



Alpha spectrometry for nuclear forensics

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IAEA Regional Training Course on
Practical Introduction to Nuclear
Forensics

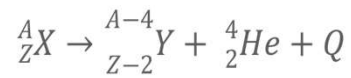
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Content

- Alpha decay
- Alpha emitting radionuclides
- Alpha particle interaction with matter
- Alpha detector system
- $^{234}\text{U}/^{230}\text{Th}$ isotope pair chronometry
- Radiochemical separation
- Characteristics of alpha spectra

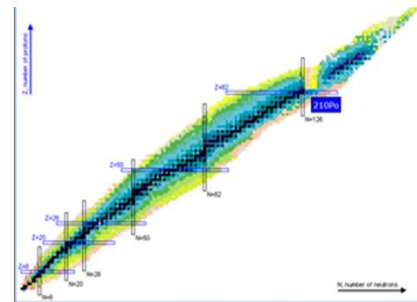


Alpha decay



${}^4_2\text{He}^{2+}$ Helium nucleus

Q = quanta of energy ~2-10 MeV



<https://www.nndc.bnl.gov/nudat2/>

Alpha emitting radionuclides

- Mono-energetic α emitters
 - Singlet α peak e.g. ${}^{210}\text{Po}$
- Dual-energetic α emitters
 - Doublet α peak e.g. ${}^{232,234,238}\text{U}$ and ${}^{228,230,232}\text{Th}$
- Complex α emitters
 - Multiplet α peak e.g. ${}^{229}\text{Th}$, ${}^{235}\text{U}$,

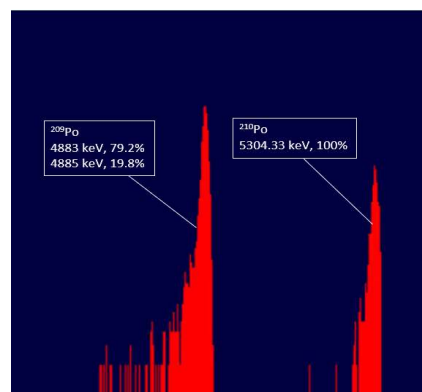
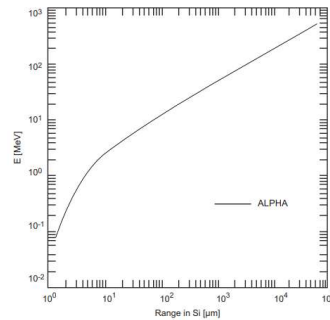


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Interaction with matter

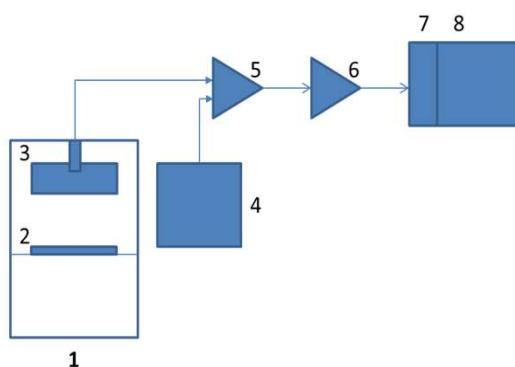
- α -particles:
 - Carry sufficient energy to ionise or excite atoms
 - Deposit their energy across a short distance
 - Typically keep their direction
 - Travel only a few cms in air
 - Travel $\sim 100 \mu\text{m}$ in Si



Range of alpha particles in Si

Image: Handbook of Radioactivity Analysis

Alpha detection system



#	Description
1	Detector chamber under vacuum
2	Source and chamber shelf
3	Detector
4	High Voltage
5	Pre-amplifier
6	Amplifier
7	Analogue to Digital Converter (ADC)
8	Multi-Channel Analyzer (MCA)

Image Source: ANSTO-image

Alpha detection system

- Considerations:
 - Size of alpha source relative to detector
 - Distance of alpha source from detector
 - Counting chamber under vacuum
 - Purity and thickness of alpha source



