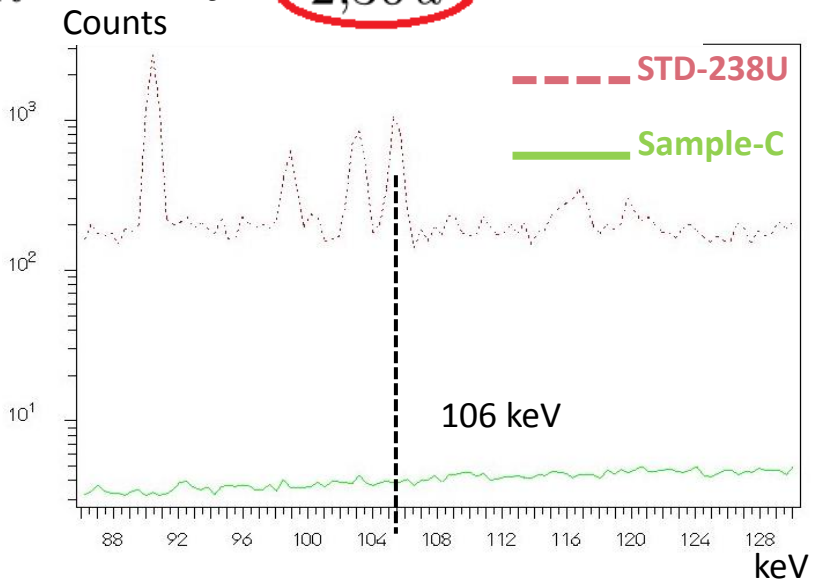
$$n + {}^{238}U \longrightarrow {}^{239}U \xrightarrow[23{,}5\,m]{\beta^-} {}^{239}Np \xrightarrow{\beta^-} {}^{239}Pu$$

$\tau = 2{,}36\,d$

Np-239



Pu-239



ϒ-ray(keV)	BR(%)
10612	26.3
228.18	11.14
277.60	14.44

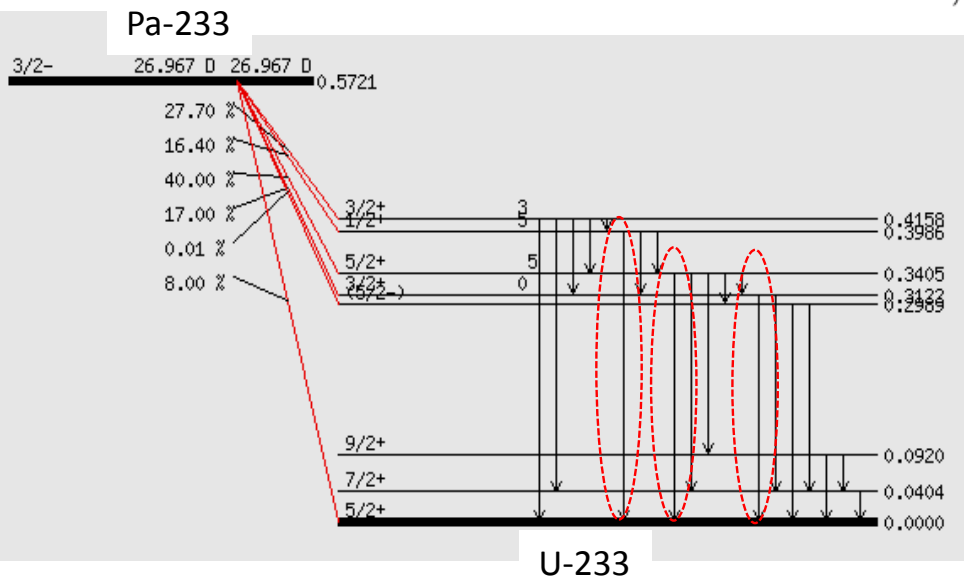
Measurements were carried out on 3 HPGe detectors

