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# Status of LAB radiopurity measurements

**Italian JUNO Meeting - PoliMi**
**5-6th May, 2022**

**Speaker: Nastasi Massimiliano**

# Radiopurity LAB measurements

**Juno baseline request for LAB:**
$^{238}U$, $^{232}Th$, $^{40}K$ $< 1 \cdot 10^{-15}$ g/g

In recent months we have **validated a measurement procedures** suitable to achieve the required sensitivity

**Two procedures** have been tested in order to identify natural contamination in LAB samples

**Uranium and Thorium**

**Potassium**

**High sensitivity measurements**

NAA + Low background detector

- Concentrate the sample
- Remove interferences

Avoiding to introduce external contamination

# Main steps for $^{238}U$ and $^{232}Th$ measurements

**Cleaning protocol (Pre-Irradiation)** → Any manipulation or treatment of the sample could introduce contaminations before irradiation

**Chemical/Radiochemical Treatments (Pre-Irradiation)** → Allows to **remove interferences** and **concentrate** the sample

**Sample irradiation** → Allows to transform long life nuclide $^{238}U/^{232}Th$ into the radioactive short life $^{239}Np/^{233}Pa$ nuclide. Sensitivity <1ppt

$$^{232}_{90}Th + \mathrm{n} \longrightarrow {}^{233}_{91}Pa \qquad {}^{238}_{92}U + \mathrm{n} \longrightarrow {}^{239}_{93}Np$$

**γ measurements** → We developed a new detector suitable to **β – γ coincidence** measurements on liquid irradiated samples

# $^{238}U$ - $^{232}Th$ procedure validation

Sample and container cleaning **preparation**

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**Chemical/Radiochemical Treatments**

↓

**Irradiation**

↓

**Extraction Chromatography**
*TEVA Resin*

↓

**β-γ coincidence measurements**

↓

**Calculation** of the **quantity of precursor** element

**Extraction**

↓

**Extraction Chromatography**
*U-TEVA Resin*

Procedure has been **validated** perfoming a measurement on a **blank** sample

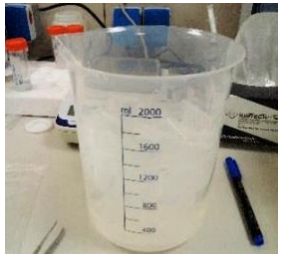
**Cleaning protocol (Pre-irradiation)**

- Tools washing
- Reagents/Containers validation

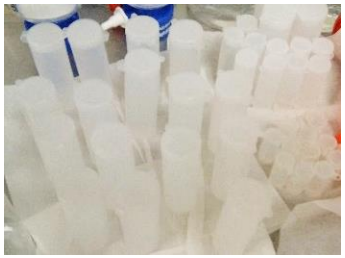
All operations are carried out in a **clean room**

All **tools are cleaned prior to sample handling** with a specific protocol

4 days of immersion in slightly nitric acid water for all the tools



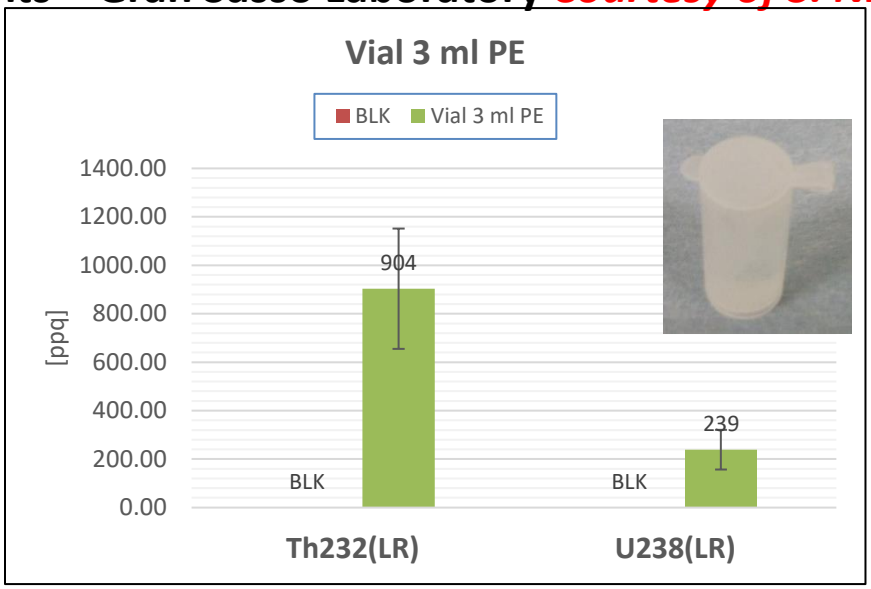
Vials are filled with nitric acid and water for several days