Image Source: ANSTO-image

$^{234}\text{U}/^{230}\text{Th}$ isotope pair chronometry

Material	Chronometer
Uranium	$^{234}\text{U} \rightarrow ^{230}\text{Th} (\rightarrow ^{226}\text{Ra}, \rightarrow ^{214}\text{Bi})$ $^{235}\text{U} \rightarrow ^{231}\text{Pa} (\rightarrow ^{227}\text{Ac})$ $^{233}\text{U} \rightarrow ^{229}\text{Th}$ $^{232}\text{U} \rightarrow ^{228}\text{Th}$
Plutonium	$^{241}\text{Pu} \rightarrow ^{241}\text{Am}$ $^{240}\text{Pu} \rightarrow ^{236}\text{U}$ $^{239}\text{Pu} \rightarrow ^{235}\text{U}$ $^{238}\text{Pu} \rightarrow ^{234}\text{U}$ $(^{242}\text{Pu} \rightarrow ^{238}\text{U})$
Th material, UOC, some bulk U	$^{232}\text{Th} \rightarrow ^{228}\text{Ra} \rightarrow ^{228}\text{Th}$

Using multiple chronometers increases confidence in the determined age value

$^{234}\text{U}/^{230}\text{Th}$ isotope pair chronometry

separation of parent and progeny isotopes (“clock” set to zero)



build up of progeny isotopes with time



determine ratio of progeny/parent isotopes (“clock”)



calculate age

$^{234}\text{U}/^{230}\text{Th}$ isotope pair chronometry

- Age = the time elapsed since the last separation of parent and progeny isotopes (e.g. production, reprocessing, purification)
- Calculation based on decay equations (Bateman equations):

$$T = \frac{1}{\lambda_{234\text{U}} - \lambda_{230\text{Th}}} \ln\left(1 - \frac{N_{230\text{Th}}}{N_{234\text{U}}} * \frac{\lambda_{230\text{Th}} - \lambda_{234\text{U}}}{\lambda_{234\text{U}}}\right)$$

- ‘Model ages’ based on assumptions:
 - Complete separation of progeny from parent during sample formation
 - Closed-system behaviour of the sample after formation

$^{234}\text{U}/^{230}\text{Th}$ isotope pair chronometry

■ Considerations

- Main radionuclides present (e.g. uranium, plutonium)
- Approximate uranium amount, activity or enrichment
- Physical and chemical form
- Closed system (no fractionation between the parent and progeny)
- Impurities, homogeneity of sample
- An initial estimate of the radioactive components is essential
- Screening by non-destructive techniques (NDA) (e.g. gamma spectrometry) useful

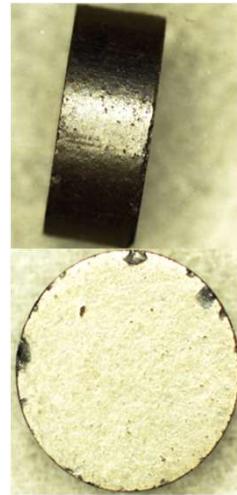


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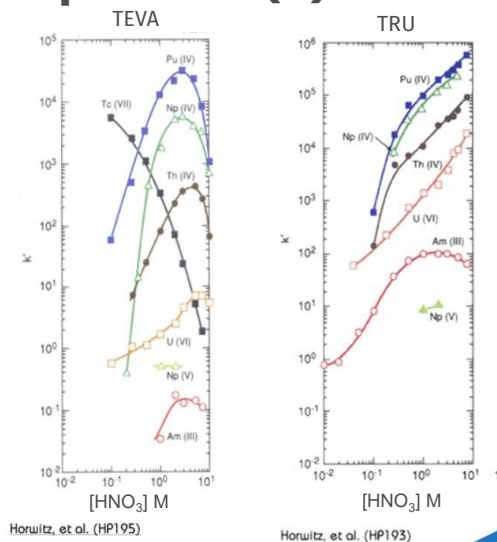
Radiochemical separation

