

Decay energy spectroscopy with solid phase extraction for radio-assay of high-purity copper

Jongseok Chung

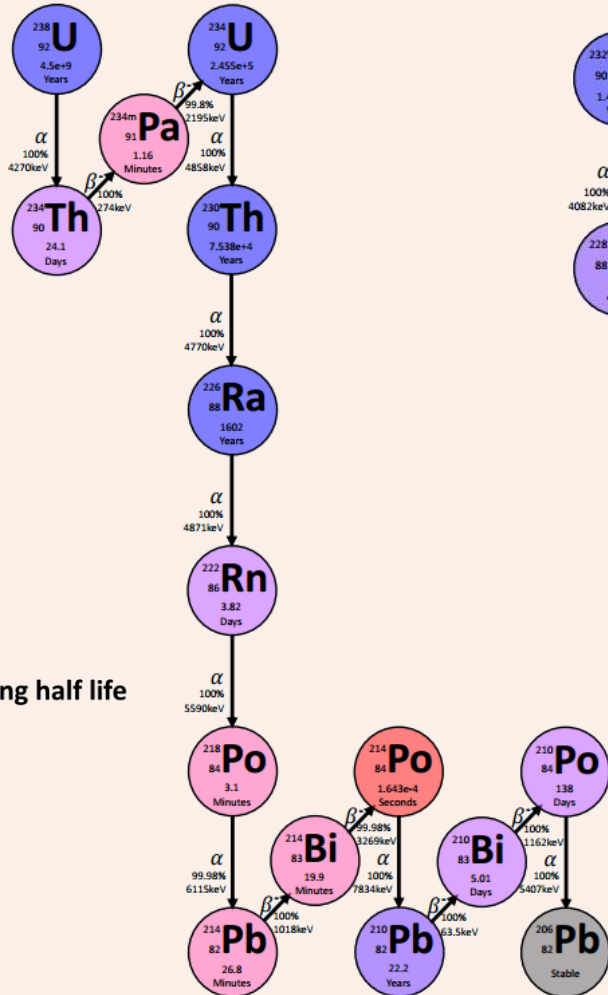
Institute for Basic Science
Chung-Ang Univ. Seoul. Korea



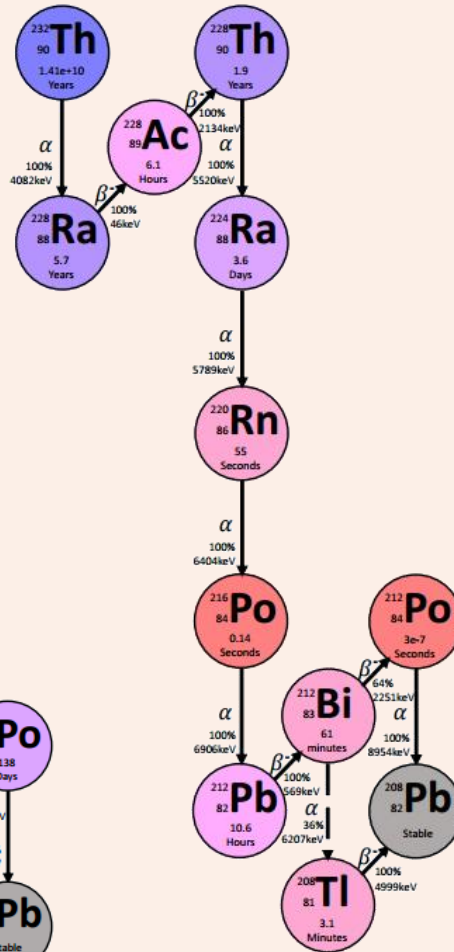
24. SEP. 2026

Motivations for the radio-assay

U-series



Th-series



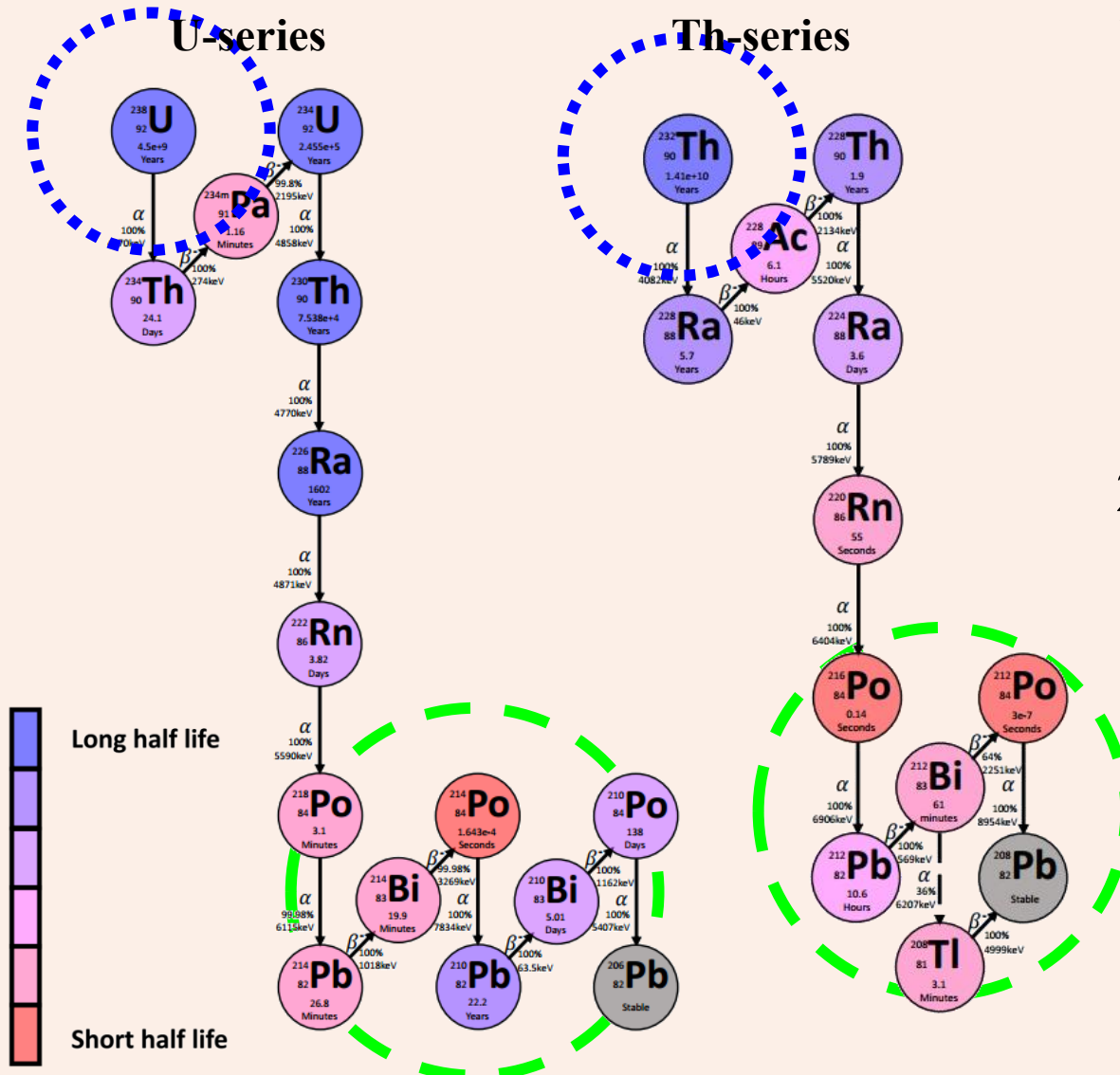
In rare event search experiments, such as **Dark Matter**, $0\nu\beta\beta$ search, **Radio active isotopes** from these U & Th series can create significant background events.



Long half life

Short half life

Conventional methods for the radio-assay



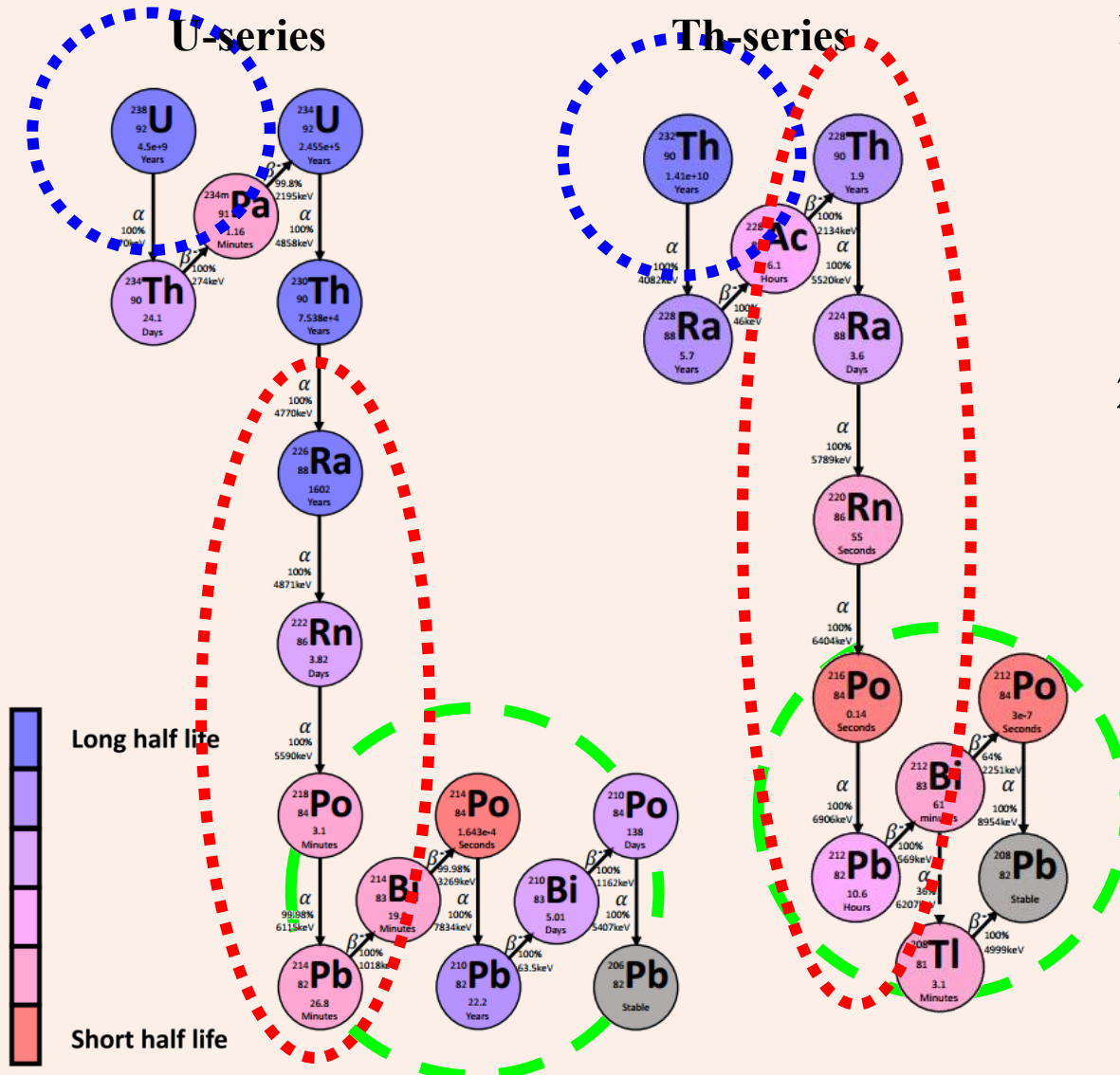
1. ICP-MS : Measure the **number of nuclei**
LIMITATION:

Their activity can be different from their descendant's activities

2. HPGe : Measure the **gamma peaks**
LIMITATION:

Detection efficiency is limited by sample self-shielding

Conventional methods for the radio-assay



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LIMITATION:

Their activity can be different from their descendant's activities

2. HPGe : Measure the **gamma peaks**

LIMITATION:

Detection efficiency is limited by sample self-shielding

We aim to achieve **sensitivity to the entire alpha-decay chain** using decay energy spectroscopy

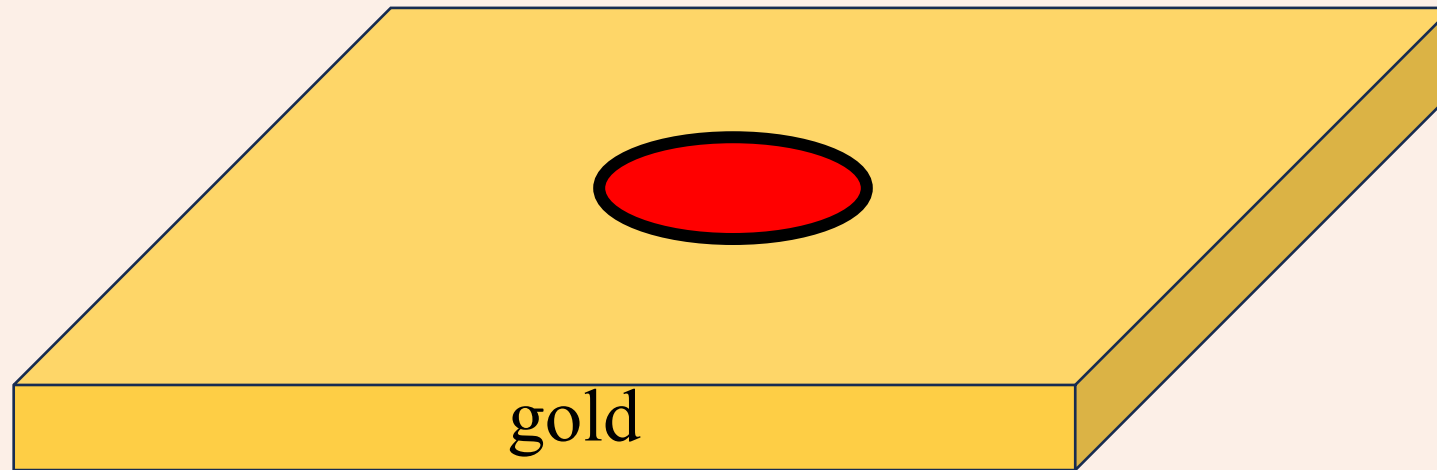
Decay energy spectroscopy(DES) introduction

Prepare the **gold foil**



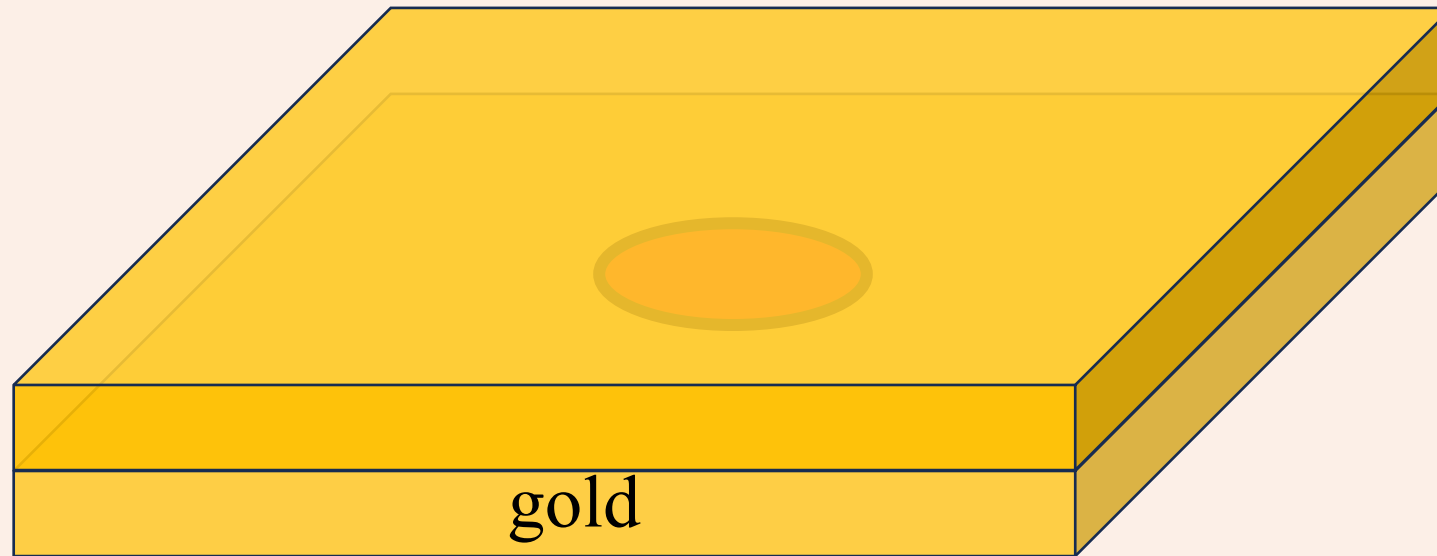
Decay energy spectroscopy(DES) introduction

Place the **radioactive isotopes** on the gold foil



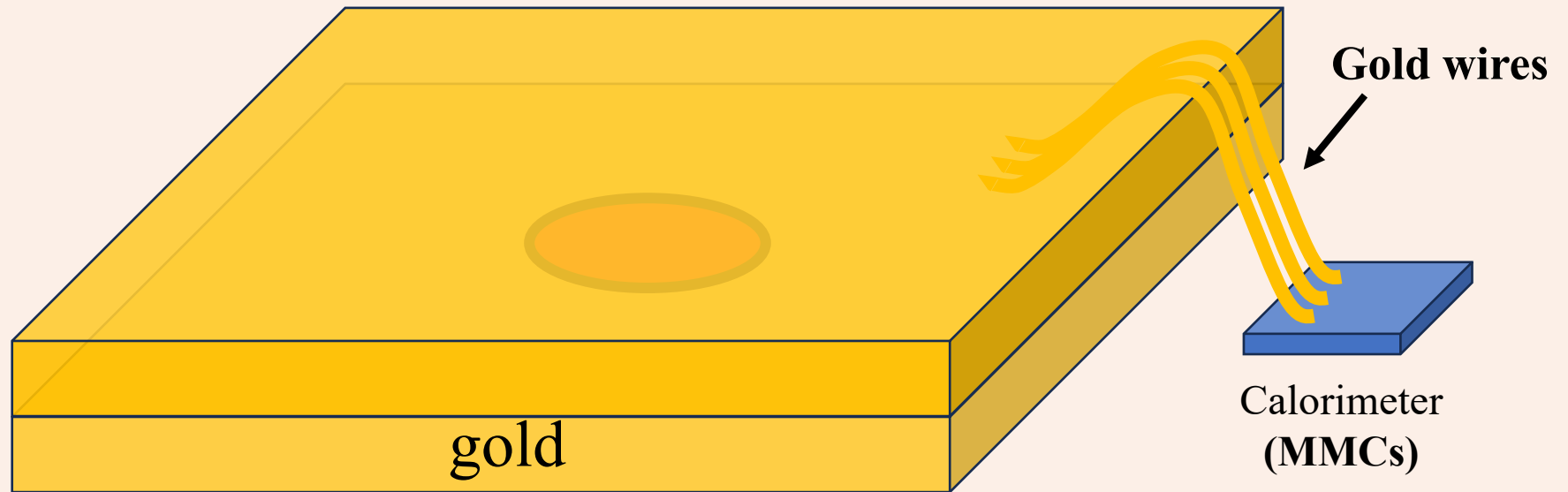
Decay energy spectroscopy(DES) introduction