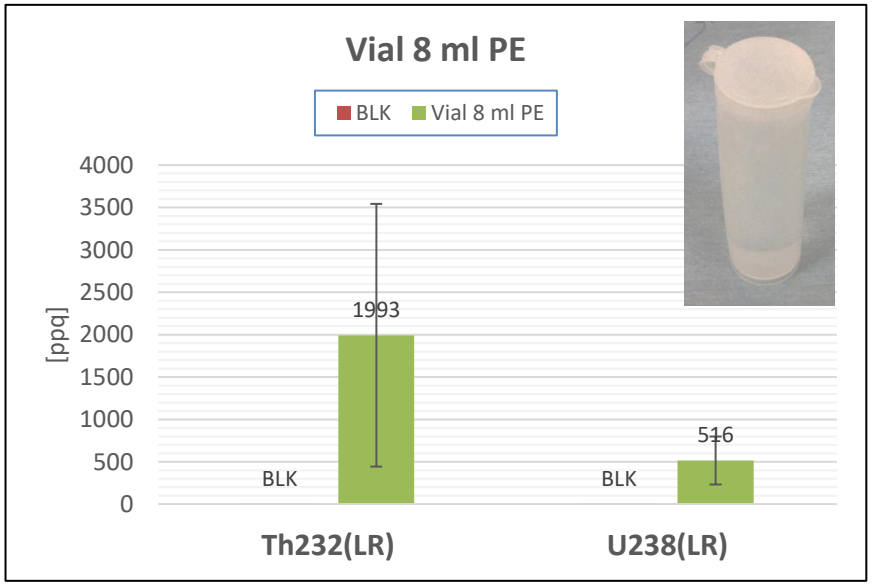


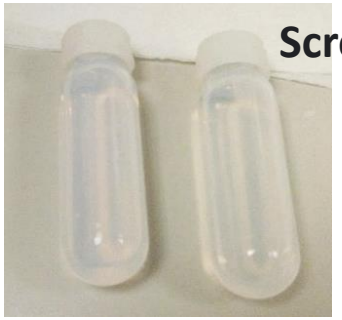
# Containers validation

**ICP-MS measurements – Gran Sasso Laboratory *Courtesy of S. Nisi***

- The containers in Polyethylene (PE) could release uranium and thorium from the walls
- The containers in with pressure cap are unsuitable for ultra-low background: the insertion of the cap has high probability to introducing contamination in the sample



**Screw cap**

It's **mandatory** to use tools and container in **PFA/PTFA material with screw cap** suitable for trace elements analysis

# Reagents validation

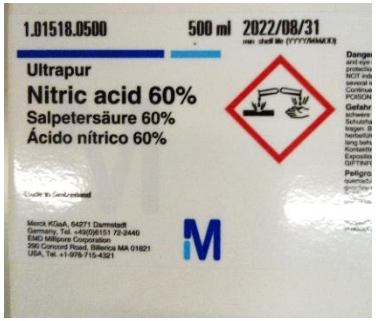
## Water

### ICP-MS measurements – Gran Sasso Laboratory

| | $^{238}U$ | $^{232}Th$ |
|---|---|---|
| $H_2O$ MilliQ | $< 0.7 \cdot 10^{-15}$ g/g | $< 0.8 \cdot 10^{-15}$ g/g |
| $H_2O$ MilliQ Element | $< 0.7 \cdot 10^{-15}$ g/g | $< 0.8 \cdot 10^{-15}$ g/g |

***Courtesy of S. Nisi***

## Nitric Acid




### ICP-MS measurements – Gran Sasso Laboratory

| | $^{238}U$ | $^{232}Th$ |
|---|---|---|
| $HNO_3$ Iper-pur | $< 3 \cdot 10^{-14}$ g/g | $< 3 \cdot 10^{-14}$ g/g |

