Detection of $^{225/226}$Ra Impurities in ^{225}Ac Isotope Products using Extraction Chromatography and Microprecipitation Techniques

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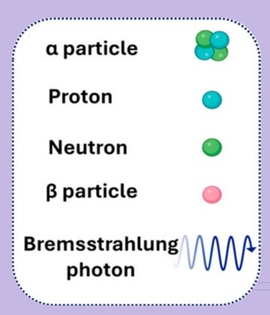
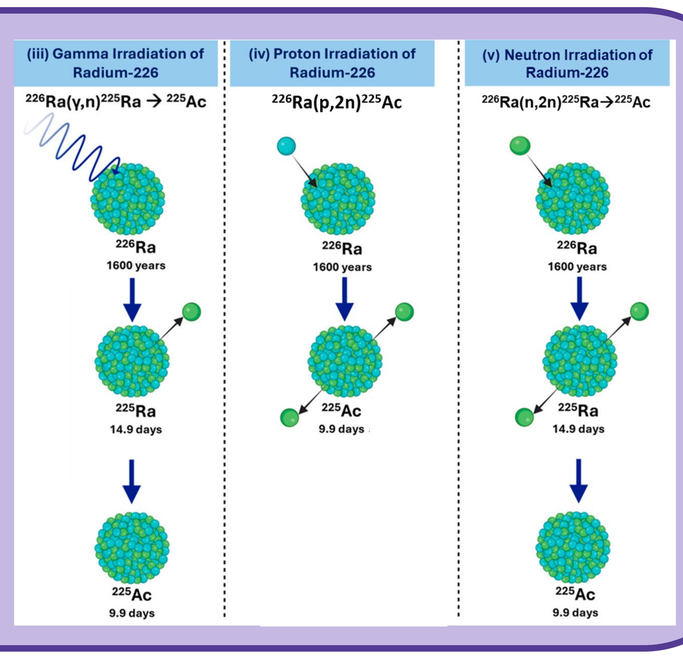
Abstract:

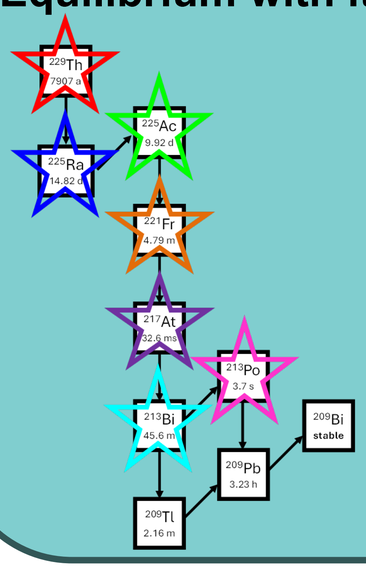
Production of ^{225}Ac may be achieved by irradiation of ^{226}Ra targets to produce intermediate ^{225}Ra via (n,2n) or (γ,n) reactions, leading to high specific activity ^{225}Ac free of $^{226/227}$Ac impurities. Using these production routes, the main impurities of potential concern for the ^{225}Ac isotope products will be long-lived ^{226}Ra target material ($t_{1/2}$ = 1603 a) and co-produced intermediate ^{225}Ra ($t_{1/2}$ = 14.9 d) parent. The work described herein employs extraction chromatography (EXC) to selectively purify radium from the ^{225}Ac and daughters for quantification by radiometric detection. Alpha spectrometry and High Purity Germanium (HPGe) gamma spectroscopy allow for the best limits of detection of ^{226}Ra and ^{225}Ra respectively, and a Ba tracer may be added for recovery correction. This method allows for the quantification of radium impurities in ^{225}Ac samples of >99.99% radionuclidic purity.

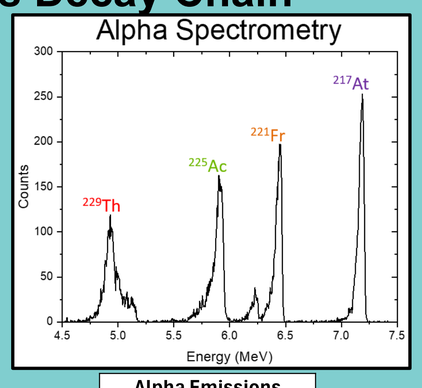
Production of ^{225}Ac from ^{226}Ra Targets

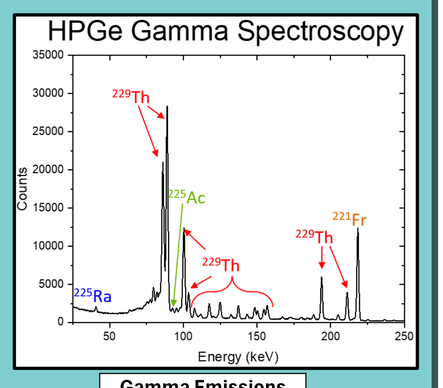




Alpha and Gamma Emissions of ^{225}Ac(^{229}Th) in Equilibrium with its Decay Chain