Enclose the radioactive isotopes in the gold foil



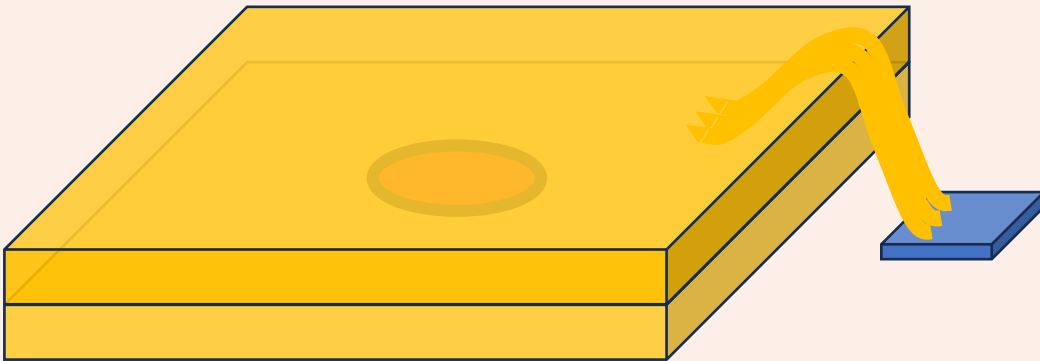
Decay energy spectroscopy(DES) introduction

Connect it to the **MMCs** using gold wires



Decay energy spectroscopy(DES) introduction

DES

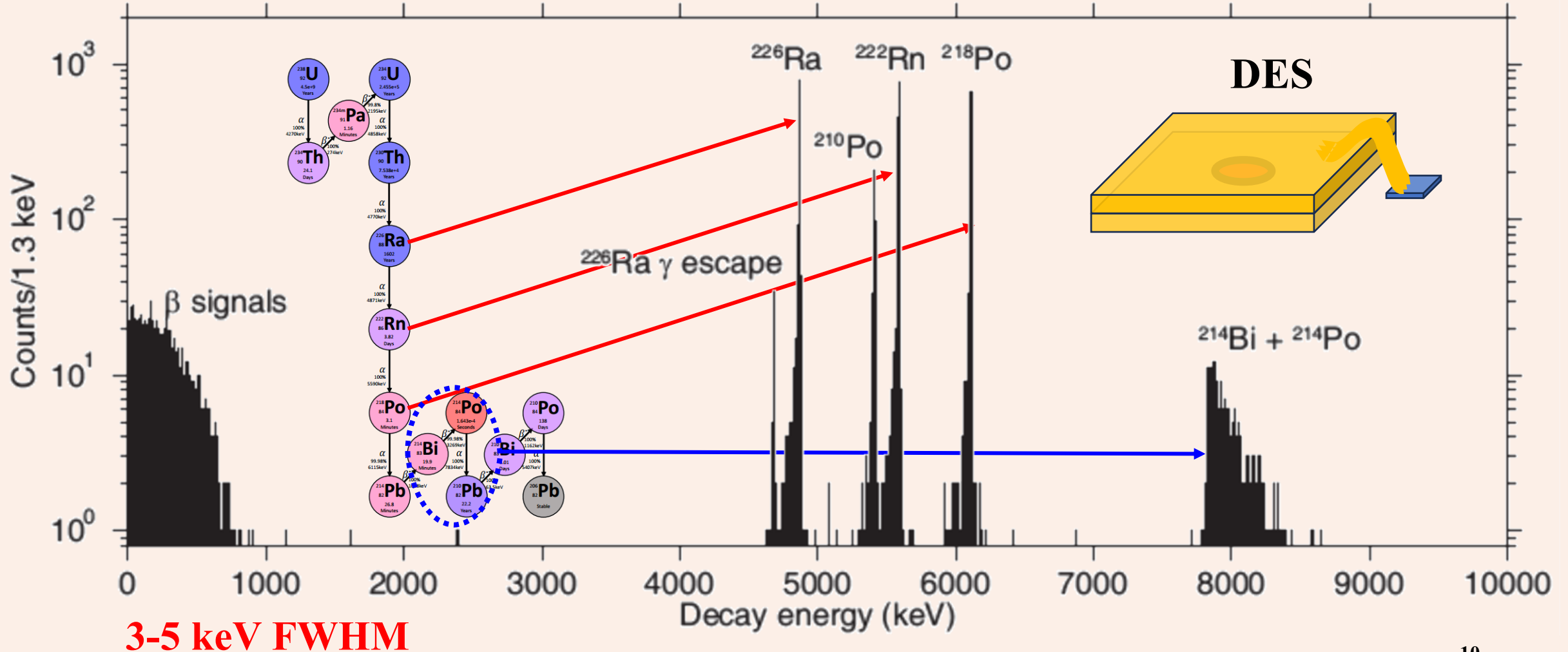


Measure the temperature change
caused by radio-decay energy
(Base temperature : $\sim 10\text{mK}$)

- 4π detection for α decay
(100% detection efficiency)
- Whole decay energy can be
measured (recoil + α)
- Children nuclei will be
staying inside of gold foil

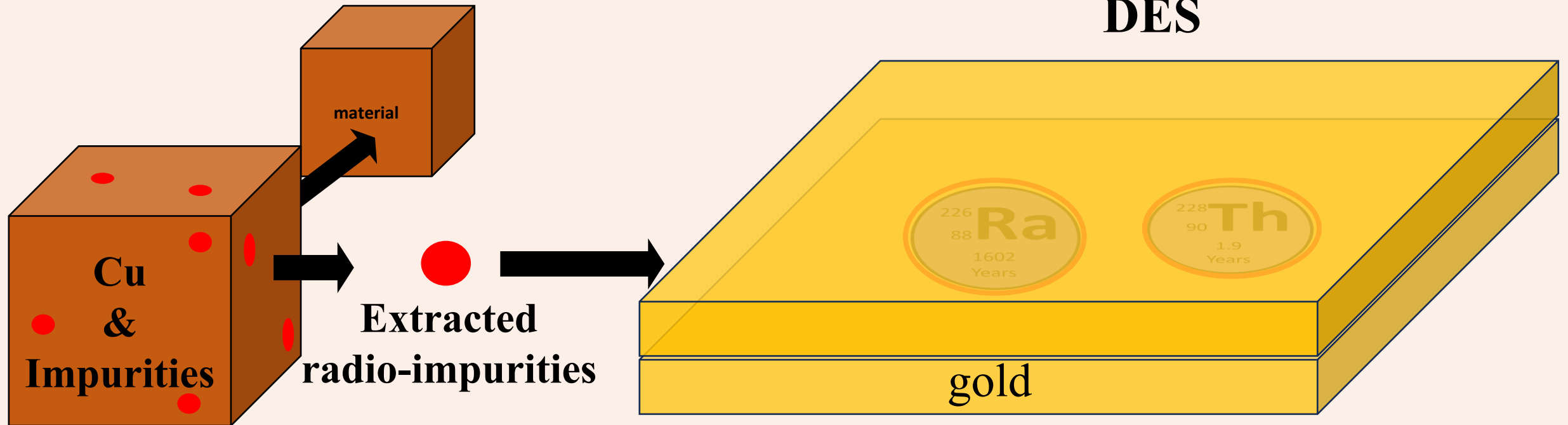
Decay energy spectroscopy(DES) introduction

DES example : ^{226}Ra reference source measurements



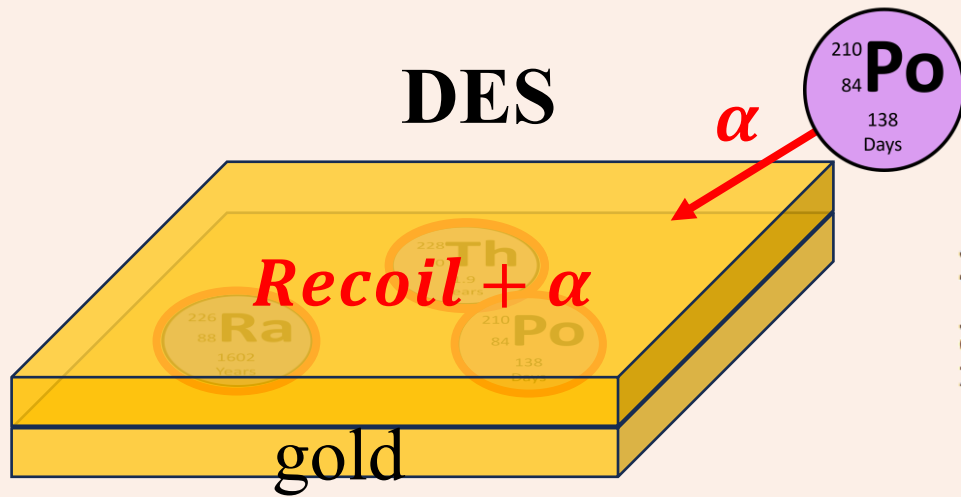
The way for radio-assay with DES

Chemical extraction

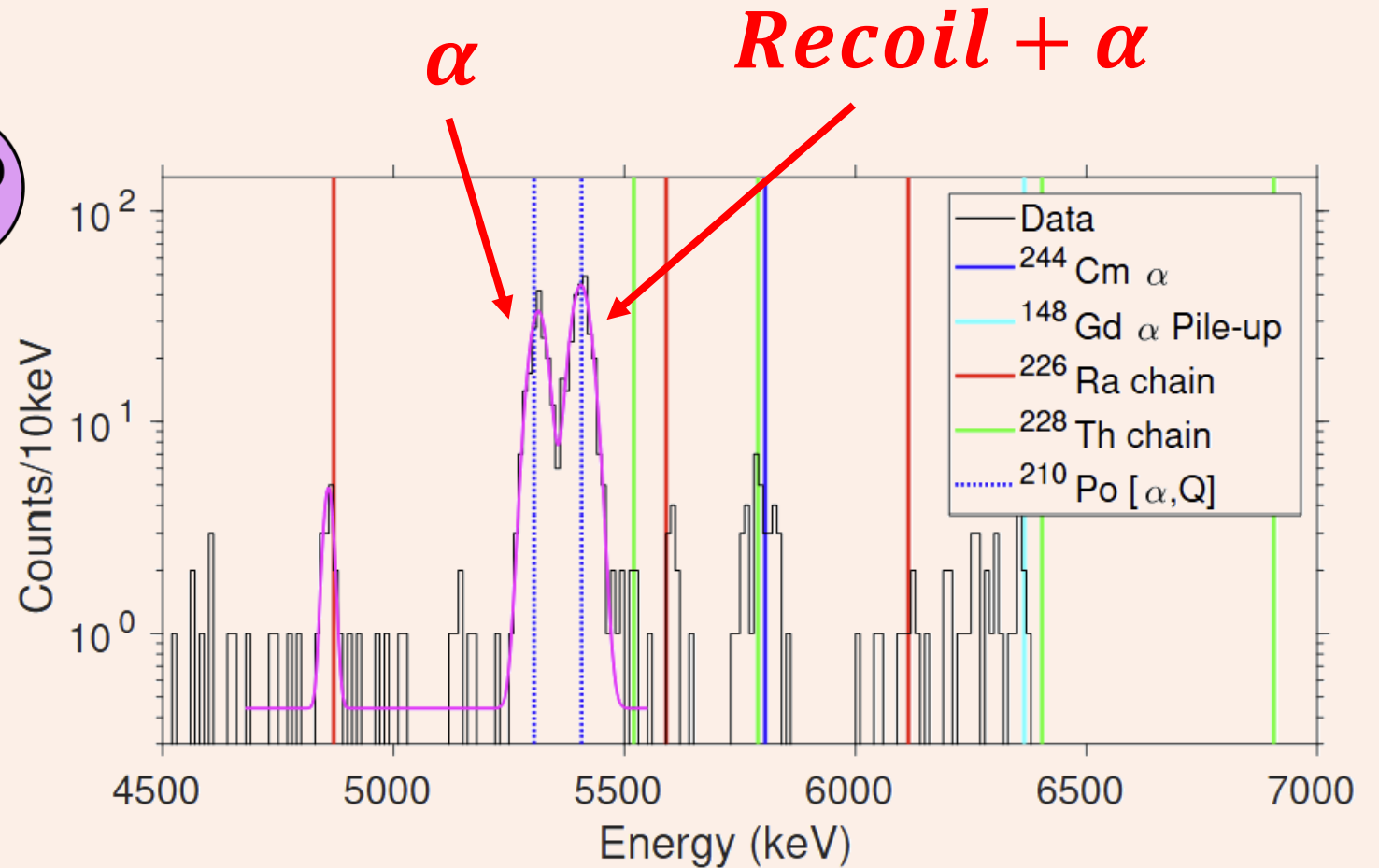


Chemical extraction isolates the radio-impurities from the bulk material for deposition onto the gold absorber

The power of radio-assay with DES

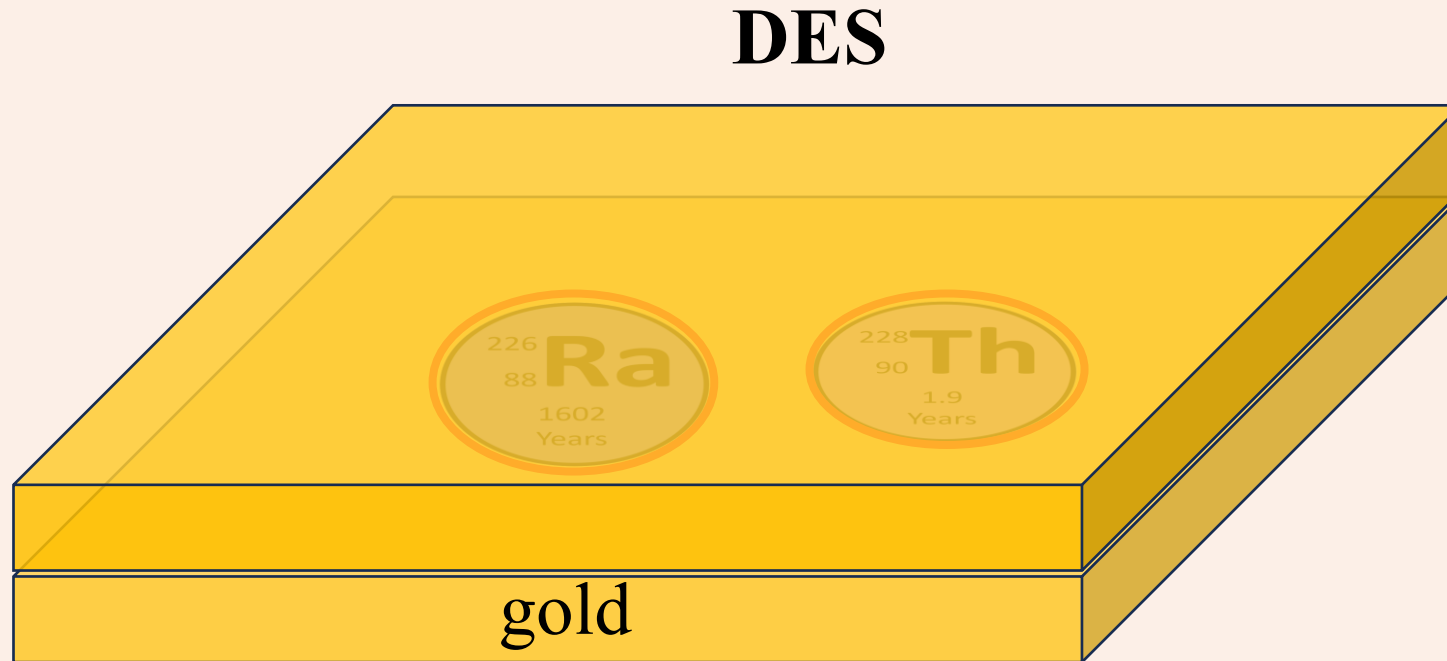


DES enables us to **distinguish the external α backgrounds**

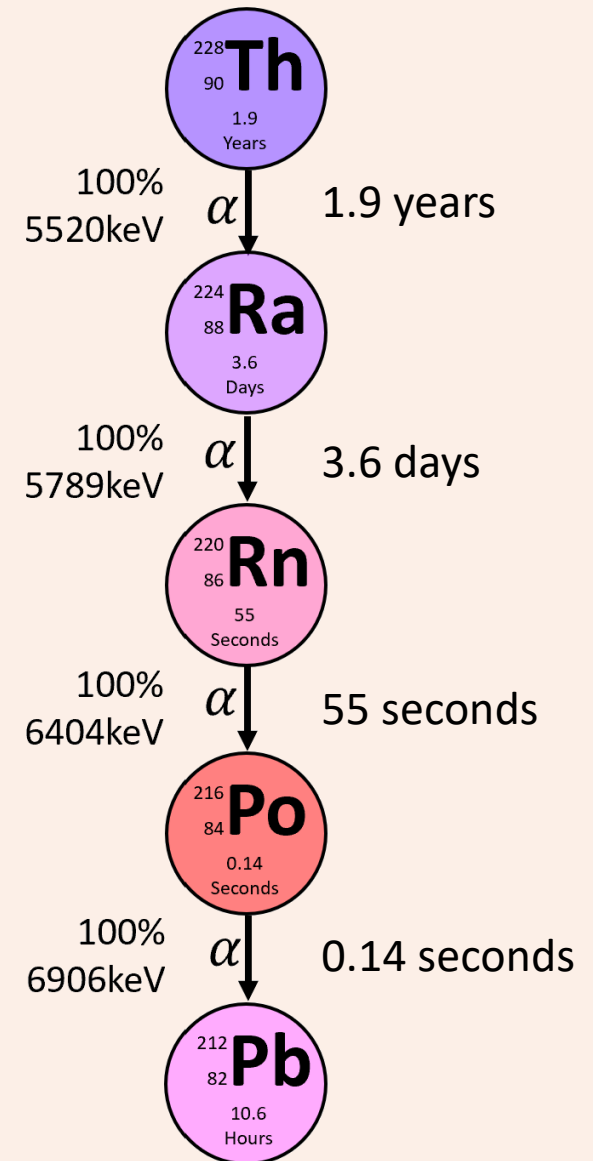


Previous work : [10.1109/TASC.2024.3515965](https://doi.org/10.1109/TASC.2024.3515965)

The power of radio-assay with DES



DES enables
Delayed-time coincidence analysis



Aurubis NOSV copper sample (Th) measurement with UTEVA

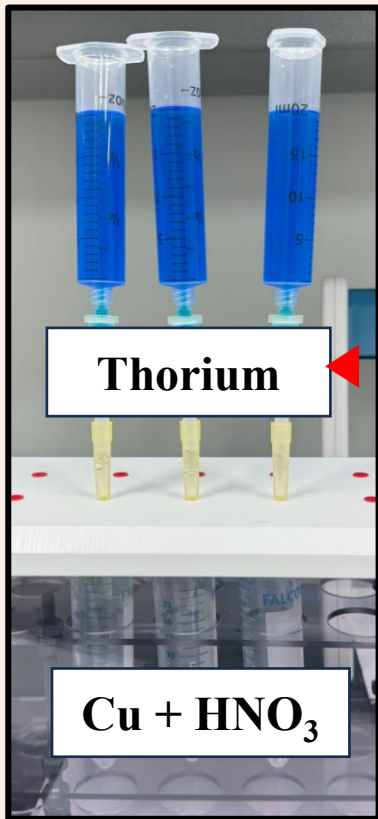
$\text{Cu(Th)} + (27\%)\text{HNO}_3$



1. Thorium is retained by UTEVA

Aurubis NOSV copper sample (Th) measurement with UTEVA

$\text{Cu(Th)} + (27\%) \text{HNO}_3$



$(0.1\%) \text{HNO}_3$



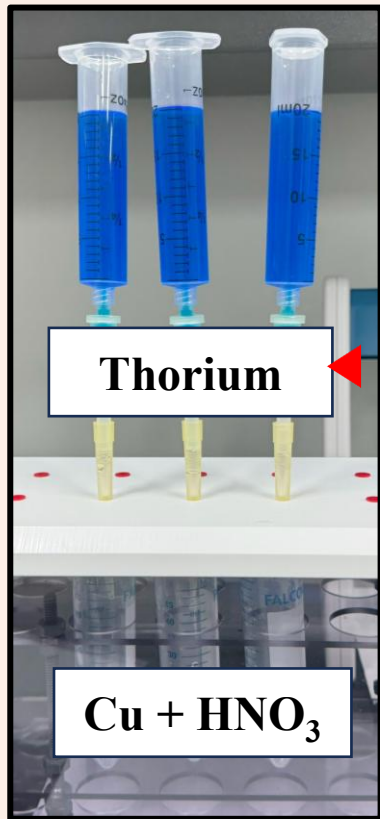
UTEVA
Uranium and
TEtraValents
Actinides