**Chemical/Radiochemical Treatments (Pre-irradiation)**

Allow to **remove interferences** and **concentrate** the sample

**Liquid-Liquid Extraction**

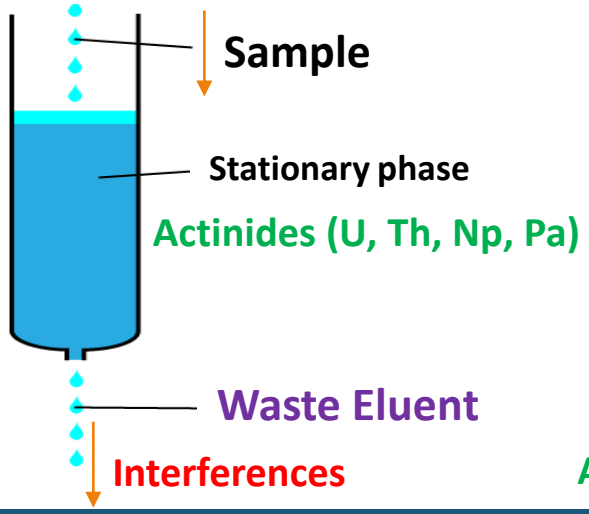
Consist in the transfer of the contaminations of U and Th from **LAB sample** into a liquid solution(**HNO3 + water**)

Proportion: HNO3+Water:LAB =1:5

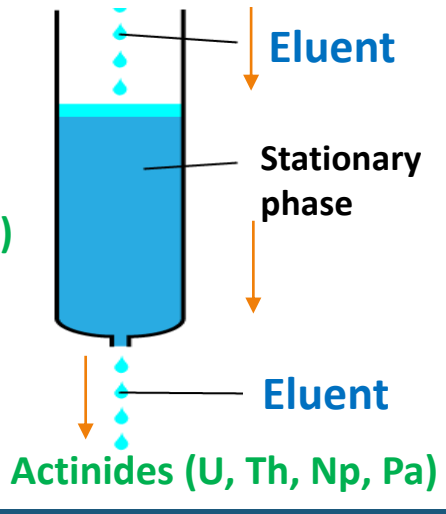
**Extraction Chromatography**

Ideally the column chromatography **selectively absorbs actinide activities** (U, Th, Pa ,Np) while allowing interferences pass through

**Charging**




**Washing** (Acid solution)




**Pre-Irradiation:** *U-TEVA Resin*
Uranium and Thorium

# Efficiency of pre irradiation steps

The effectiveness of the chemistry and radiochemistry treatments has been studied considering **spiked LAB samples**

*L-L* Extraction

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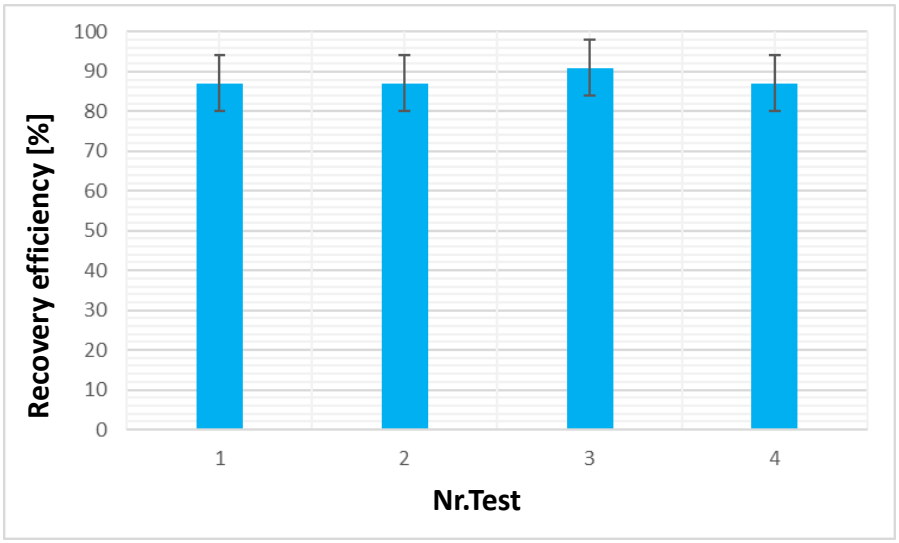
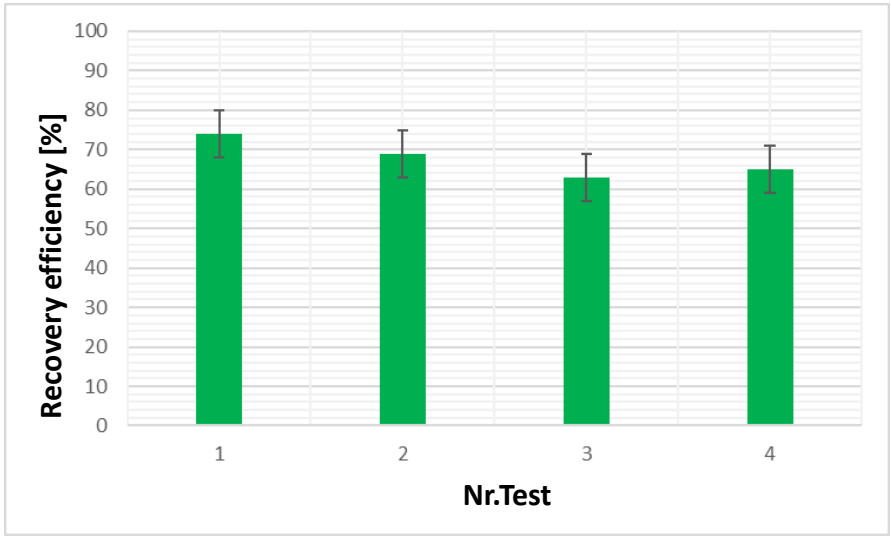
Extraction Chromatography ***U-TEVA Resin***