- Destructive technique (e.g. pellet dissolution)
- Long half lives of parent U isotopes - very small amounts of progeny isotope grow-in:
 - need radiochemical separation with high chemical recovery of progeny nuclide (to purify and concentrate the progeny)
- Consider detection limits, laboratory activity handling limits
- Radiotracer required to track chemical losses
- Spiked and unspiked analysis pairs required

Radiochemical separation (U)



Image Source: ANSTO



Radiochemical separation (Th)

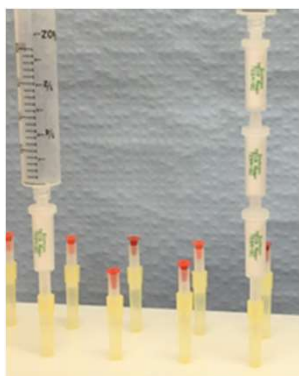
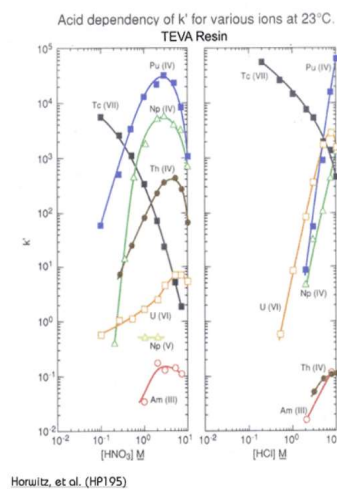


Image Source: ANSTO



Radiochemical separation (α source prep)

Step 1

- Add cerium carrier
- Add 1 mL **hydrofluoric acid***
- Mix and place in refrigerator for 30 minutes

Step 2

- Assemble filter components: vacuum box, yellow and white manifold tips, filter base, 0.1 μ m filter paper, filter funnel.
- Turn on vacuum
- Condition filter with ethanol and water and ensure no leaking is occurring

Step 3

- Remove solution from step 1 from refrigerator (after 30 mins)
- Pour solution into filter funnel

Step 4

- Once solution from step 3 has passed through the filter, rinse solution vessel with 5 mL of water and pour into filter funnel
- Rinse filter funnel with a further 5 mL of water
- Rinse filter funnel with 5 mL of ethanol

Step 5

- Once solution has passed through filter paper turn off vacuum
- Remove filter paper from base with tweezers and centre on a 1" stainless steel disc that has a thin layer of glue
- Air dry or dry under IR lamp and label

Characteristics of alpha spectra (U)

Uranium

Energy (KeV), Intensity (%)

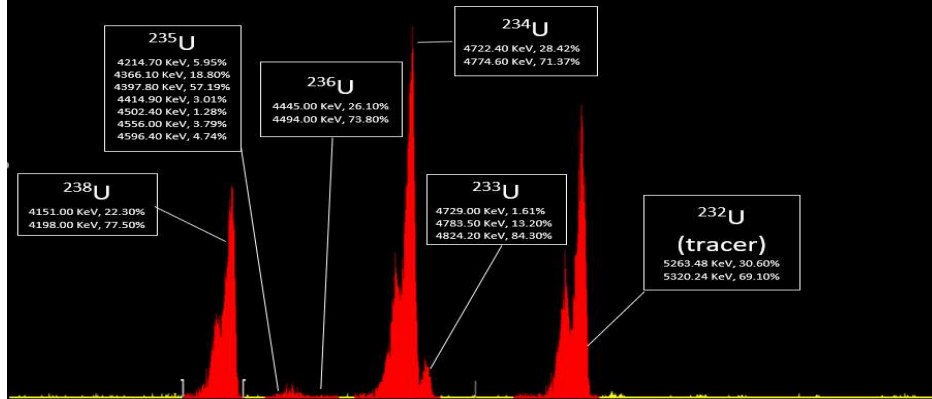


Image Source: ANSTO

- Peaks in spectra are expected and at correct energy
- Low background
- Low energy tail
- Peak resolution and separation