<https://doi.org/10.1016/j.apradiso.2023.110673>
95% extraction efficiency

1. Thorium is retained by UTEVA
2. Thorium is eluted from UTEVA

Aurubis NOSV copper sample (Th) measurement with UTEVA

$\text{Cu(Th)} + (27\%) \text{HNO}_3$



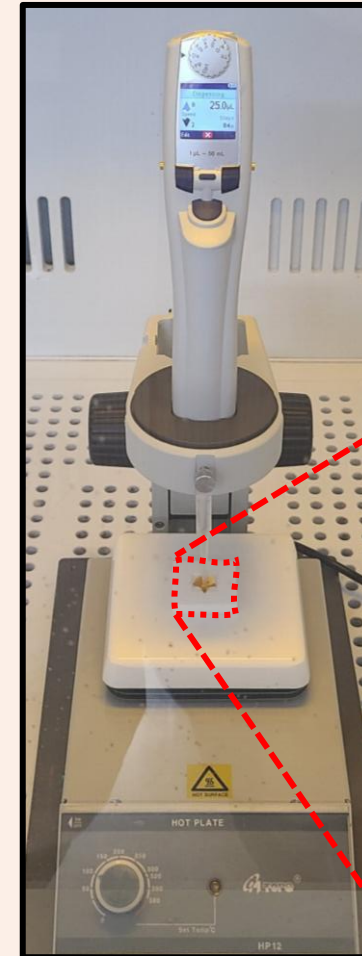
UTEVA
Uranium and
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$(0.1\%) \text{HNO}_3$



95% extraction efficiency
<https://doi.org/10.1016/j.apradiso.2023.110673>

Preconcentration
50mL to 2.5mL

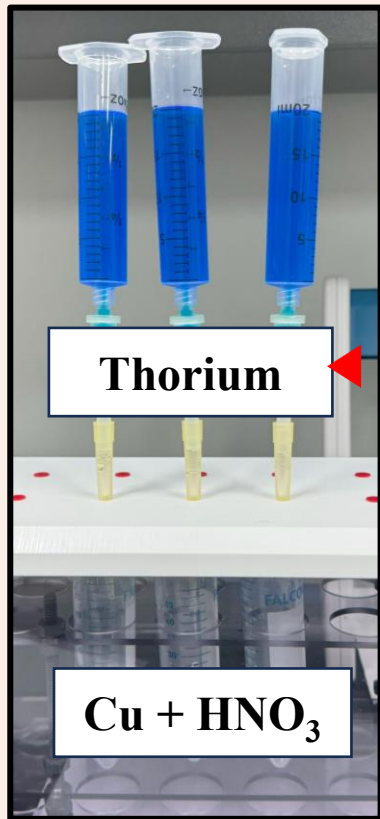


1. Thorium is retained by UTEVA 2. Thorium is eluted from UTEVA

3. Drop the eluted Thorium solution(2mL) & Dry 16

Aurubis NOSV copper sample (Th) measurement with UTEVA

$\text{Cu(Th)} + (27\%) \text{HNO}_3$



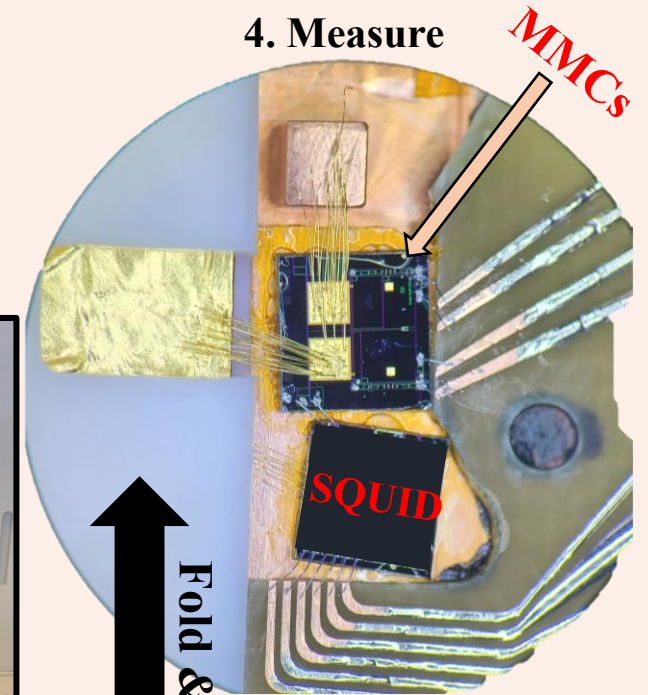
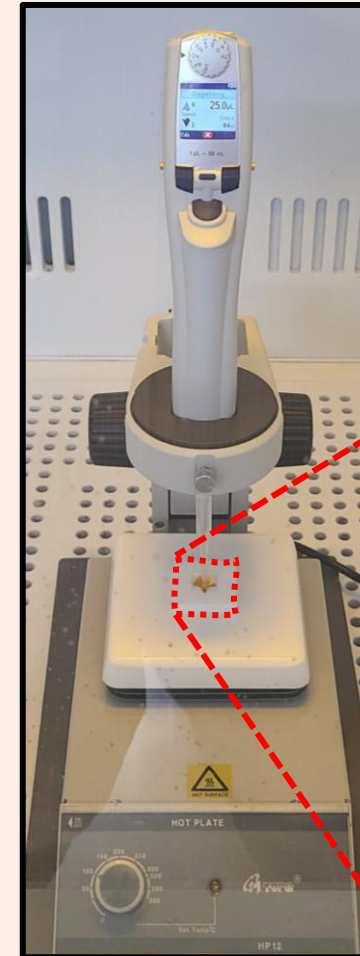
UTEVA
Uranium and
TEtraValents
Actinides

$(0.1\%) \text{HNO}_3$



95% extraction efficiency
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Preconcentration
50mL to 2.5mL



Fold & Weld



1. Thorium is retained by UTEVA 2. Thorium is eluted from UTEVA

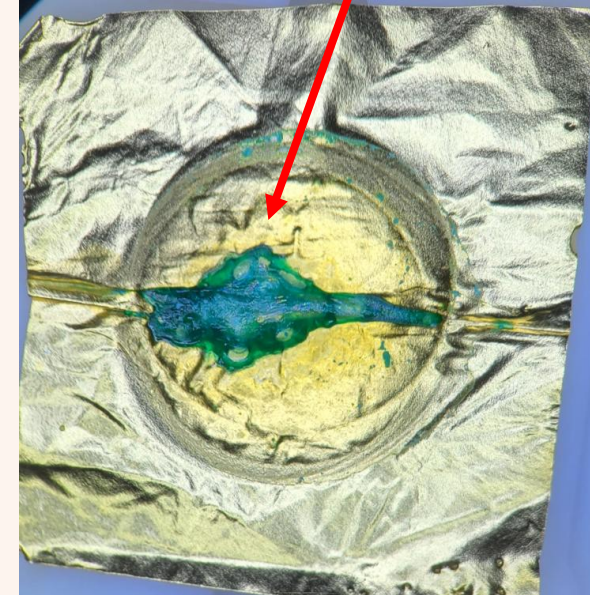
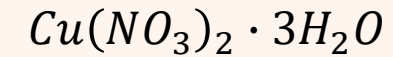
3. Drop the eluted Thorium solution(2mL) & Dry 17

Some discoveries during the dropping



Gold was damaged by dropping solution
(HCl used for extraction)

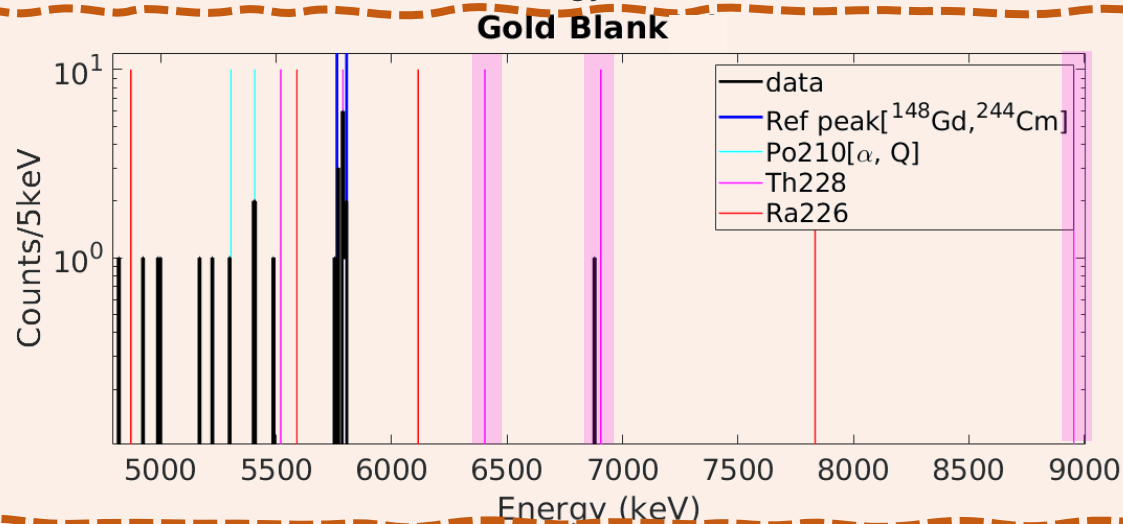
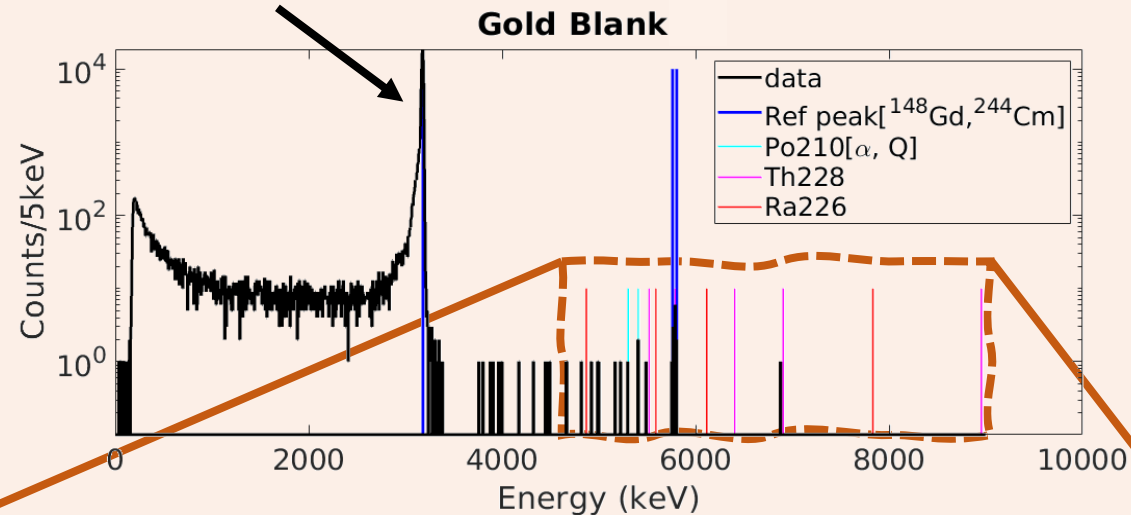
Solved!



0.00001% Cu, 95% Th remained
after extraction
(Only HNO₃ used for extraction)

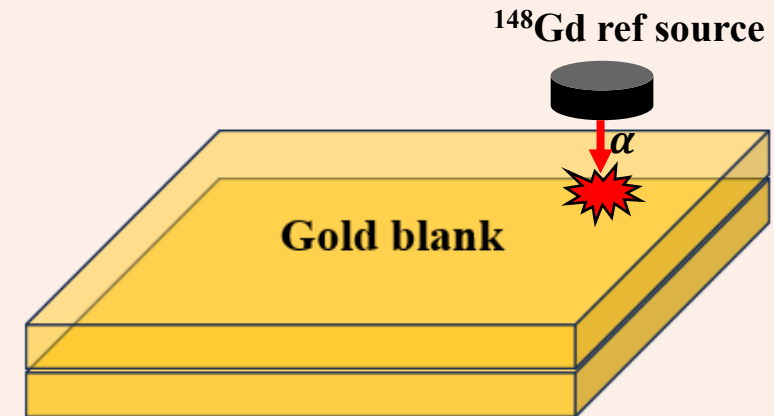
Gold blank purity

^{148}Gd is used for the calibration



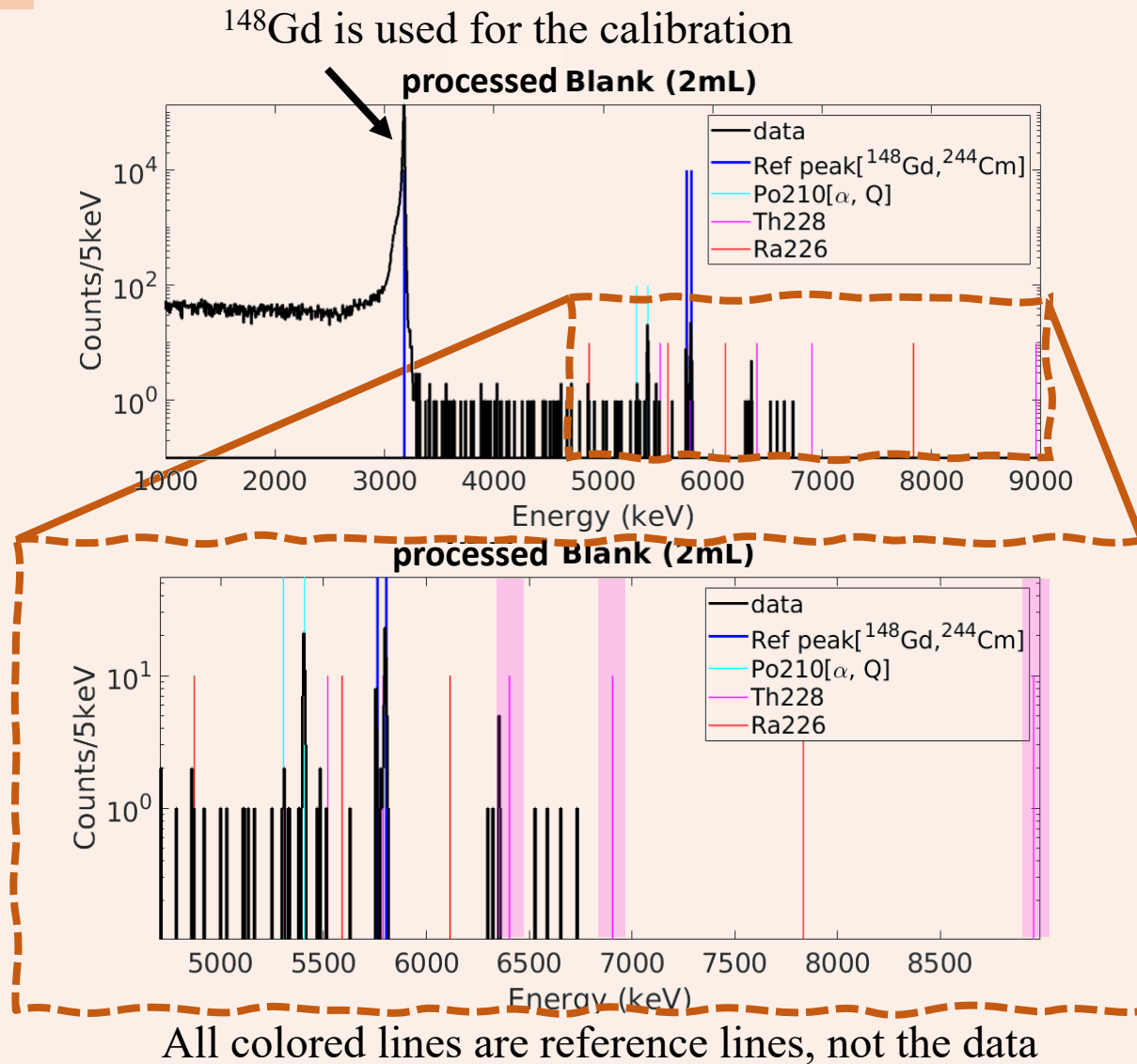
All colored lines are reference lines, not the data

Confirm the gold is clean itself for the ^{228}Th radioassay

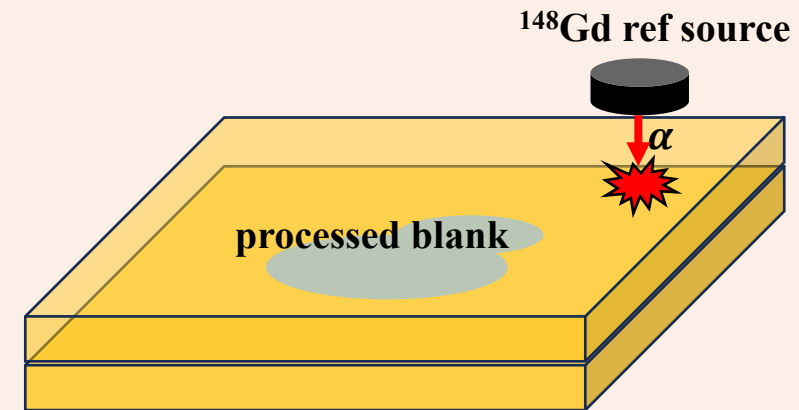


0 counts of ^{228}Th
was observed in **10days** of
gold blanks measurement

Processed blank purity



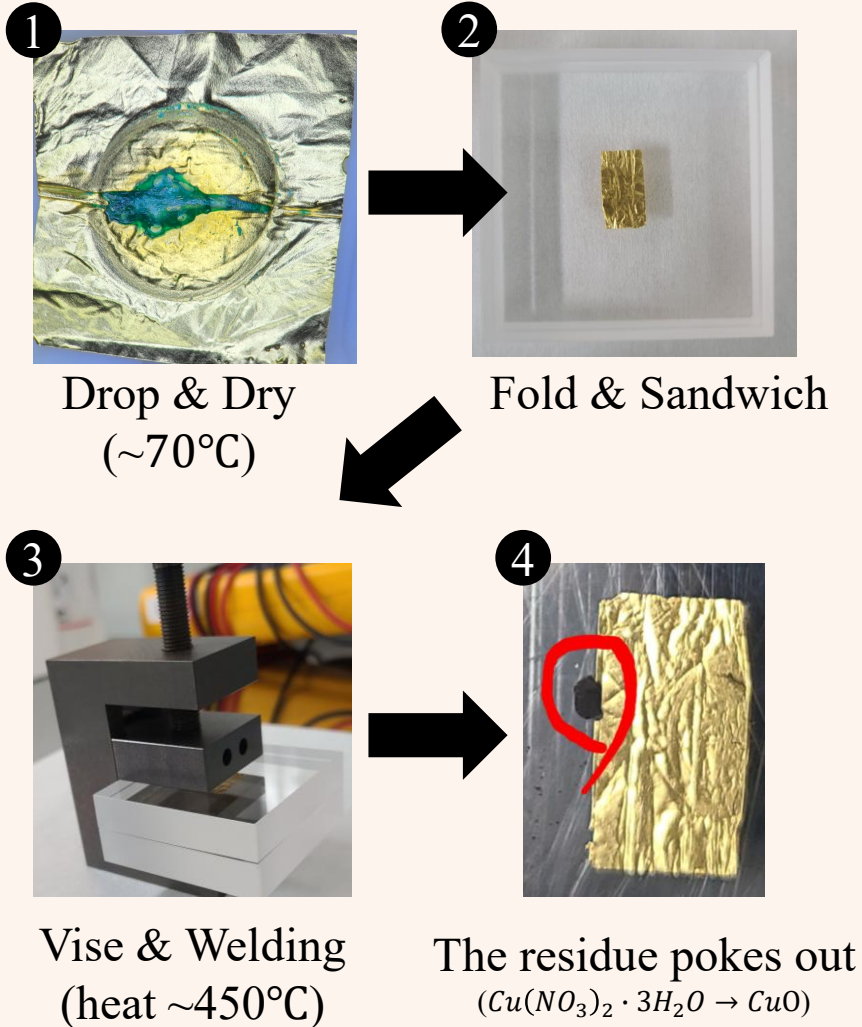
Processed blank undergoes the same procedure as the sample, but without the sample itself