**STDs** containing a known amount of the element are used as **tracers**


$^{238}$U Recovery Efficiency: **(88 ± 5)%**




$^{232}$Th Recovery Efficiency: **(68 ± 7)%**



# Neutron Activation Analysis (NAA)

**Sample irradiation**

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

The NAA technique consists of several steps:

Sample and STD reference are **exposed** to a neutron flux

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

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

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

**Extraction** of the irradiated sample and **measurement** of induced ϒ radioactivity → **Calculation** of the **quantity of precursor** element $({}^{A}_{Z}X)$

**TRIGA Mark II**

**Research reactor (250 kW) - Pavia, Italy**

**LAZY SUSAN facility**:

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

**Irradiation Time**: 6 hours

# $^{238}U$ - $^{232}Th$ steps post irradiation

**Irradiation**

↓

**Extraction Chromatrography**
*TEVA Resin*

Removes any **remaining interferences**

$\epsilon_{Removal} \sim 99\%$

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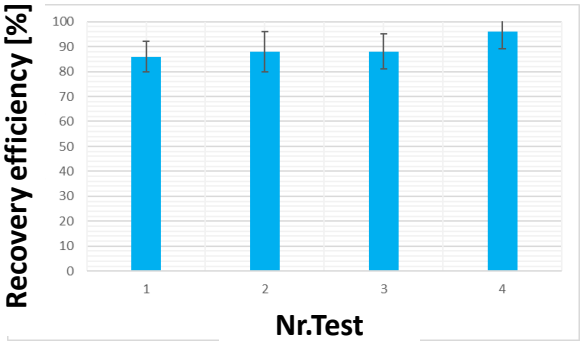
$^{238}U$ Recovery Efficiency: **(89 ± 6)%**




**β-γ coincidence measurements**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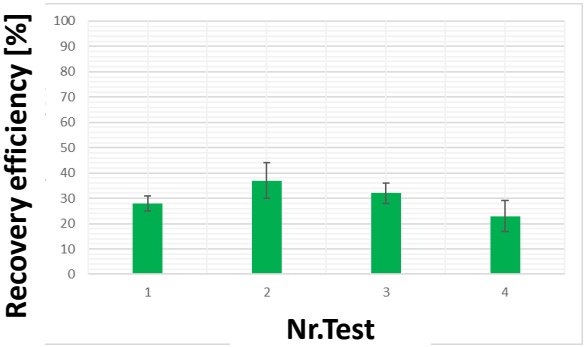
$^{232}Th$ Recovery Efficiency: **(30 ± 8)%**