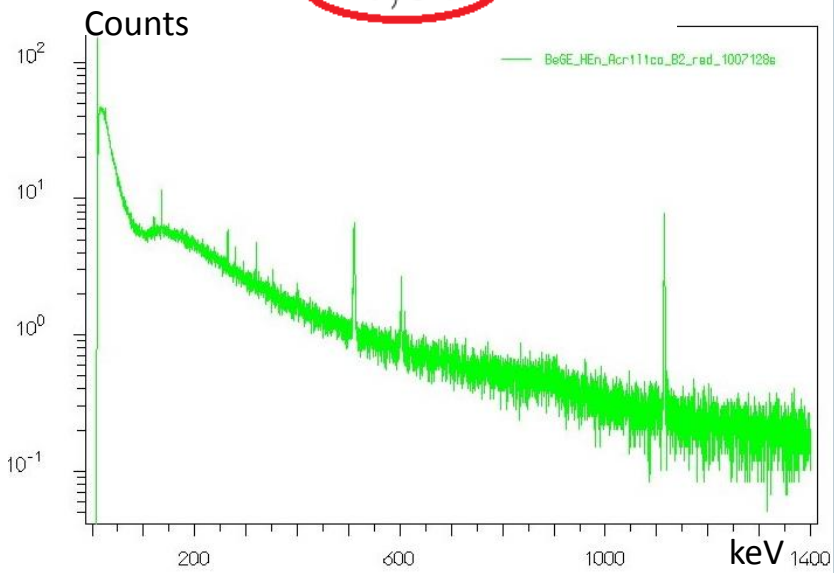
Sample	^{238}U(ppt)
A	<2.0
B1	<3.2
B2	<2.0
C	<0.7

limits @ 90% C.L.

NAA results: ^{232}Th

$$n + {}^{232}Th \longrightarrow {}^{233}Th \xrightarrow[22{,}3\,m]{\beta^-} {}^{233}Pa \xrightarrow[\tau = 27{,}0\,d]{\beta^-} {}^{233}U$$





ϒ-ray(keV)	BR(%)
300.13	6.63
311.90	38.5
340.48	4.45

Measurements were performed
~ 1 month after the irradiation

Sample	^{232}Th(ppt)
A	<2.9
B1	<2.4
B2	<2.5
C	<1.4

limits @ 90% C.L.

Conclusions

A methodology based on
neutron activation analysis (NNA) combined with treatment of the sample surfaces
has been developed to determine K, Th and U content in
Acrylic samples

By this methodology:

→ 10^{-12} g/g level has been achieved for ^{238}U and ^{232}Th

→ **10^{-13} g/g level has been achieved for ^{40}K**

Backup slides

Summary

		GePV Untreated	GePV treated	GeKan	GeGEM
Sample	STD	^{40}K(ppt)	^{40}K(ppt)	^{40}K(ppt)	^{40}K(ppt)
A	2	0.7±0.3	<0.1	<0.1	
B1	2	0.7±0.2	<0.5		<0.2
B1	4	0.7±0.2	<0.5		<0.2
B2	4	0.7±0.2	<0.4		<0.4
B2	6	0.7±0.2	<0.4		<0.4
C	6	0.9±0.2	<0.1	<0.1	

		GeKan	GeGEM	BeGE
Sample	STD	^{238}U(ppt)	^{238}U(ppt)	^{238}U(ppt)
A	2	<2	<2	
B1	2	<7	<3	
B1	4	<8	<3	
B2	4		<2	
B2	6		<2	
C	6	<1		<0.7