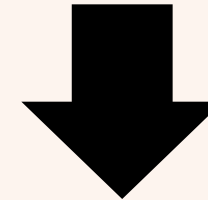
0 counts of ^{228}Th
was observed in **50days**
processed blanks measurement

Foil Sealing for 4π Detection

Original method



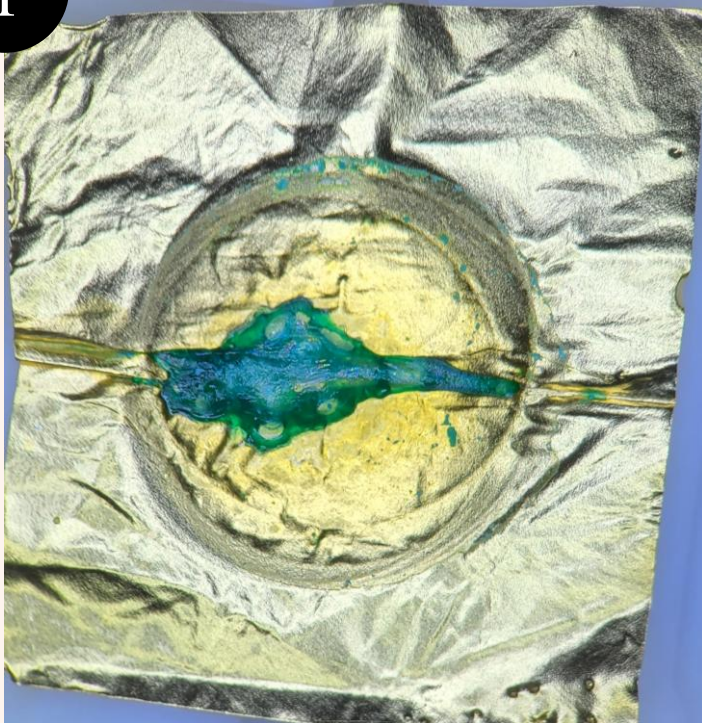
We found that mechanical compression and high-temperature welding cause the residue to protrude from the foil.



1. Do the double extraction to reduce the residue amounts!
2. Change the sealing method!

Reducing the residue amounts

1



Drop & Dry
(~70°C)

double
extraction

1

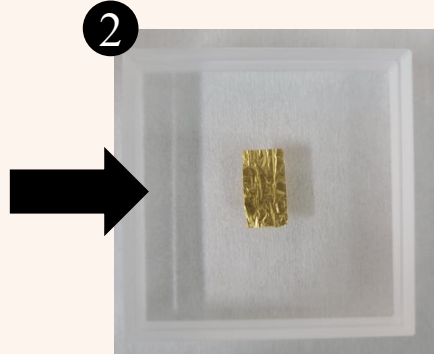


Drop & Dry
(~70°C)

Change the sealing method



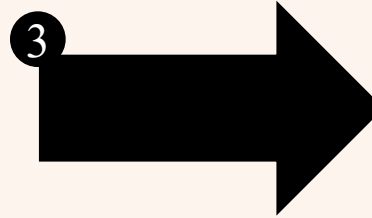
Drop & Dry
(~70°C)



Fold & Sandwich

Idea 1(full drying) :

Drying with 200°C before welding

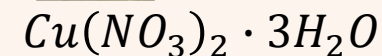
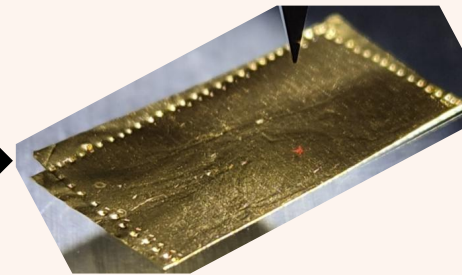
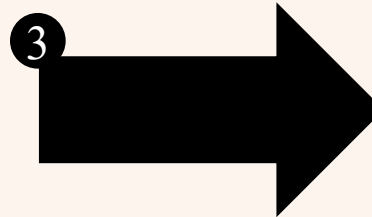


The residue goes like powder

Hard to control

Idea 2(~70°C drying & point welding) :

Point welding without humidity exposure (by bonding machine)



**Easy to control
&
no problem to seal**



Vise & Welding
(heat ~450°C)

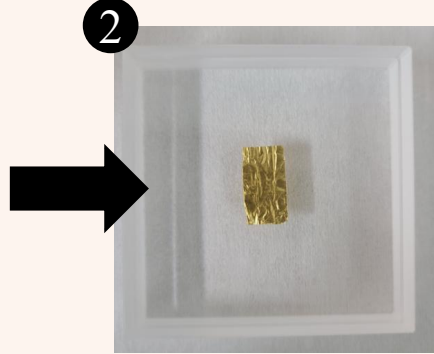


The residue pokes out
($\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O} \rightarrow \text{Cu}(\text{NO}_3)_2$)

Change the sealing method



Drop & Dry
(~70°C)



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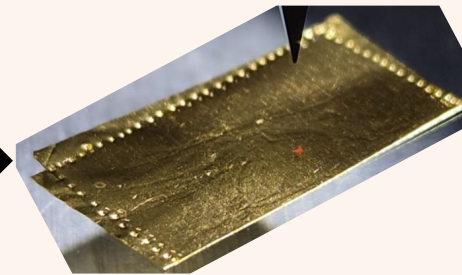
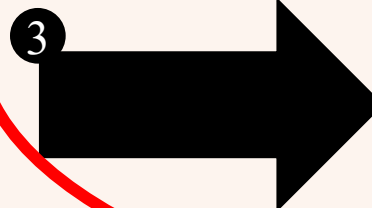
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Hard to control

$\text{Cu}(\text{NO}_3)_2$

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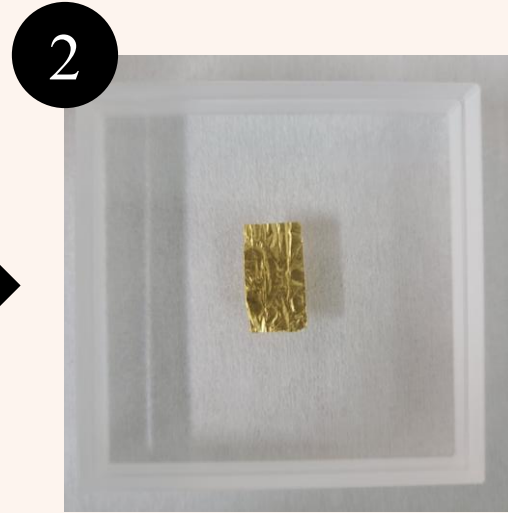
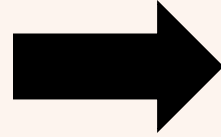
**Easy to control
&
no problem to seal**

$\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$

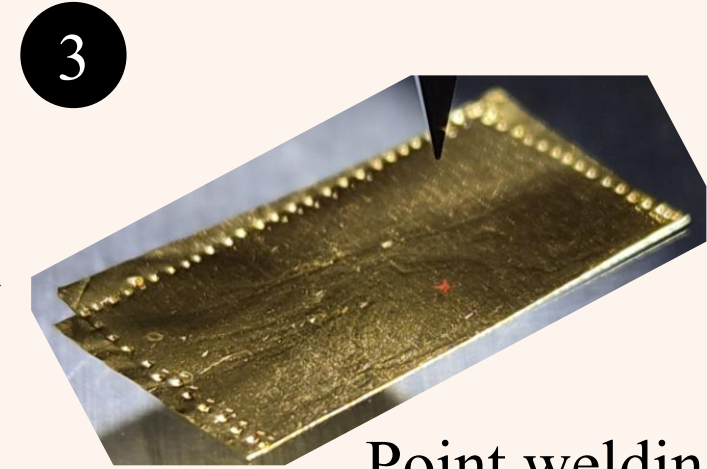
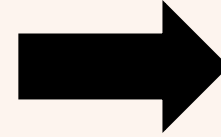
Change the sealing method



Drop & Dry
(~70°C)



Fold & Sandwich

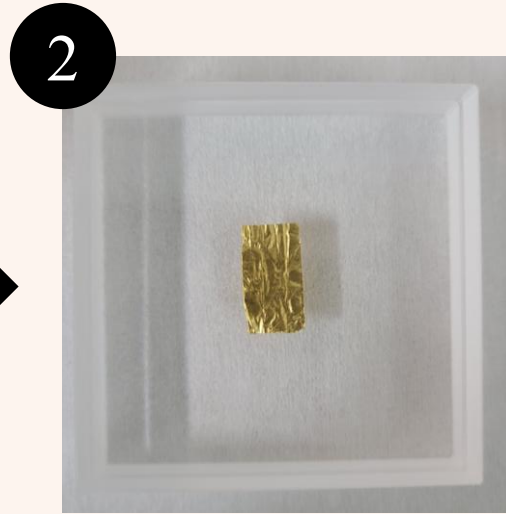
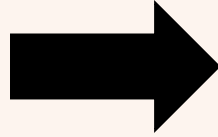


Point welding
without humidity exposure
(by bonding machine)

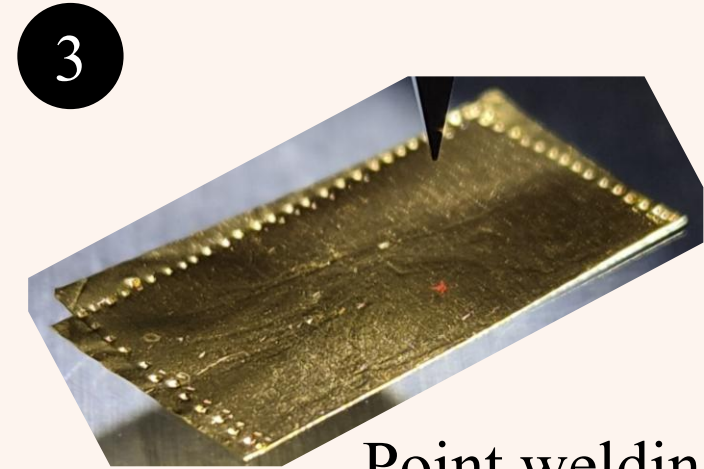
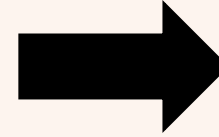
Change the sealing method



Drop & Dry
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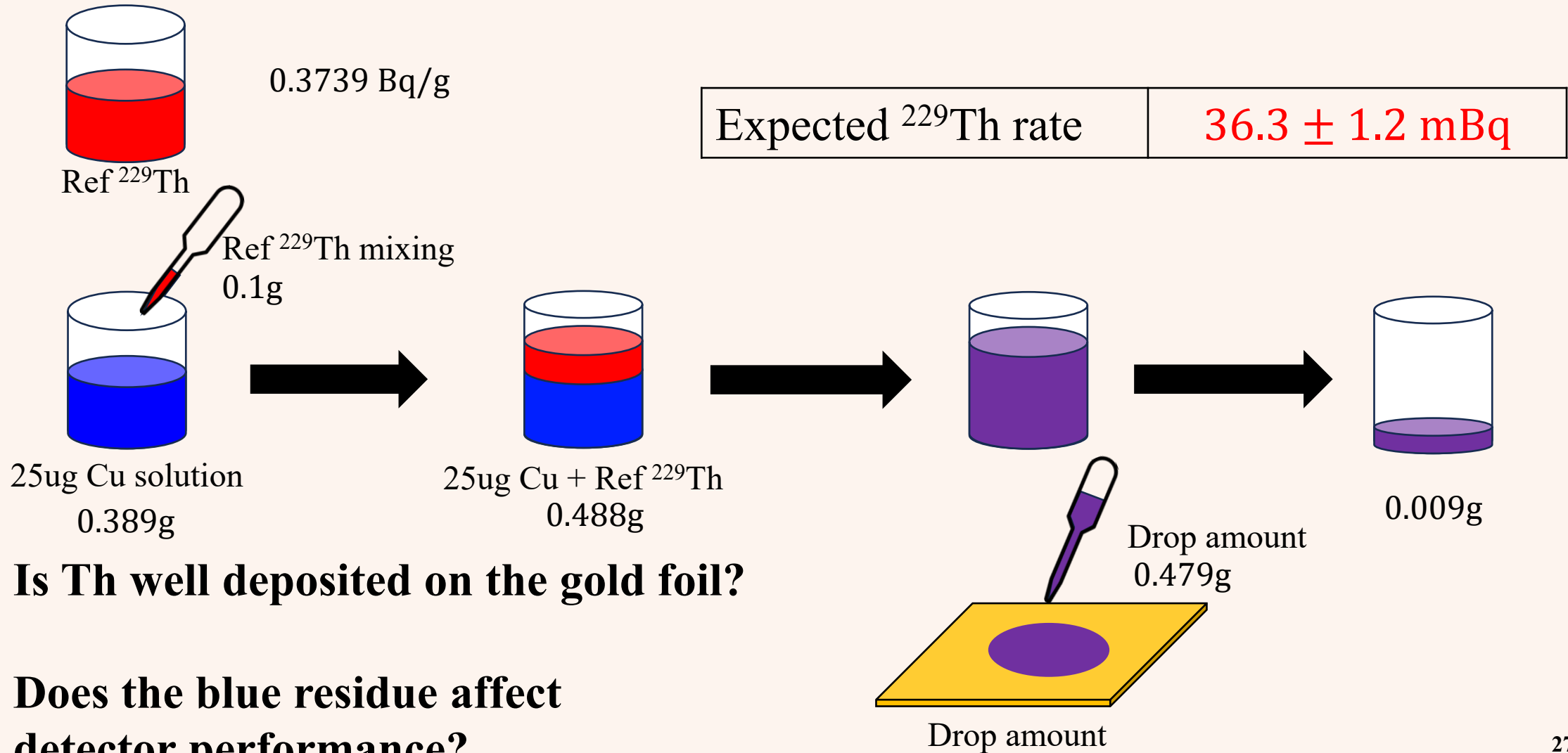


Point welding
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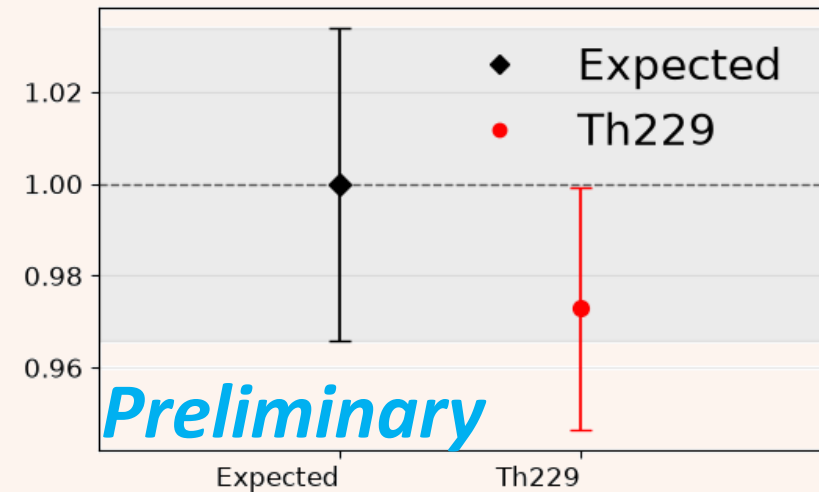
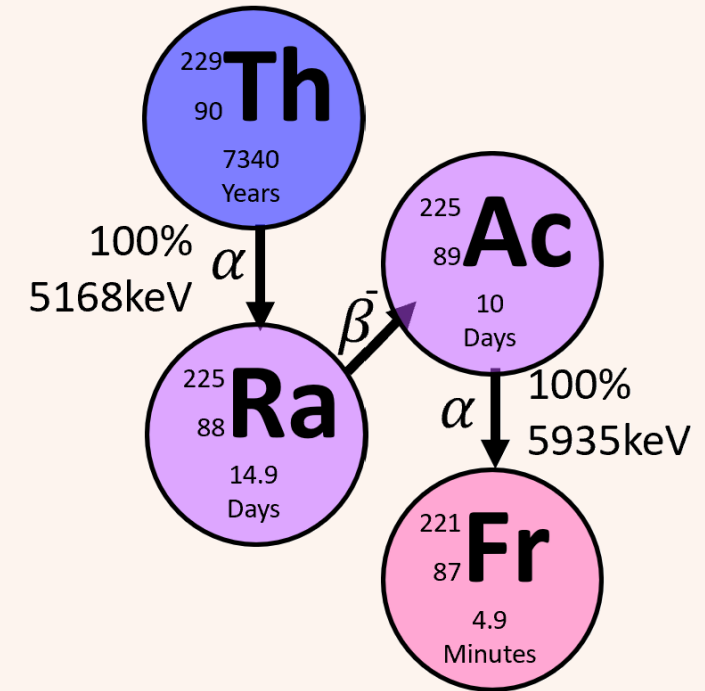
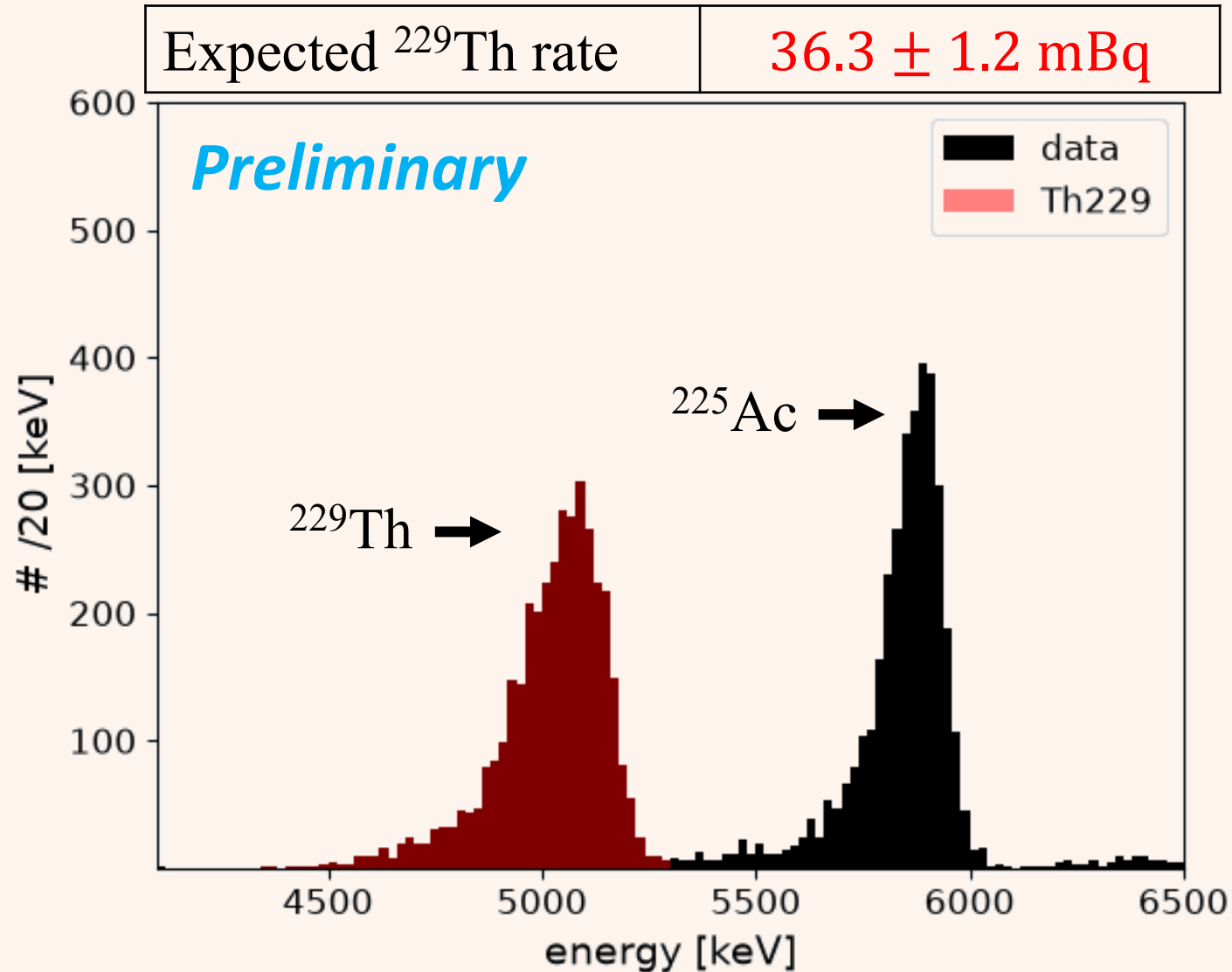
1. Is Th well deposited on the gold foil?