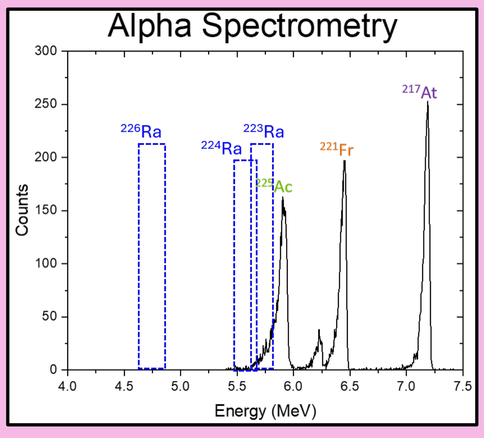


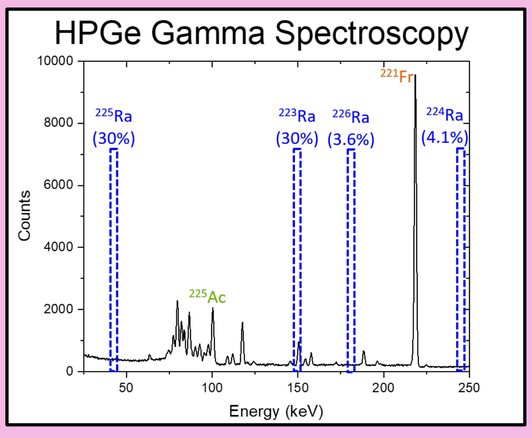
Alpha Emissions		
Isotope	Energy	Intensity
^{229}Th	5.168	100%
^{225}Ac	5.935	100%
^{221}Fr	6.457	100%
^{217}At	7.201	99.99%
^{213}Bi	5.988	2.14%
^{213}Po	8.536	100%

Gamma Emissions		
Isotope	Energy	Intensity
^{229}Th	88.47	23.90%
^{225}Ra	40.09	30.00%
^{225}Ac	99.8	1.00%
^{221}Fr	218.1	15.61%
^{213}Bi	440.4	30.77%

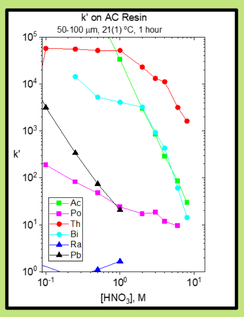
Alpha and Gamma Emissions of Ra Impurities

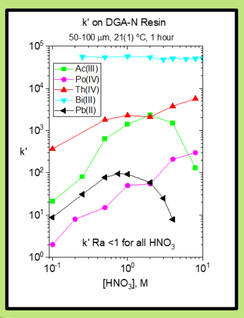
(iii) Gamma Irradiation of Radium-226	(iv) Proton Irradiation of Radium-226	(v) Neutron Irradiation of Radium-226
^{226}Ra(γ,n)^{225}Ra → ^{225}Ac	^{226}Ra(p,2n)^{225}Ra → ^{225}Ac	^{226}Ra(n,2n)^{225}Ra→^{225}Ac
225/226Ra	223/225/226Ra	223/224/225/226Ra





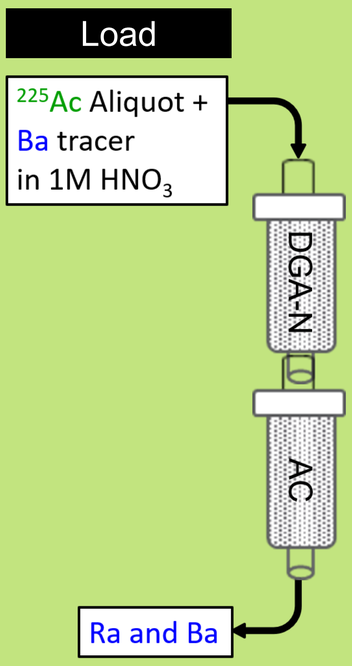
Isolation of Ra Impurities from ^{225}Ac





Load Solution:
- Small Aliquot of ^{225}Ac product (approx. 10 µCi)
- Stable or ^{133}Ba yield tracer
- 10 mL 1 M HNO_3
- Mix to homogenize

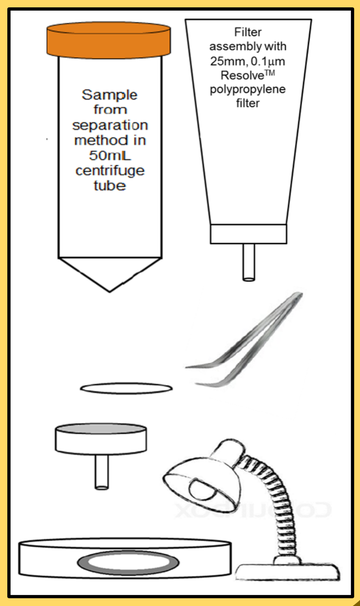
Cartridge Elution:
- Actinide (AC) resin on top of DGA-Normal (DNS) resin
- Pre-condition with 10 mL 1 M HNO_3 and discard as waste
- Load sample and collect effluent for Ra analysis
- Rinse with 10 mL1 M HNO_3 and collect with load effluent for Ra analysis