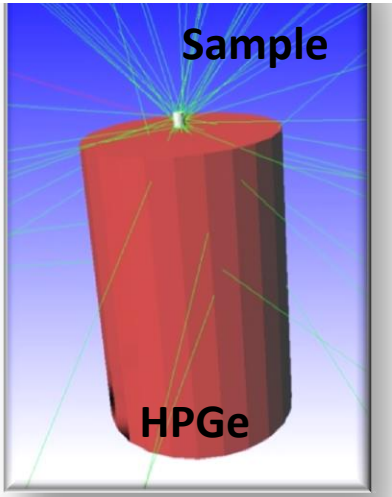
		GeKan	GeGEM	BeGE
Sample	STD	^{232}Th(ppt)	^{232}Th(ppt)	^{232}Th(ppt)
A	2	<3		<3
B1	2			<2
B1	4			<3
B2	4			<3
B2	6			<3
C	6		<3	<1

Measurements made with different standards are independent and are always compatible

STD solid

Channel	LS1	LS2	LS3	LS4	LS5	LS6	LS7
Sample/ STD liquid	A	STD2	B1	STD4	B2	STD6	C
STD solid	SS1	SS2	SS3	SS4	SS5	SS6	SS7

MC simulation



Al-Co wires

Integral flux

$$\Phi = \frac{R}{N\sigma_{eff}}$$

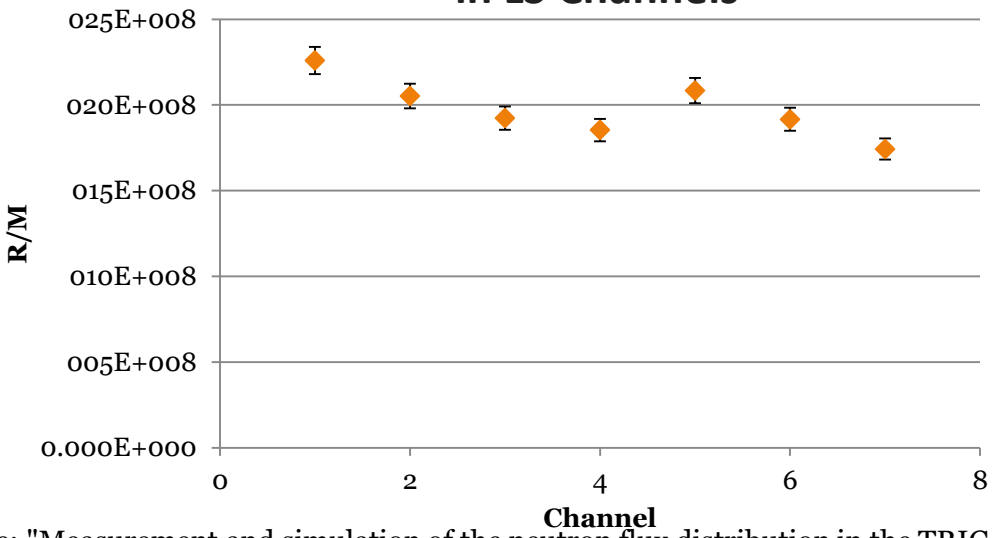
σ_{eff}: calculated from MCNP

Activation Rate

$$R \sim n_{dec} = \frac{C_{meas}}{C_{sim}} n_{sim}$$

Channel	Mass(mg)
1	34.25
2	29.92
3	31.44
4	37.56
5	32.30
6	34.25
7	31.43

Verification of the uniformity of the flux in LS Channels



Reference: "Measurement and simulation of the neutron flux distribution in the TRIGA Mark II reactor core" Correspondig author: D.Chiesa

Efficiency: corrective factor

Acrylic Sample and STD liquid have a different geometry

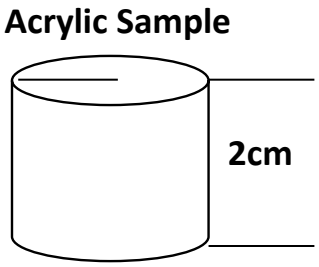
$$Efficiency_{Sample} \neq Efficiency_{STD}$$