**2. Does the blue residue affect
detector performance?**

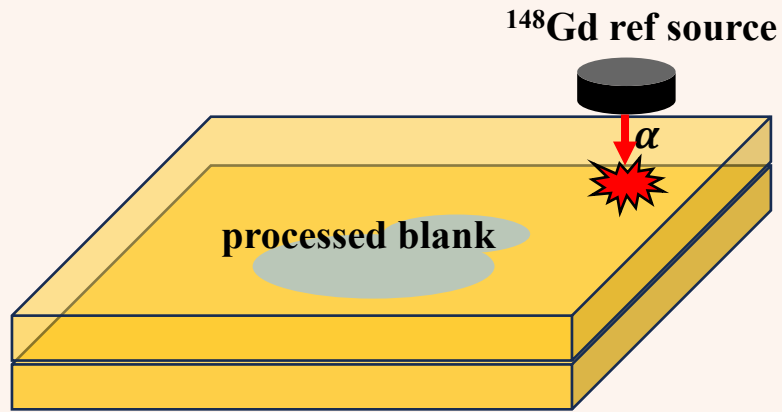
^{229}Th tracer with Cu



^{229}Th tracer with Cu



Potential sensitivity of the DES technique



0 counts of ^{228}Th
was observed in **50days**
processed blanks measurement

- Assume that if we extract the thorium from **50g copper**:
Sensitivity $\rightarrow 11.3 \mu\text{Bq/kg}$ (Bulk copper)
- Assume that if we extract the thorium from **500g copper**:
Sensitivity $\rightarrow 1.13 \mu\text{Bq/kg}$ (Bulk copper)

ONLY 50 days measurements!

Summaries

- **We established a DES-based radio-assay procedure** combining chemical extraction and 4π decay-energy measurement.
- The UTEVA **extraction achieved ~95%** Th recovery, with no ^{228}Th events observed in the process blank.
- The ^{229}Th tracer measurement demonstrated a preliminary **deposition efficiency of ~97%**.
- Our sensitivity can reach to the **sub-uBq/kg level sensitivity** in a month measurements

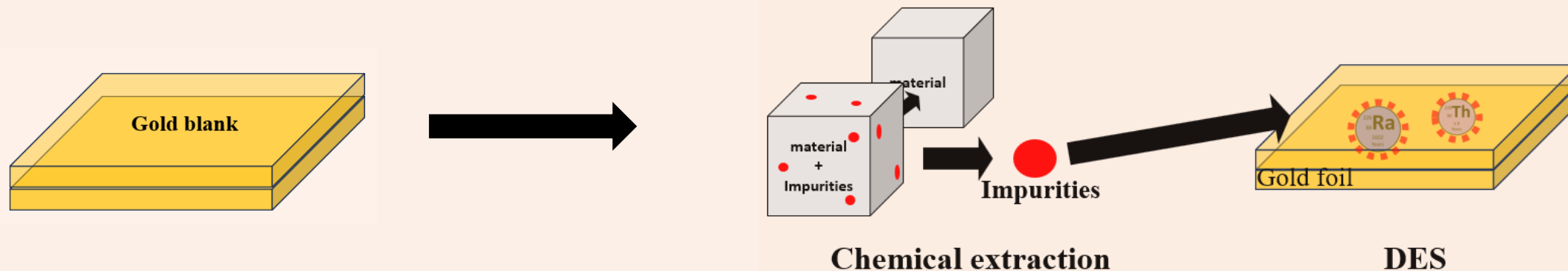
These results demonstrate the feasibility of
DES-based radio-assay for high-purity copper!



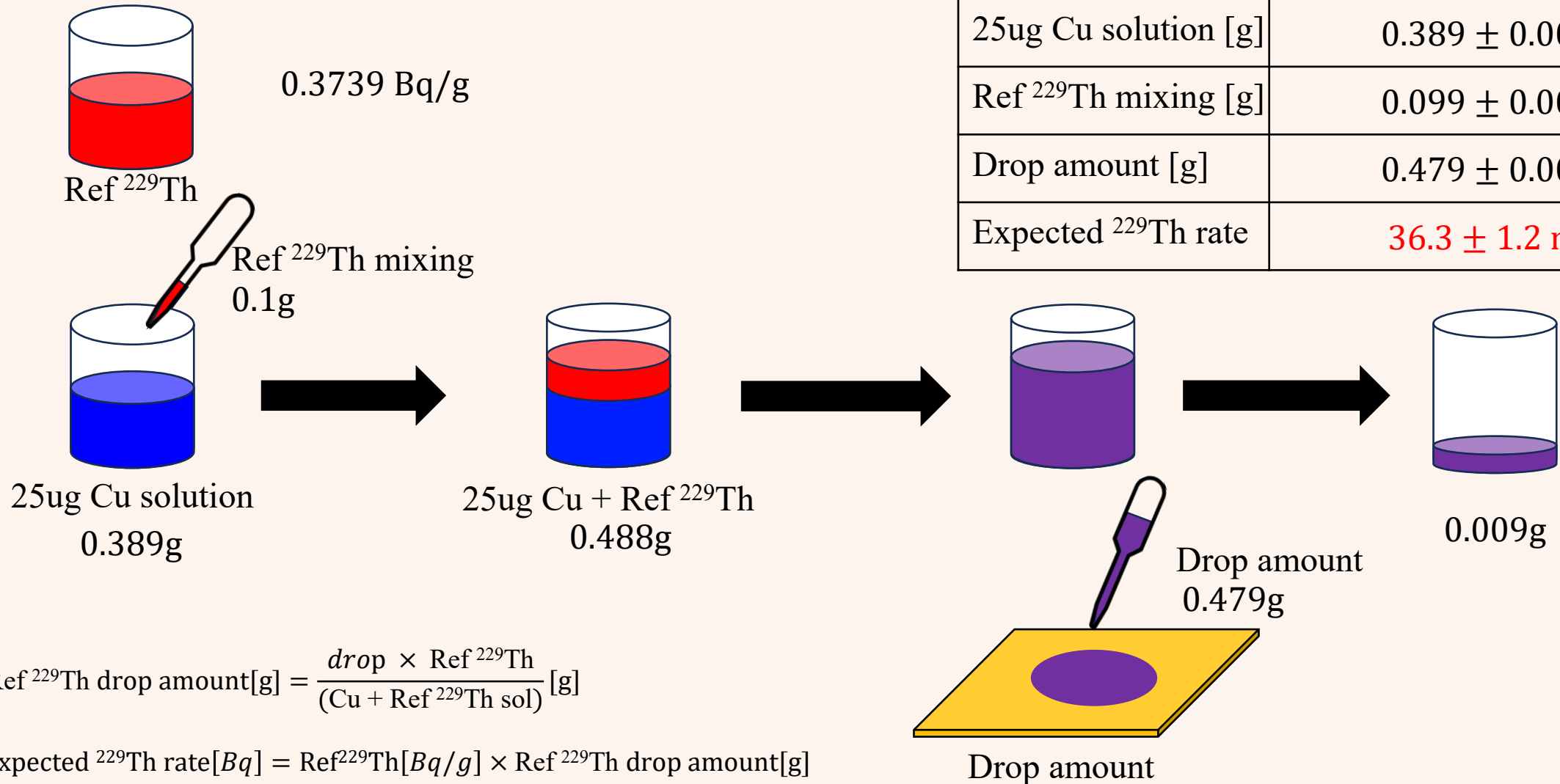
Back up

Summary of the procedure for radio-assay

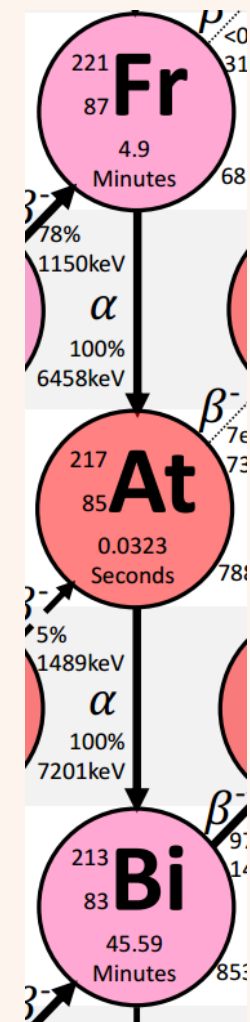
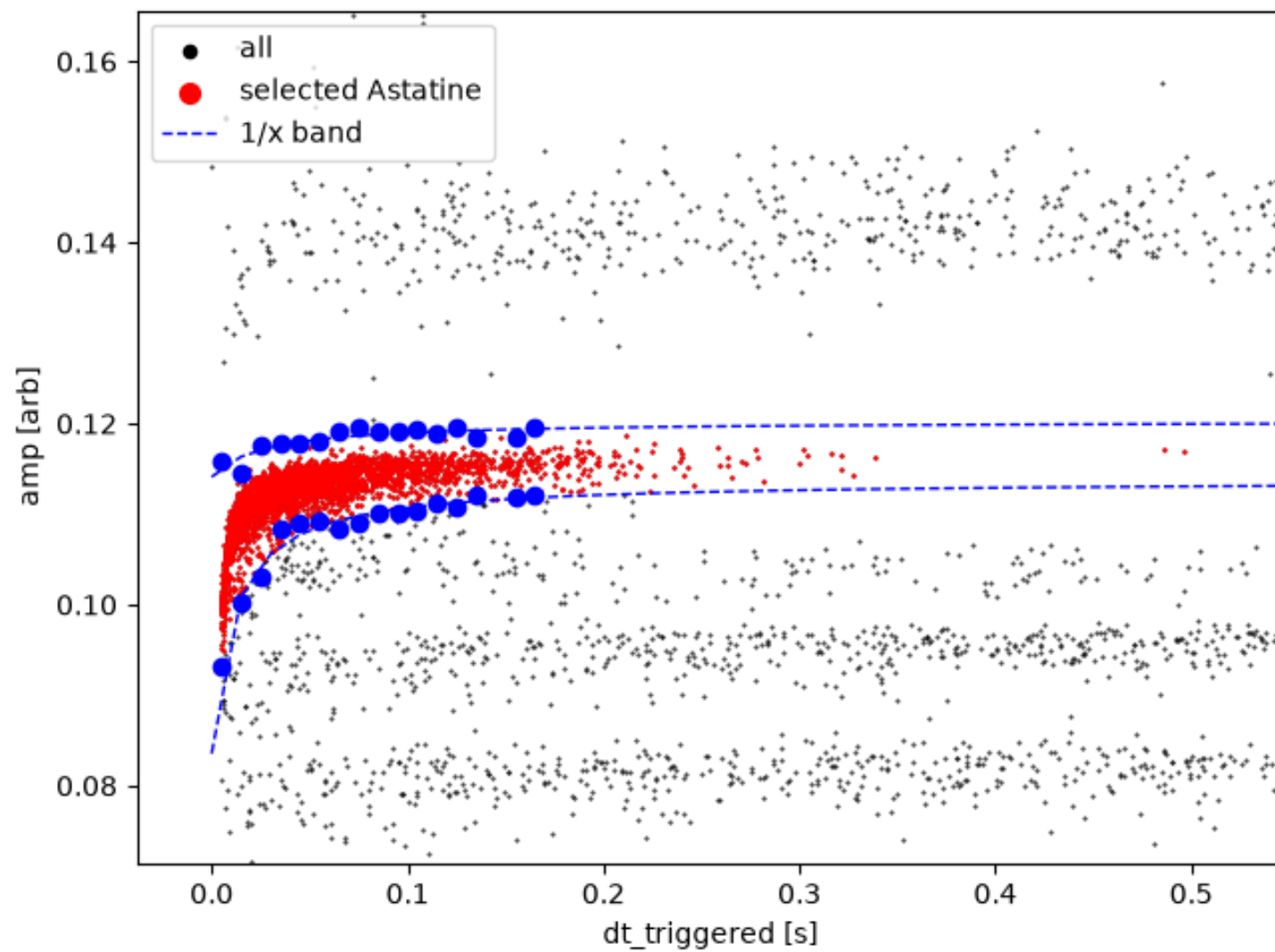
1. **Blank gold foil purity measurement** (~40days)
2. **Chemically extracting**
3. **Impurity solution drop & dry**
4. **Gold foil sealing** with bonding machine (ultrasonic power)
5. **Sample impurity measurement** (~40days)



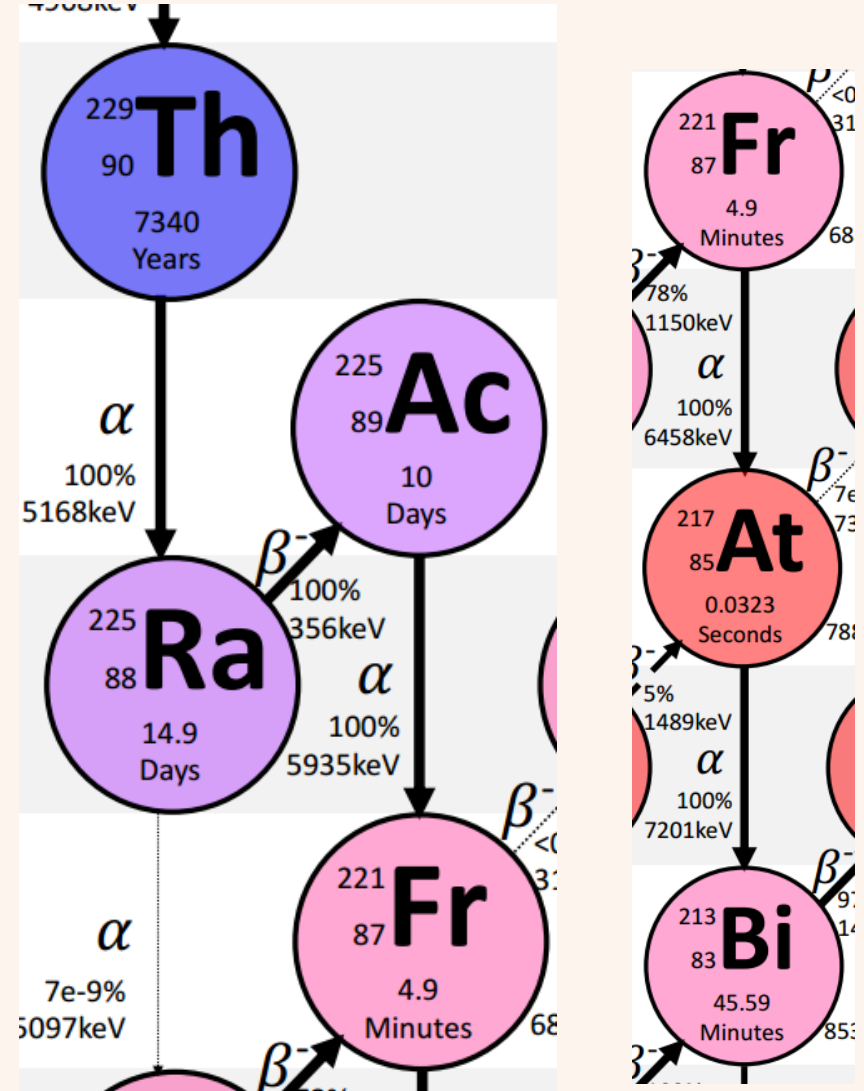
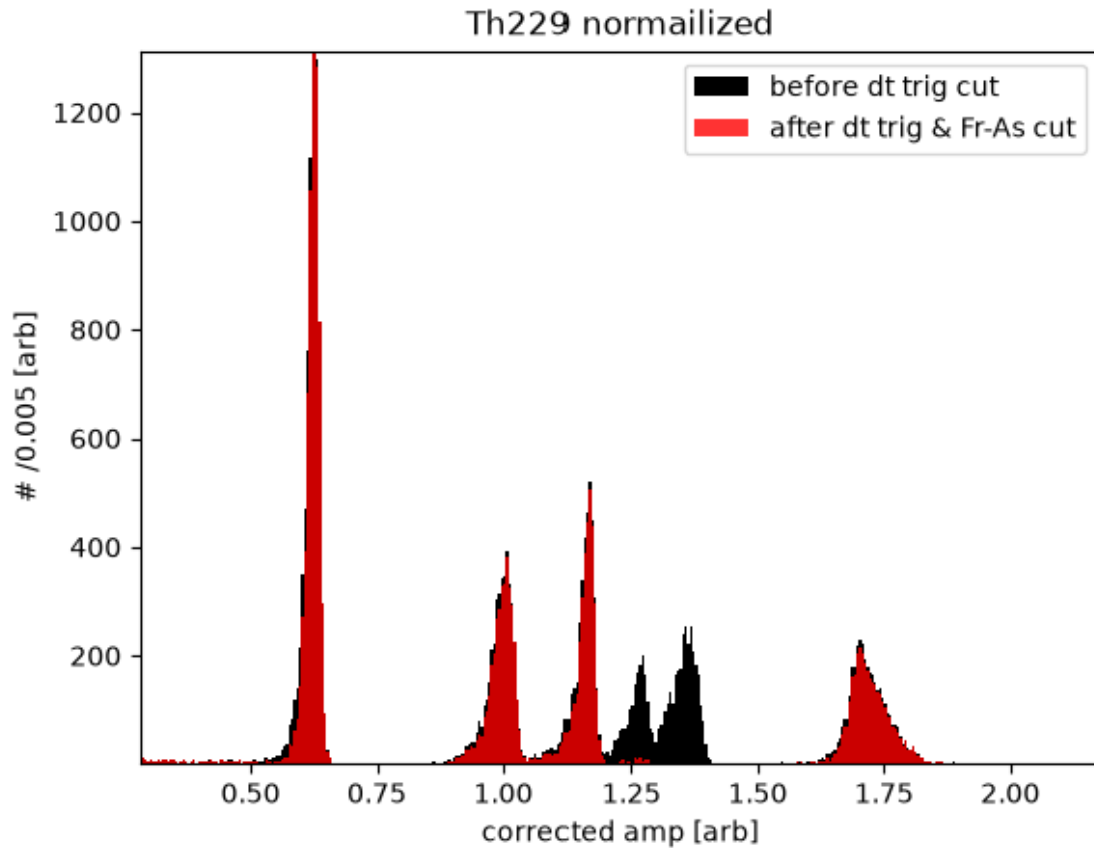
^{229}Th tracer with Cu