Poor example of alpha spectrum (U)

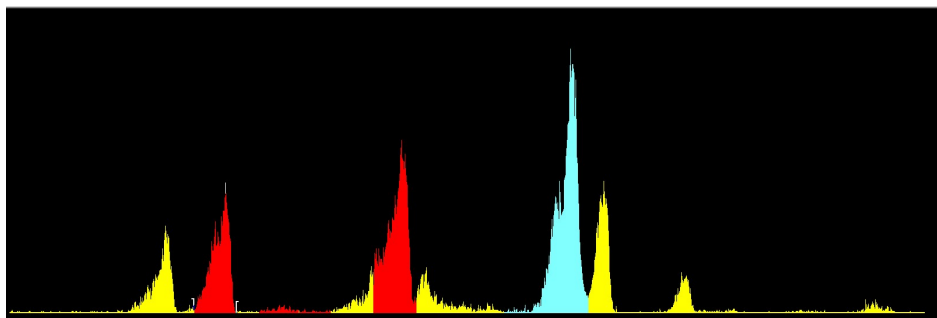


Image Source: ANSTO

Th peaks interfering with U peaks



Characteristics of alpha spectra (Th)

Thorium

Energy (KeV), Intensity (%)

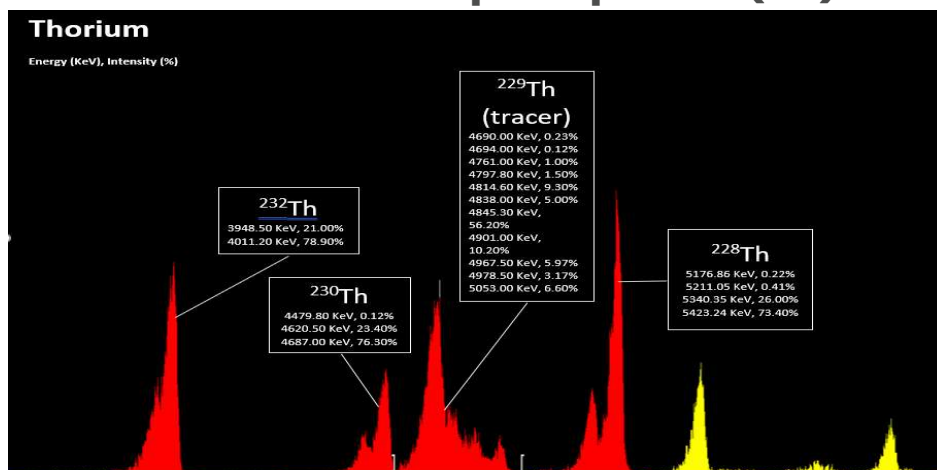


Image Source: ANSTO

- Low energy tail
- Peak resolution and separation
- Peaks in spectra are expected and at correct energy (incl progeny)
- Low background esp. between ^{229}Th and ^{230}Th



Poor example of alpha spectrum (Th)

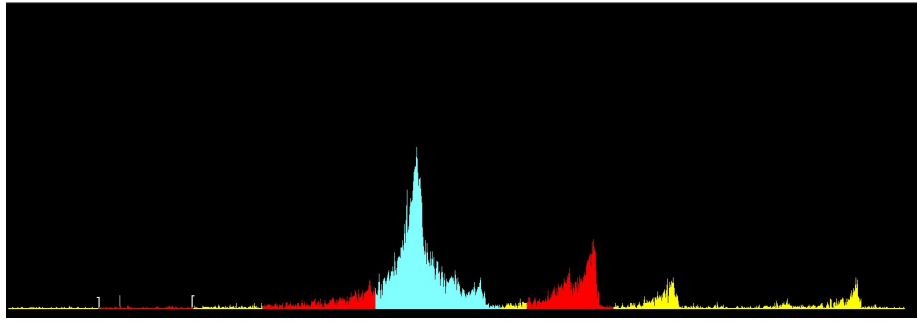


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Noisy baseline leads to poor resolution