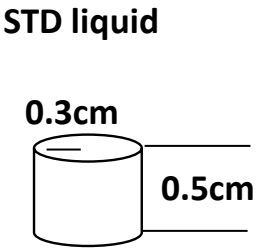
↓

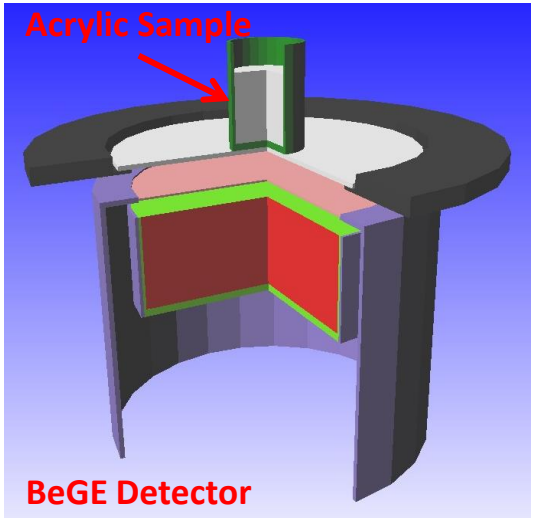
Corrective factor via MonteCarlo simulation

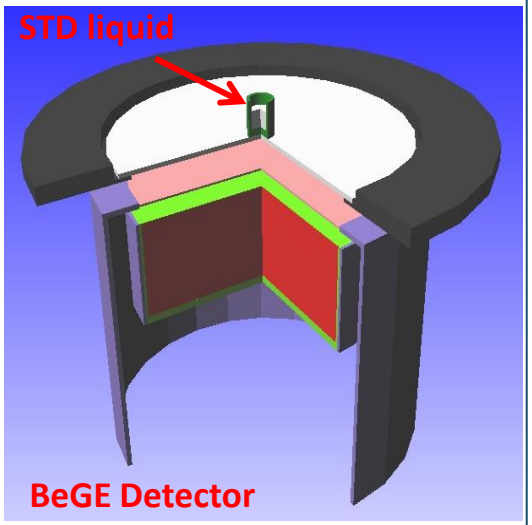
↓

$$\boldsymbol{E_{Sample}}/_{\boldsymbol{STD}} = \frac{Efficiency_{Sample}}{Efficiency_{STD}}$$









	GeGEM	GeKan	BeGE
$\boldsymbol{E_{Sample}}/_{\boldsymbol{STD}}$	1.55	1.3	1.31

$$C_{Sample}\left(\frac{g}{g}\right) = I.A.\frac{Counts_{Sample}}{Counts_{STD}}\frac{C_{STD}M_{STD}}{M_{Sample}}\frac{e^{-\lambda T_{Wait-STD}}}{e^{-\lambda T_{Wait-Sample}}}\frac{\left(1-e^{-\lambda T_{Mis-STD}}\right)}{\left(1-e^{-\lambda T_{Mis-Sample}}\right)}\boldsymbol{E_{Sample}}/_{\boldsymbol{STD}}$$

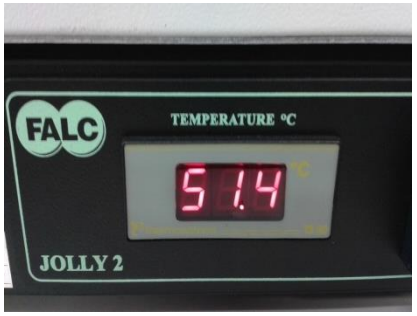
NAA: preparation of irradiation (December 2015)

Samples were prepared together with the NAA standards for neutron irradiation in the **Central Thimble** (Flux of neutrons: $10^{13} cm^{-2} s^{-1}$)

Preparation of the standards inside quarz vials

Acrylic sample + standards in the irradiation containers (one per sample)

The Acrylic cylinders were destroyed!



Not a question of temperature inside the reactor core (~70-80 °C)