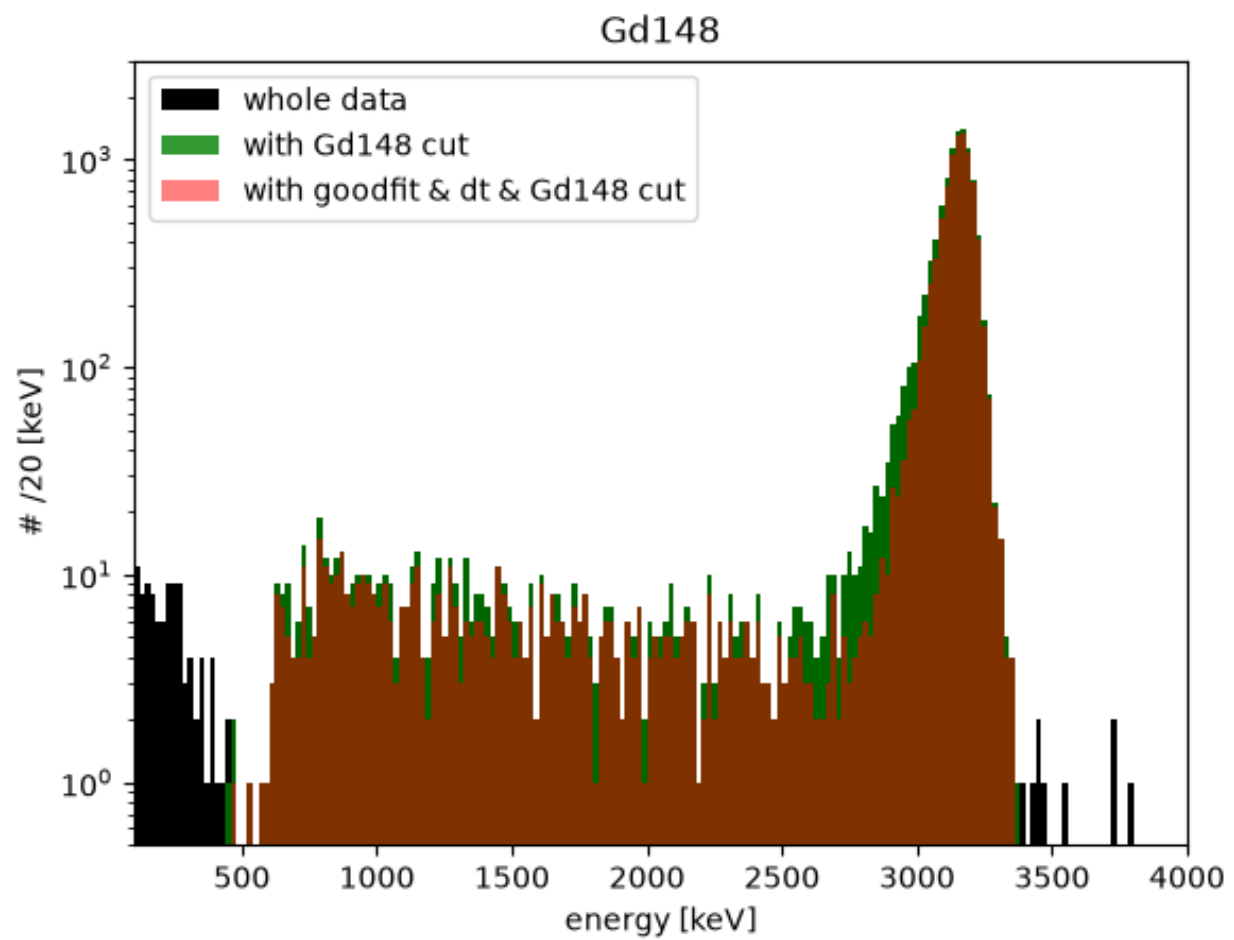


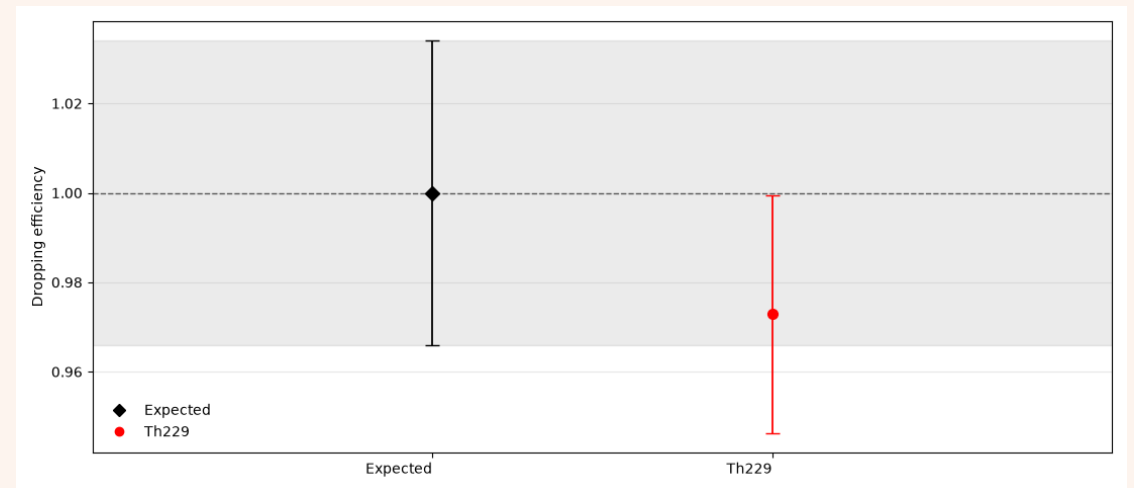
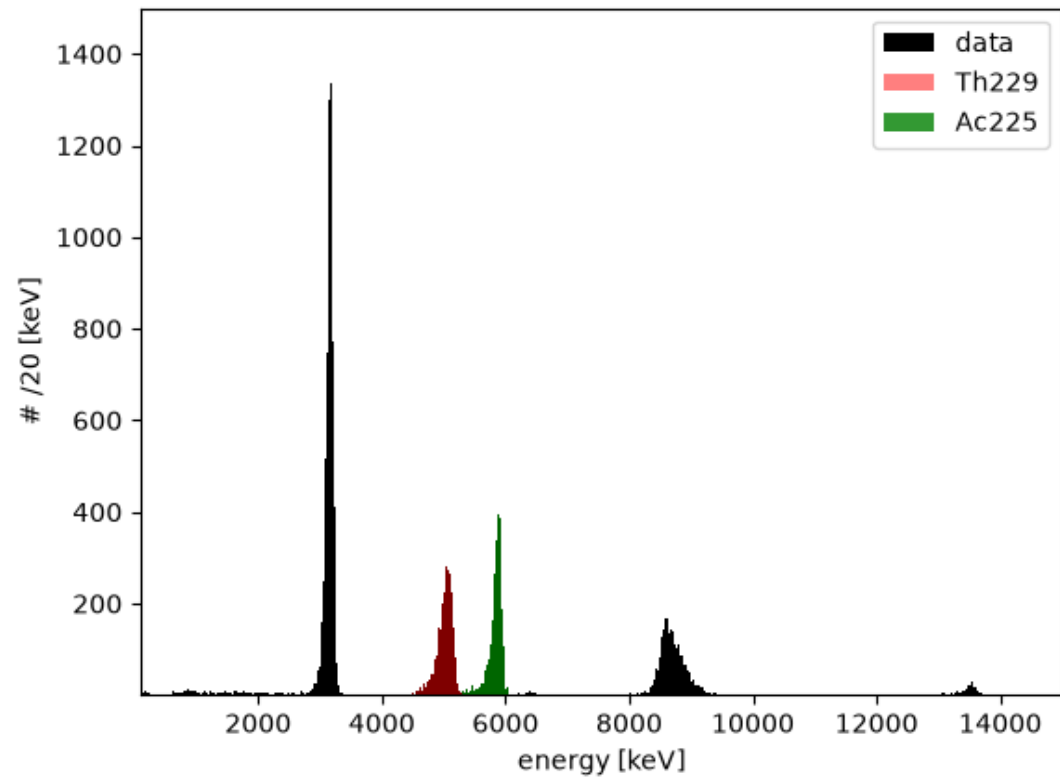
Ref ^{229}Th [Bq/g]	$0.3739 \pm 0.012 \text{ Bq/g}$
25ug Cu solution [g]	$0.389 \pm 0.001 \text{ g}$
Ref ^{229}Th mixing [g]	$0.099 \pm 0.001 \text{ g}$
Drop amount [g]	$0.479 \pm 0.001 \text{ g}$
Expected ^{229}Th rate	$36.3 \pm 1.2 \text{ mBq}$



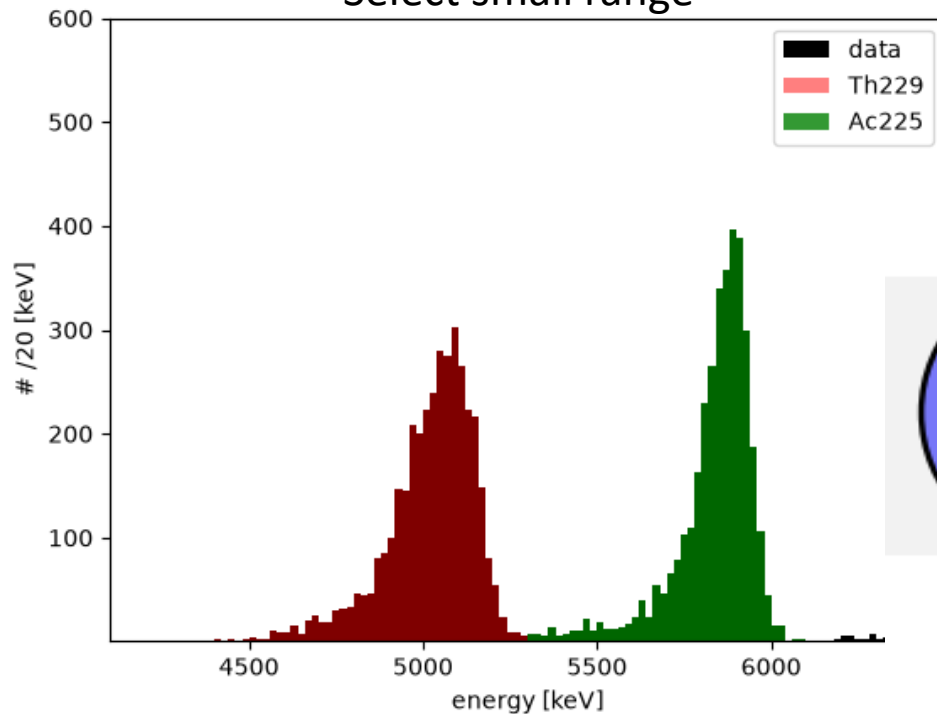
Dropping efficiency test with ^{229}Th 40mBq





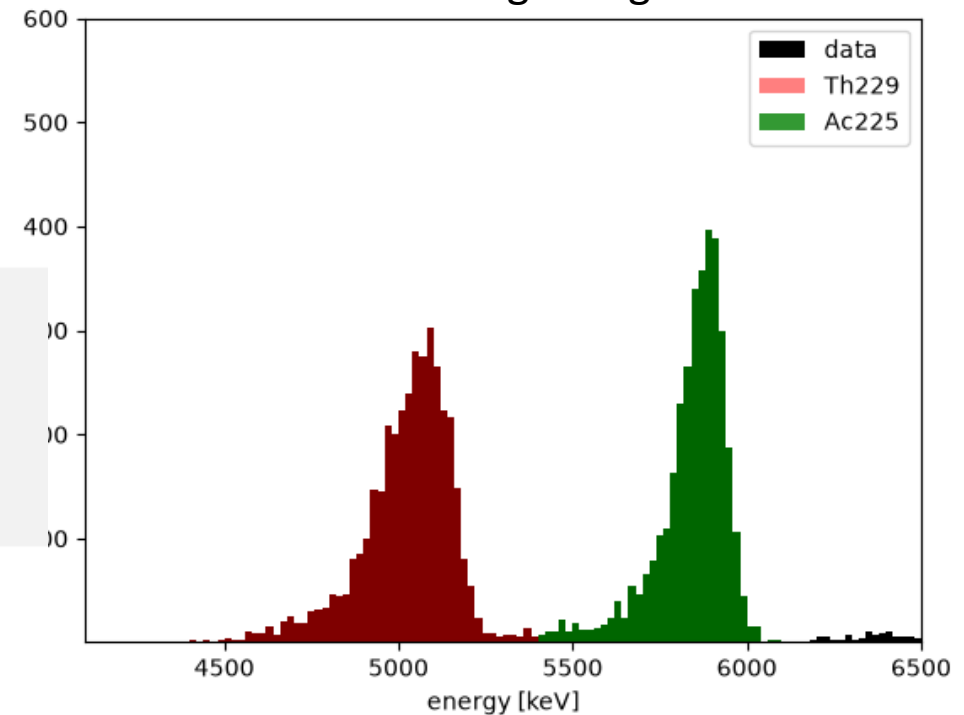


Select small range

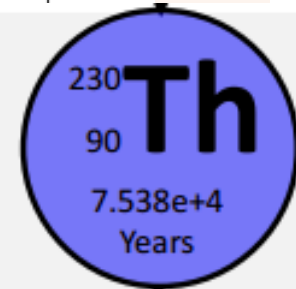


```
is_th229_small = (
    (tmp_calibrated_energy > 4200)
    &(tmp_calibrated_energy < 5300)
)
is_ac225_small = (
    (tmp_calibrated_energy > 5300)
    &(tmp_calibrated_energy < 6100)
)
```