**Calculation** of the **quantity of precursor** element

# β-γ coincidence detector

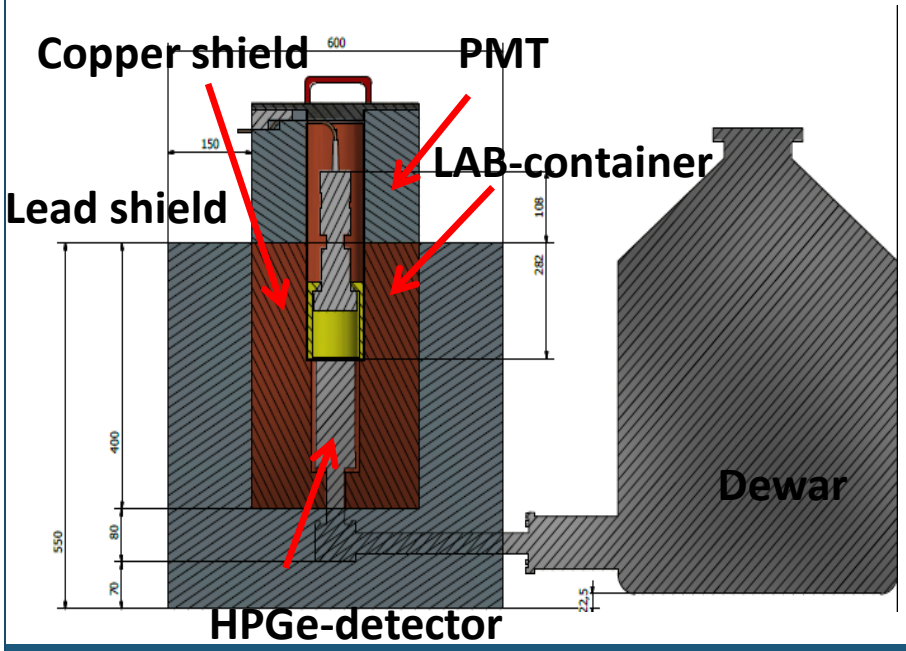
**β-γ measurements**

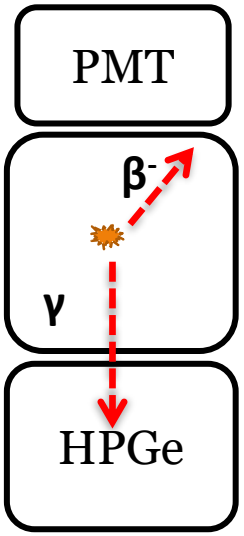
**Neutron Activation Reaction**

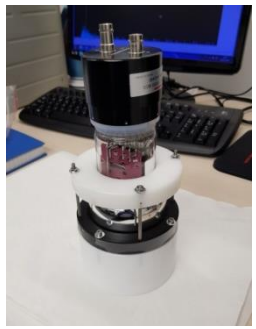
$$ {}^{A}_{Z}X + n \longrightarrow {}^{A+1}_{Z+1}Y + \gamma_{cascade} $$

($\beta^-$)

Irradiated sample is mixed with not irradiated liquid scintillator



β-γ coincidence detector allows a strong **background reduction**

# Sensitivity for $^{238}U$ - $^{232}Th$

**The blanks** are critical for determining the contamination introduced during pre-treatment and are essential for evaluate the sensitivity

The blank went through all processing steps **just without LAB**

Nitric acid water
Blank mass: 228g

*It is representative of the measure of*
***1 kg of Liquid scintillator (LAB)***

Proportion: LAB:HNO3+Water = 1: 5

| Blank | $^{238}U$[g/g] | $^{232}Th$[g/g] |
|---|---|---|
| Mass sample 228g | **$(9,5\pm2,4)\cdot10^{-15}$** | **$<7,7\cdot10^{-14}$** |

limits @ 90% C.L.

In the hypotesis that we got a LAB sample of 1 kg without contaminations we could achieve a sensitivity of: **$2\cdot10^{-15}$g/g** for **$^{238}U$ - $1,5\cdot10^{-14}$g/g** for **$^{232}Th$**

# Radiopurity LAB – $^{40}$K

Sample and container cleaning **preparation**

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**LAB + STD reference has irradiated**

↓

**γ measurements (HPGe)**

↓

**Calculation** of the **quantity of precursor** element

To study potassium contamination we irradiated a LAB sample

Sample mass: **19g**

$$n + {}^{41}K \longrightarrow {}^{42}K \xrightarrow[12{,}36\,h]{\beta^-} {}^{42}Ca$$

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

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

| $^{40}$K [g/g] |
|---|
| **< 8,3·10$^{-15}$** |

limits @ 90% C.L.

# Summary

$^{238}U$ and $^{232}Th$ measurements

- The cleaning protocol has been defined
- Chemistry procedure has been tested

→ Processes are reliable and consistent

Recovery efficiency has been determined

For **Uranium** and **Thorium** a representative **blank sample** of the whole procedure has been mesured

| Blank | $^{238}U$[g/g] | $^{232}Th$[g/g] |
|---|---|---|
| Mass sample 228g | **$(9,5\pm2,4)\cdot10^{-15}$** | **$<7,7\cdot10^{-14}$** |

Rescaling these results for a mass of 1 kg we could achieve a **sensitivity**: **$2\cdot10^{-15}$g/g** for **$^{238}U$** and **$1,5\cdot10^{-14}$g/g** for **$^{232}Th$**

Radiopurity LAB – Potassium

| LAB 19g | $^{40}K$ | **$< 8,3\cdot10^{-15}$g/g** |
|---|---|---|

# Future plan:

- We plan to perform measurements on new blank samples

- We will try out to increase sensitivity

**Uranium and Thorium** → β – γ measurements with a higher efficiency

**Potassium** → Radiochemical treatments post irradiation