Alpha Spectrometry Source Preparation

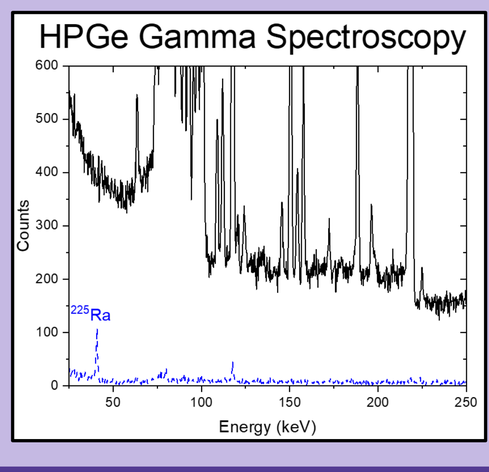
Barium Sulfate Precipitation:
- 50 µg Ba carrier
- 3 g $(NH_4)_2SO_4$
- 5 mL isopropanol
- Place in ice bath

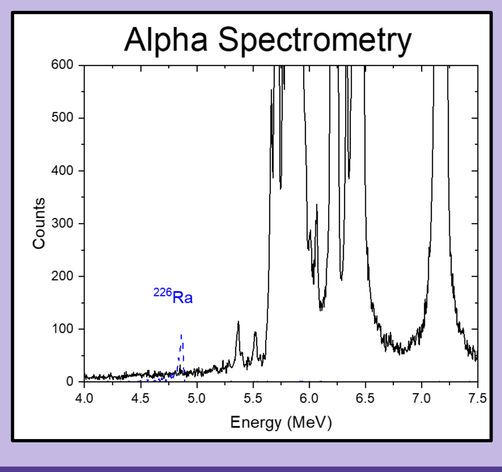
Source Preparation:
- Wait 20-30 min
- Filter over Eichrom's Resolve® Filter
- Mount on adhesive planchet
- Dry under heat lamp
- Analyze using a calibrated alpha spectrometer



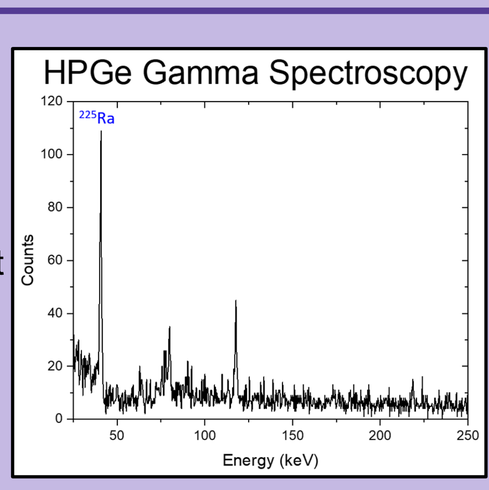
Results

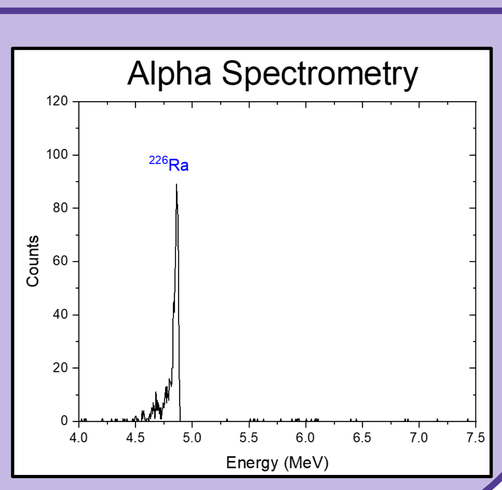
^{225}Ac Aliquot *without* separation





Radium impurities in ^{225}Ac Aliquot *after* separation





Future Work

1. Identify the optimal yield tracer
   - ^{133}Ba increases Compton, reducing HPGe LOD
   - Stable Ba could be precipitated for gravimetric yield determination
2. Improve $BaSO_4$ precipitation conditions
   - Remove the need for ice bath while maintaining high Ba/Ra recovery
3. Partner with industry leaders to test real, production-scale samples
   - Eichrom only has 10 µCi ^{225}Ac
   - Need higher activity source materials to test real, accelerator-produced samples for Ra impurities

References

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4. McAlister, Daniel; Horwitz, E. Philip, *Radiochimica Acta,* 99**(3)**, 151, (2011).
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