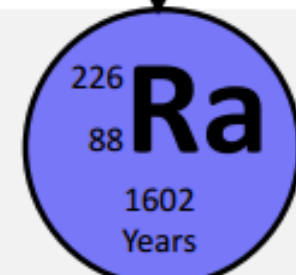
Select large range

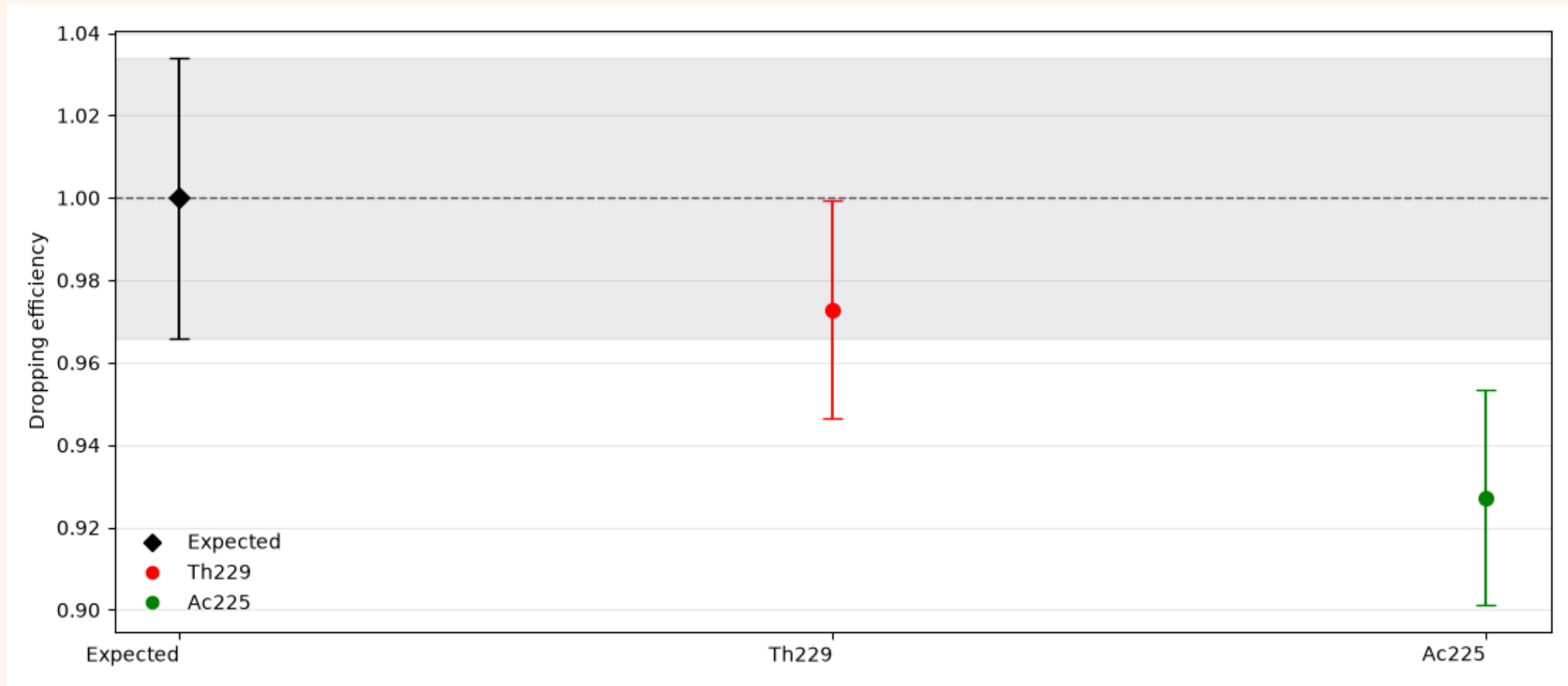


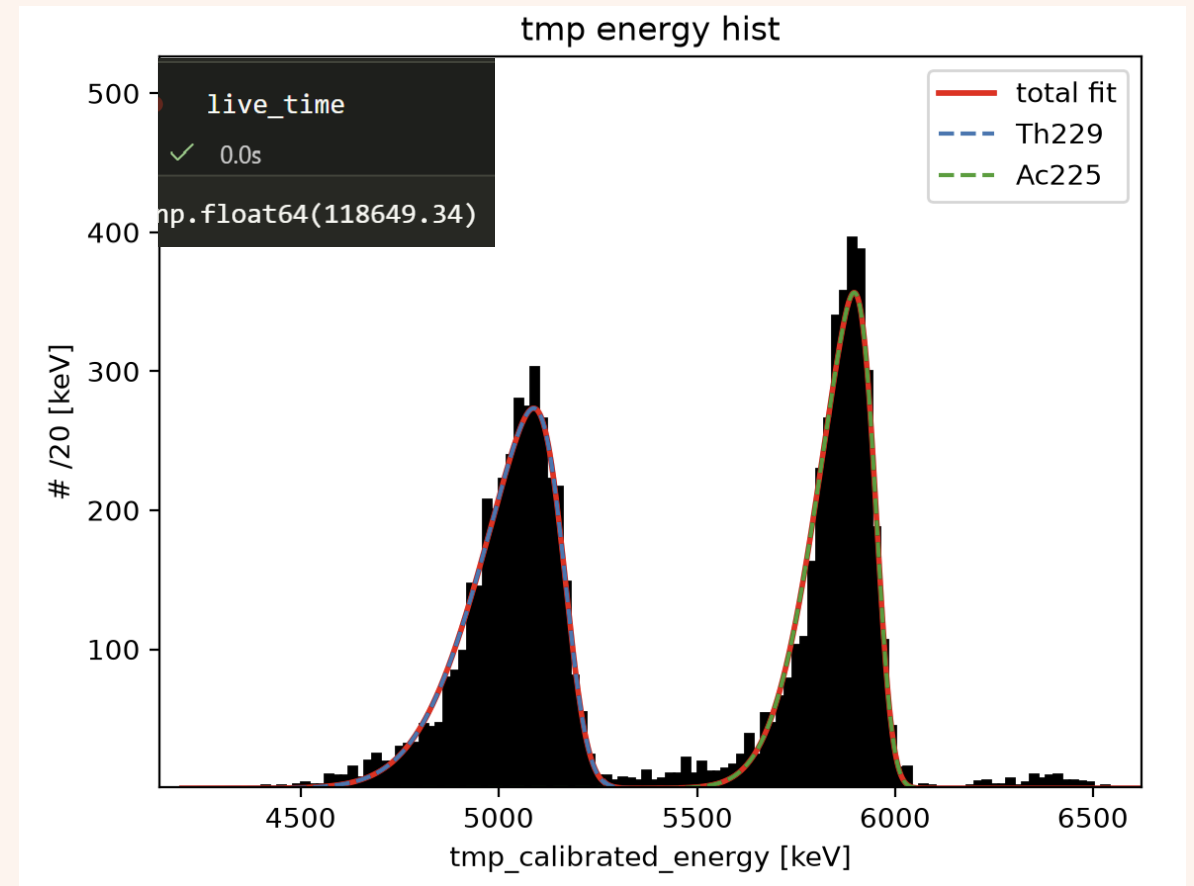
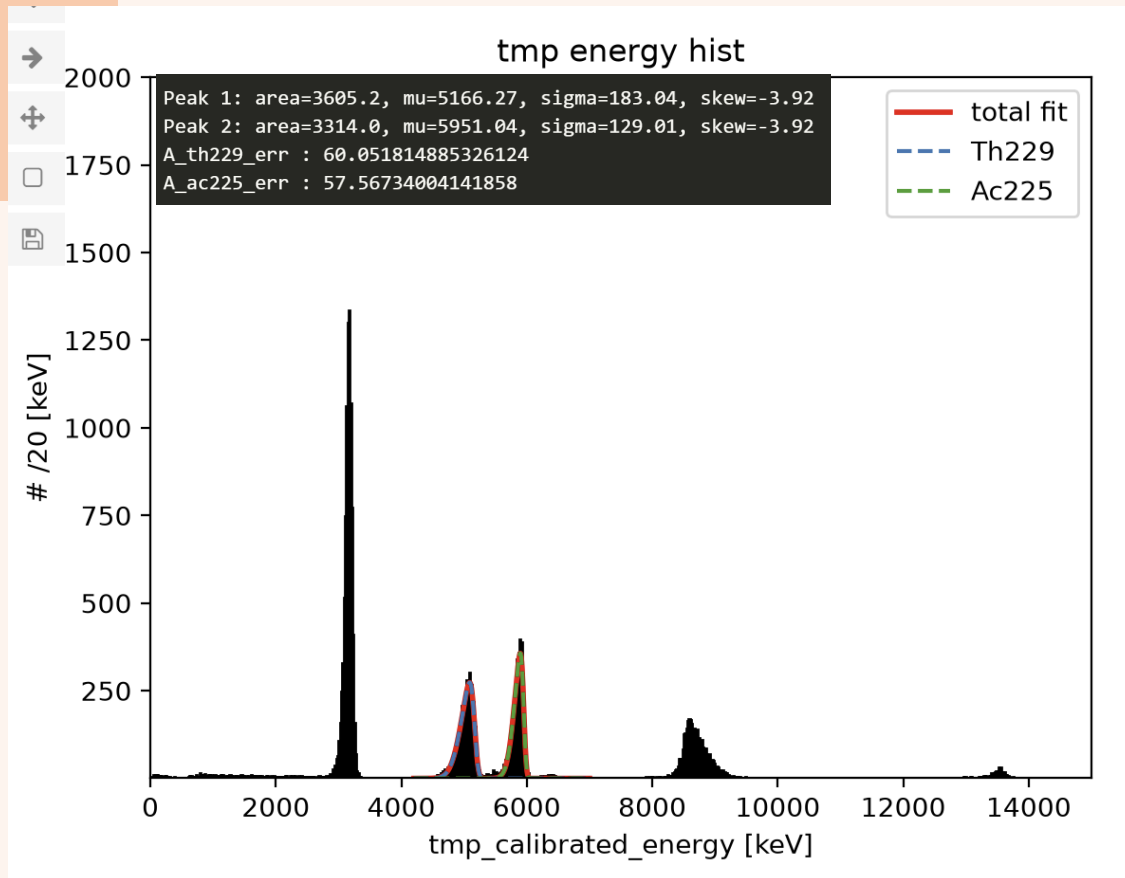
```
is_th229_big = (
    (tmp_calibrated_energy > 4200)
    &(tmp_calibrated_energy < 5400)
)
is_ac225_big = (
    (tmp_calibrated_energy > 5400)
    &(tmp_calibrated_energy < 6100)
)
```



α
100%
4770keV







From fitting results:

Th229 = 3605 +- 60

Ac225 = 3314 +- 57.6

Livetime = 118649.34

Summaries

Gd x-ray

42keV

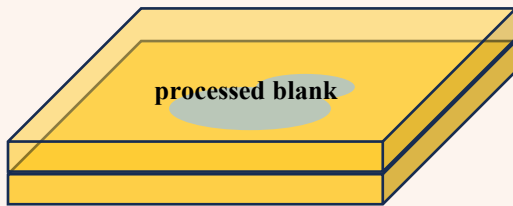
1. Extraction efficiency →

~95% extraction efficiency

<https://doi.org/10.1016/j.apradiso.2023.110673>

2. Purity of processes →

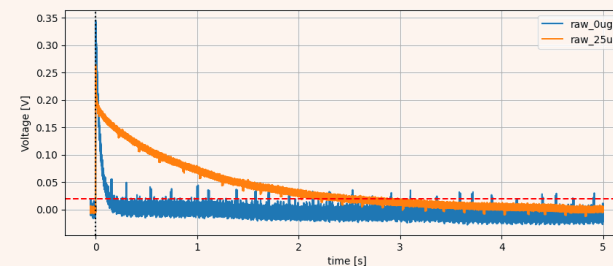
0 counts in our measurement time



3. Dropping efficiency →

~97 %

4. Cu residue affection →



Dropping efficiency test with ^{229}Th 40mBq

Set 1,
0ug Cu



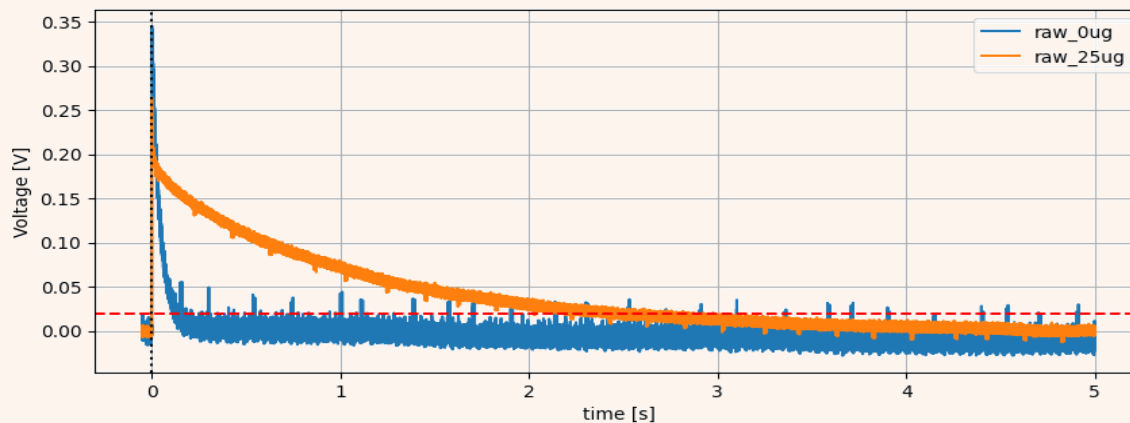
Set 2,
25ug Cu



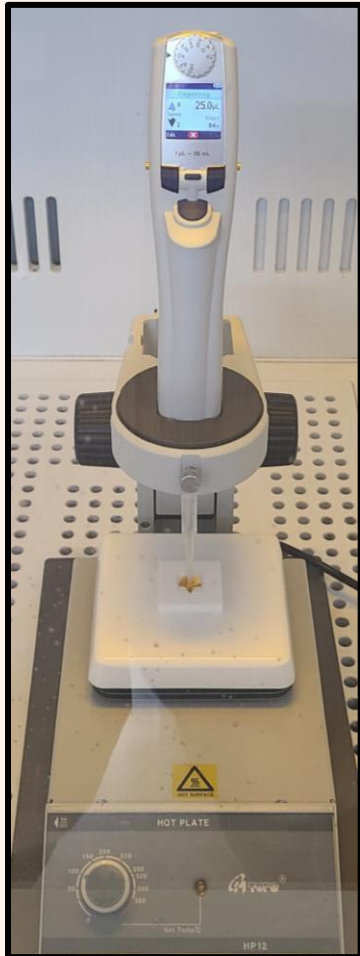
Set 3,
50ug Cu



Sensor dead

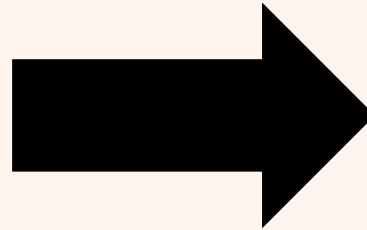


Development of auto drop machine



The pipette has no option to drop automatically (manually push the button every 8min, 100 times...)

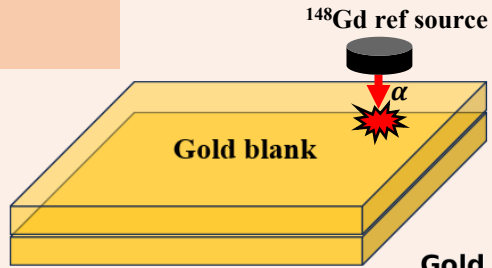
**Button
pusher!**



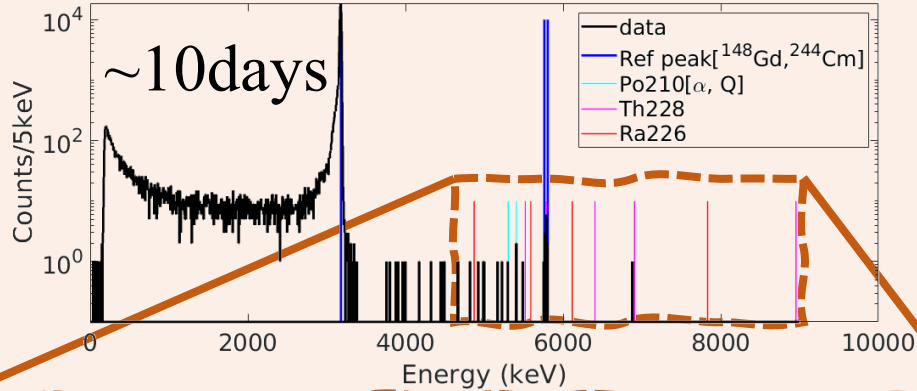
Test drop was already done

Blanks & processed purity

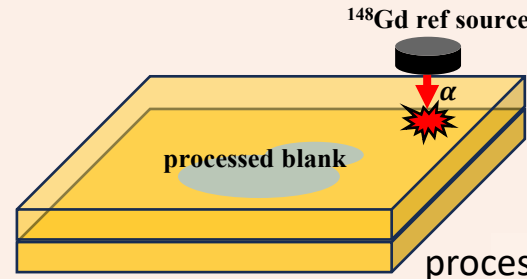
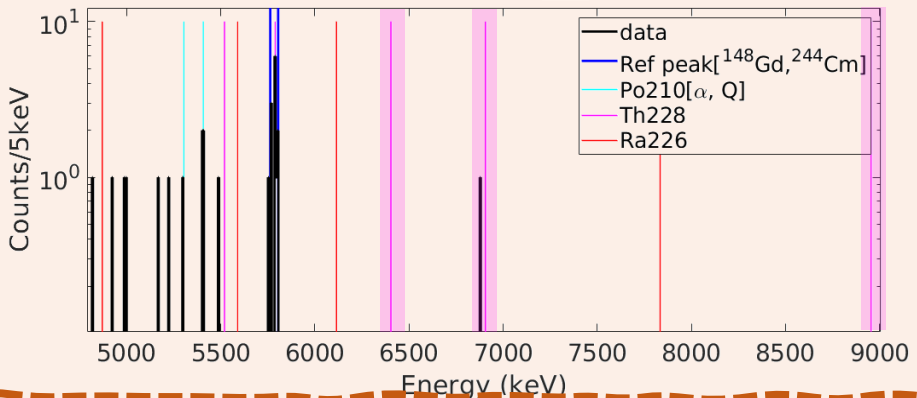
0 counts of ^{228}Th was observed in gold blanks or process blanks.



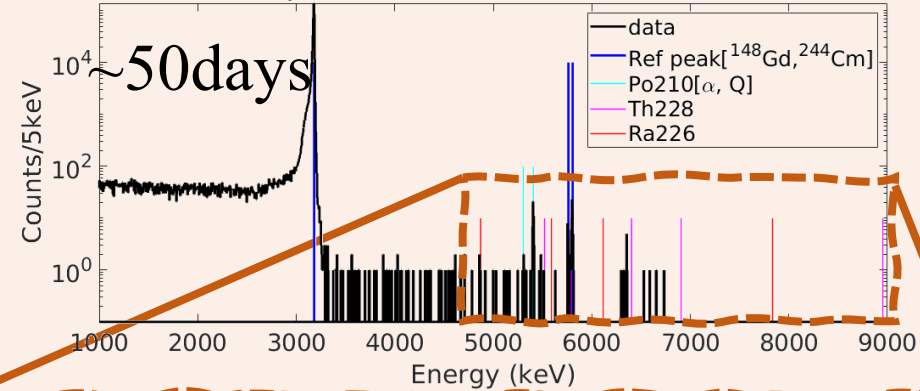
Gold Blank



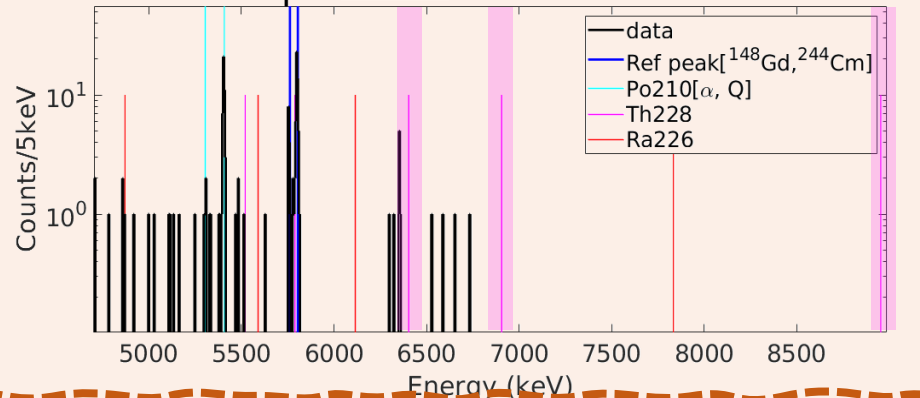
Gold Blank



processed Blank (2mL)

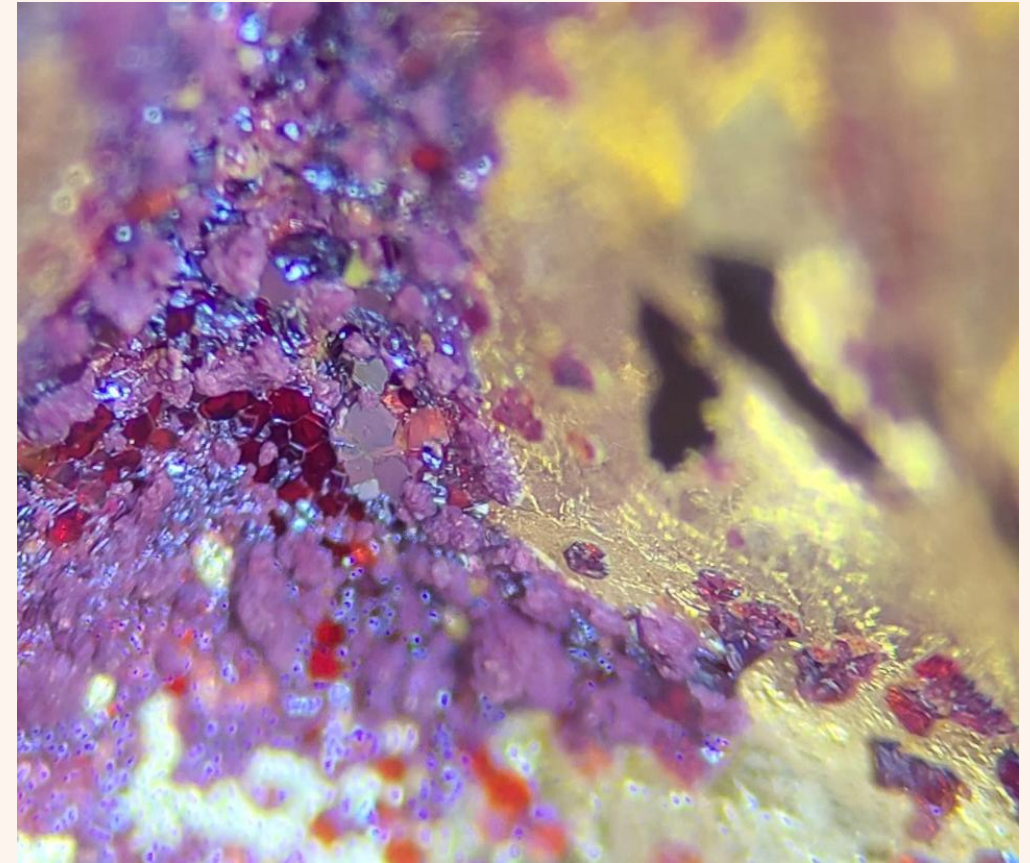


processed Blank (2mL)

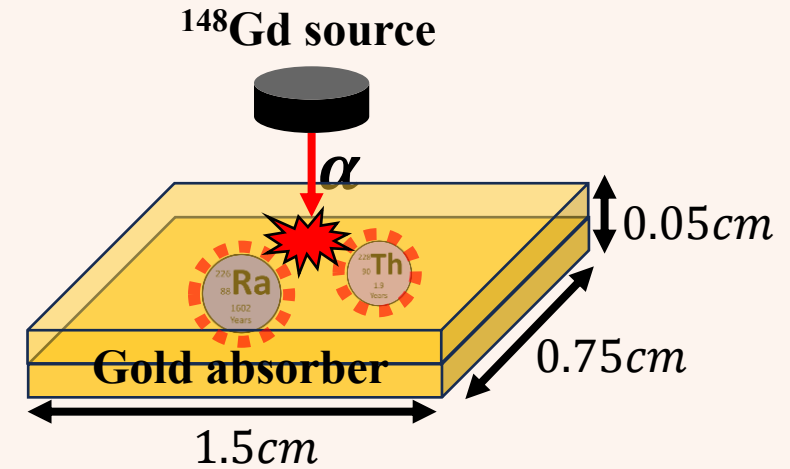
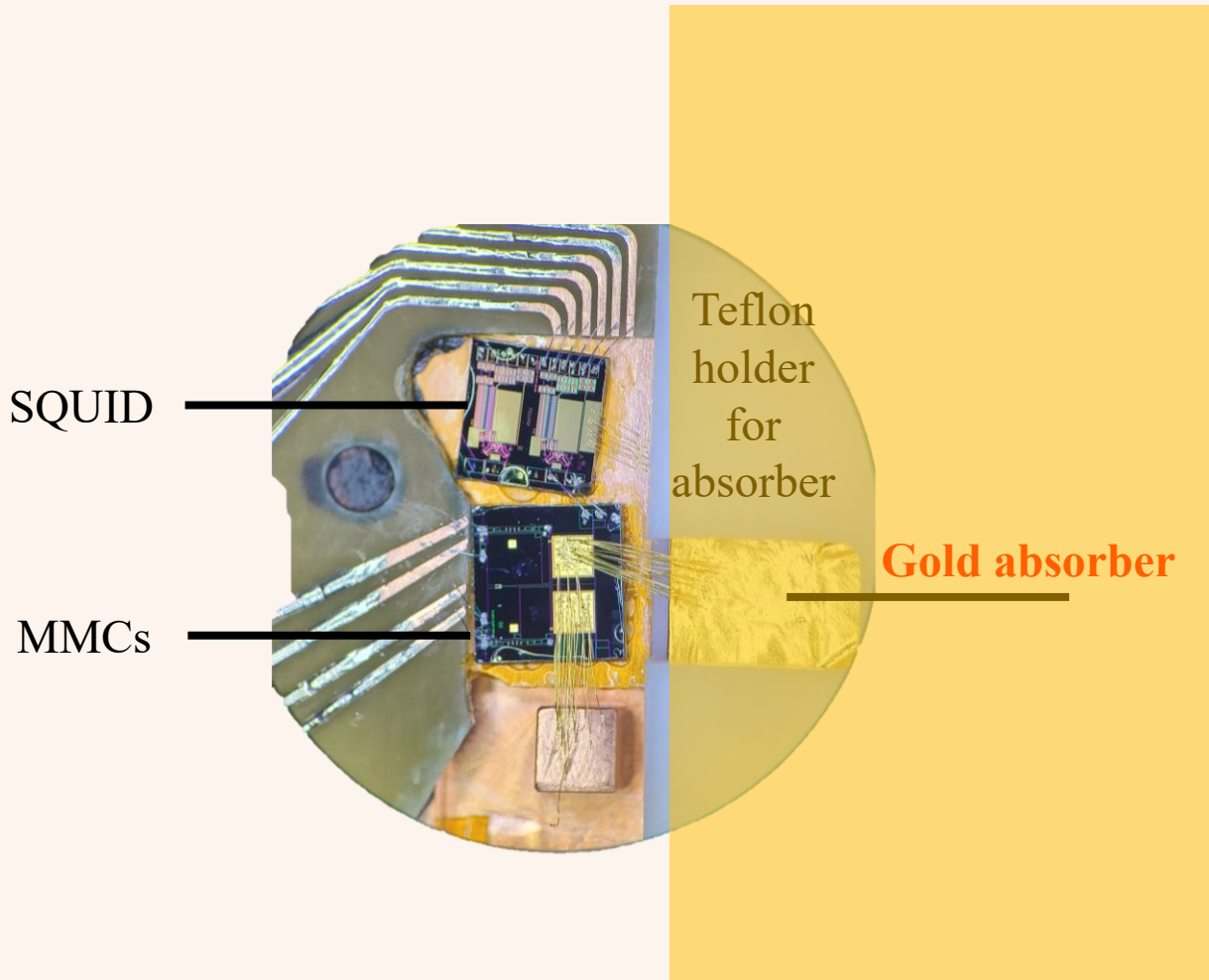


Some discovery

- $\text{HCl} + \text{HNO}_3$ can melt the gold and make red residue
- $\text{HCl} + \text{Cu}$ can exchange with gold and make red residue
- Our dropping solution have **both of that conditions** → **Sad**
- We will change eluting solution as HNO_3 ,
But it will reduce extraction efficiency (~80%)



Detector configuration

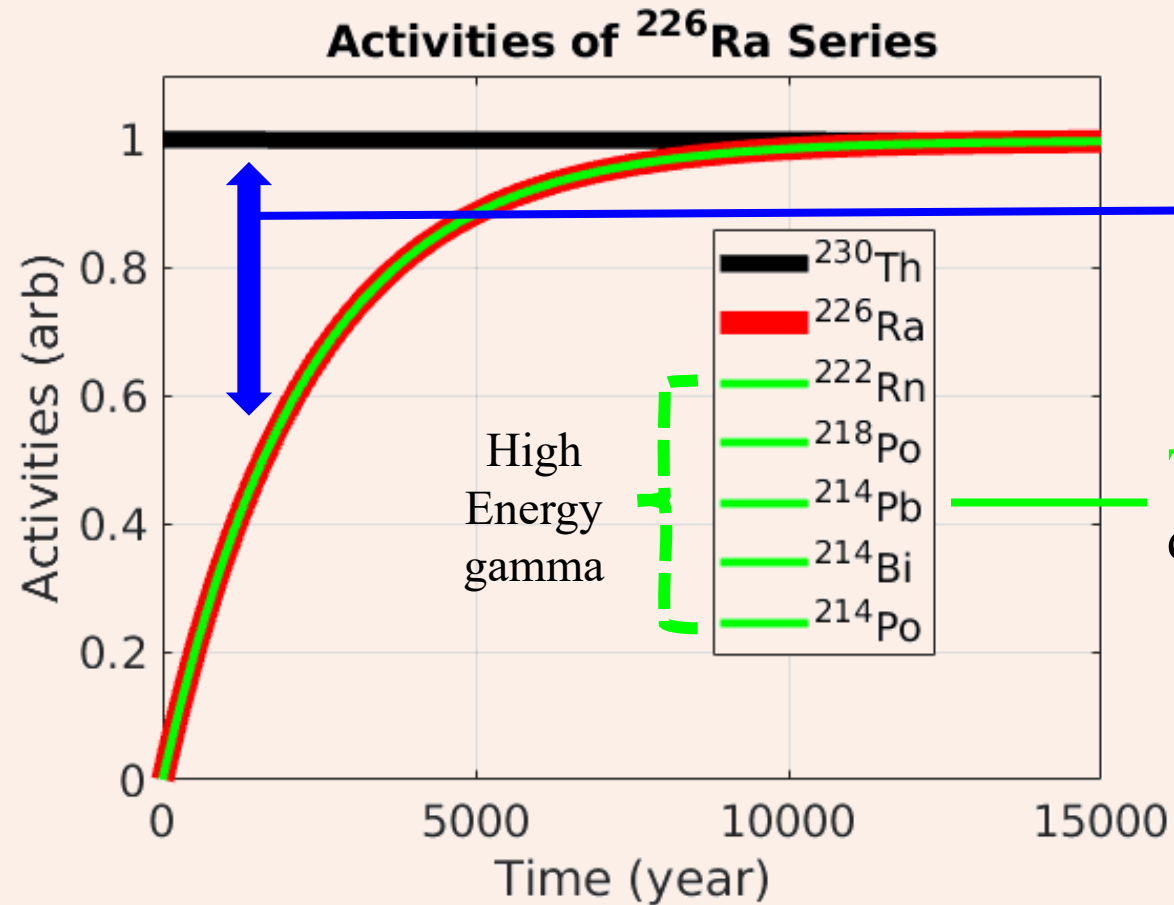
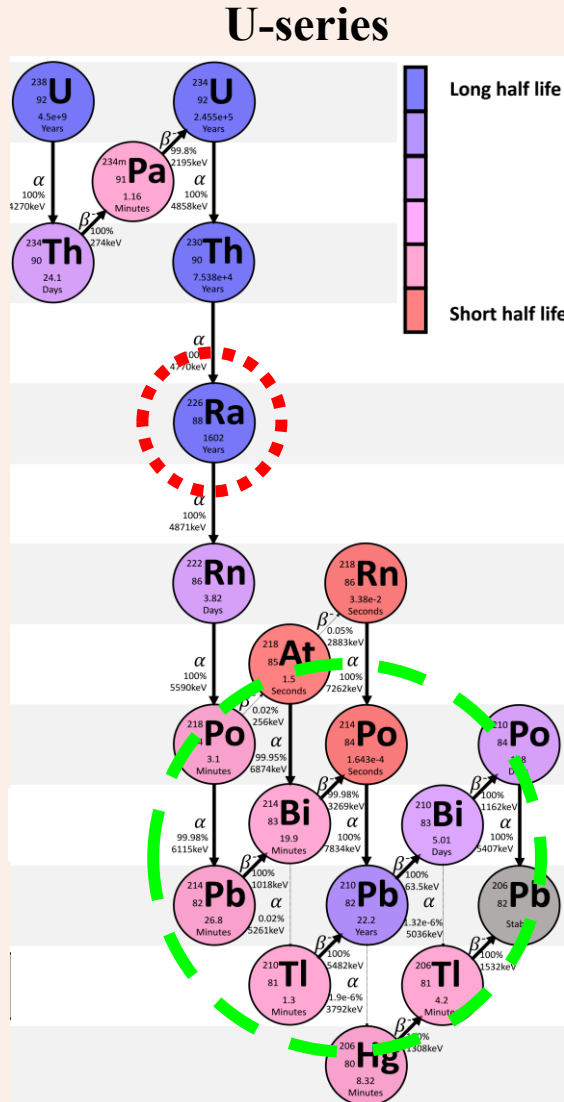


<i>Absorber</i>	Gold foil [1.5 cm × 1.5 cm × 0.05 cm]
<i>Absorber Holder</i>	2 plates of PTFE(Teflon)
<i>Calibration source</i>	3182 keV α of ^{148}Gd
<i>Temperature sensor</i>	Magnetic microcalorimeters (MMC's & SQUID)

Reference

- UTEVA 95% extraction efficiency
→ <https://doi.org/10.1016/j.apradiso.2023.110673>
- How we choose the absorber size & confirming the detection methods
(Previous work)
→ **DOI:** [10.1109/TASC.2024.3515965](https://doi.org/10.1109/TASC.2024.3515965)

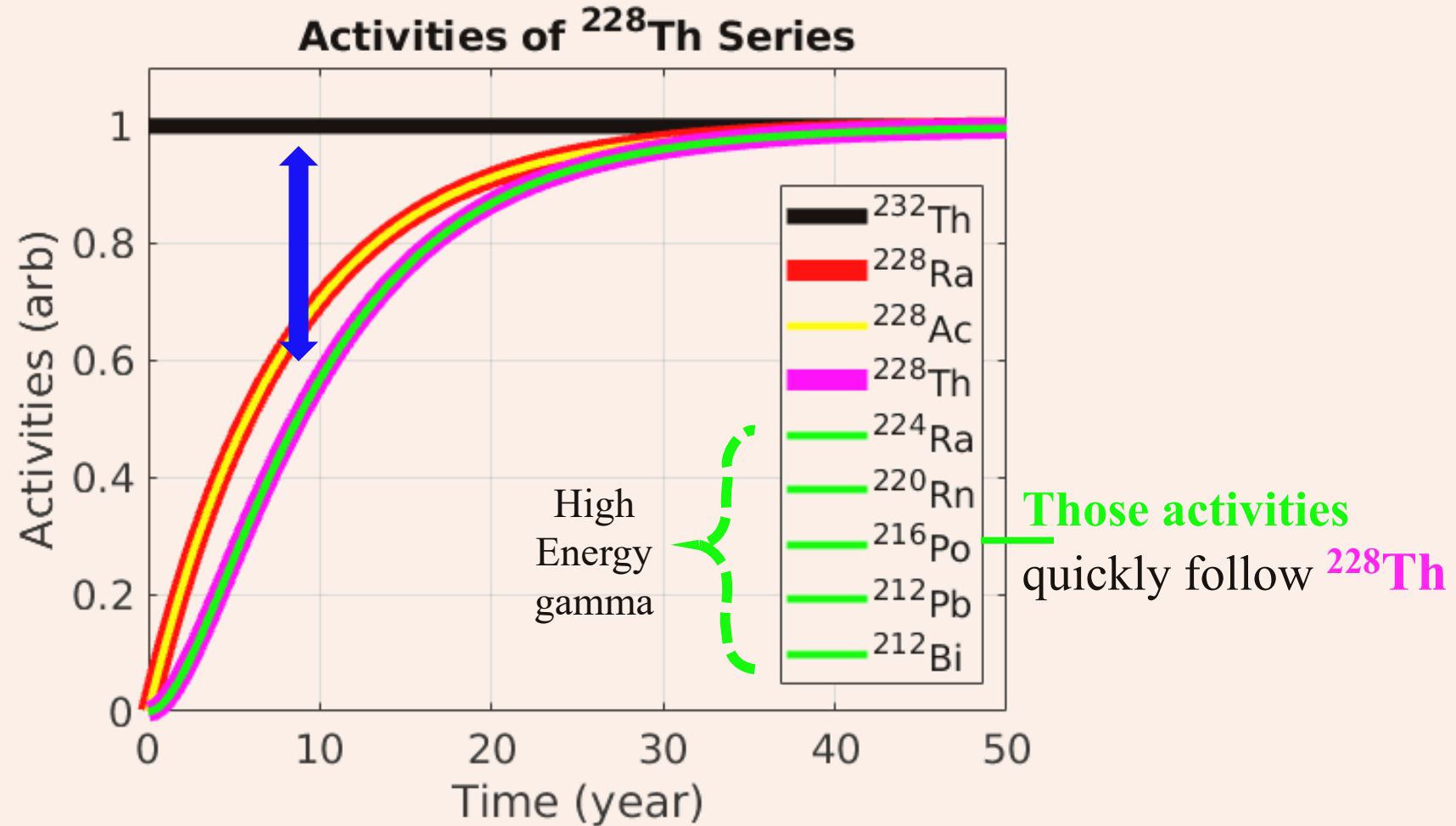
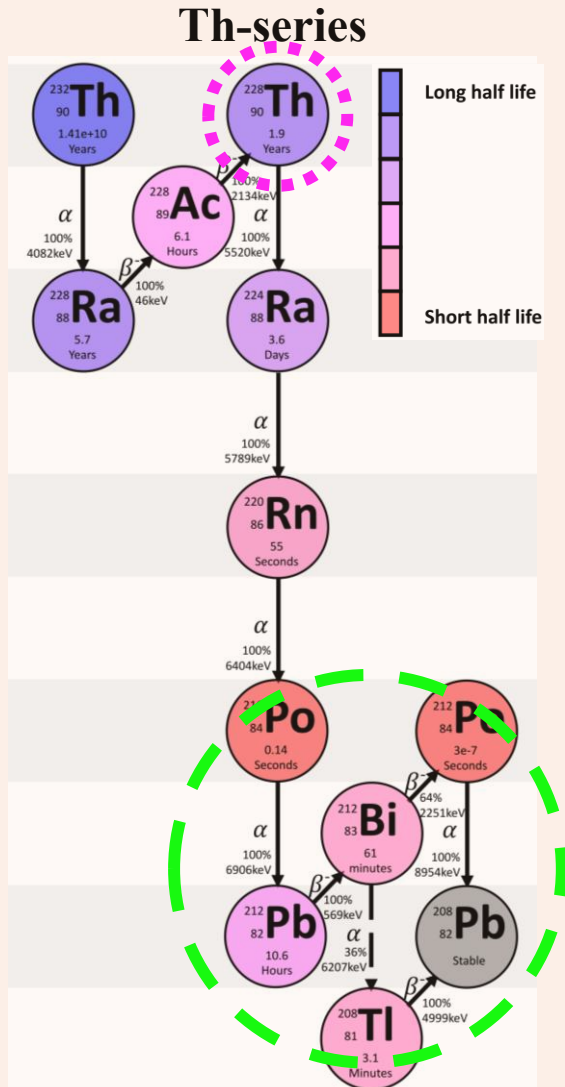
Why ^{226}Ra & ^{228}Th is important



Their activity can be different

Those activities quickly follow ^{226}Ra

Why ^{226}Ra & ^{228}Th is important



Back up

A Unified Approach to the Classical Statistical Analysis of Small Signals

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(November 26, 2024)

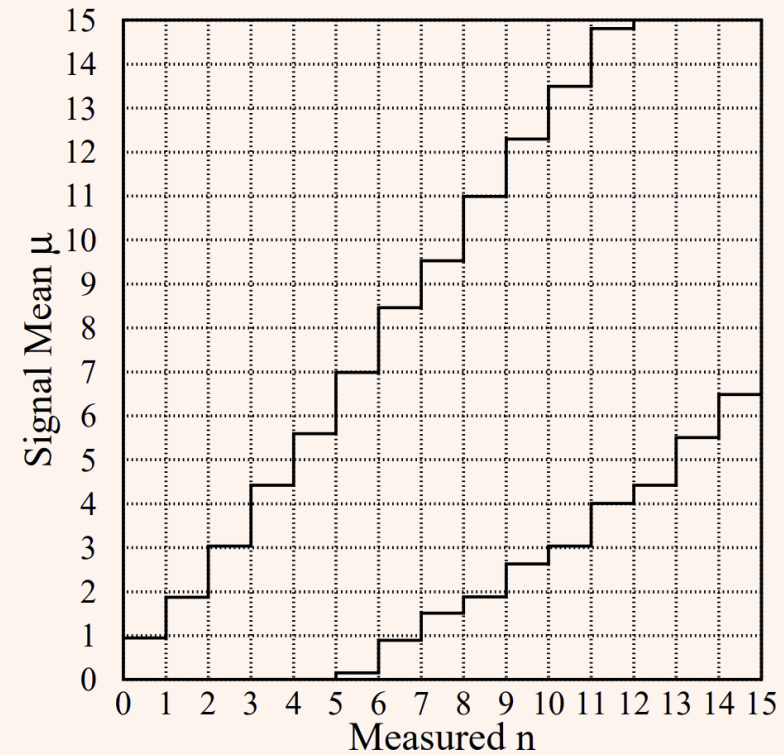


FIG. 7. Confidence belt based on our ordering principle, for 90% C.L. confidence intervals for unknown Poisson signal mean μ in the presence of Poisson background with known mean $b = 3.0$.

Counts If events are in the 95% contour

The **time difference** between two events are set in **y - value**

