Selective Separation of Radium and Actinium from Bulk Thorium Target Material

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U beam-stop from ZG-Synchrotron (ANL), 12 MeV

2.5 x 10^{24} atoms U in target

Protons delivered ???

Atoms Produced

Mass Number

Flowsheet $^{233}\text{U}$, $^{229}\text{Th}$, $^{225}\text{Ac}$

Aged $^{233}\text{U}$ with $^{229}\text{Th}$ in HNO$_3$

Anion Exchange

Dilute HNO$_3$

$^{233}\text{U}$

$^{229}\text{Th}$
Flowsheet $^{233}\text{U}$, $^{229}\text{Th}$, $^{225}\text{Ac}$

Aged $^{233}\text{U}$ with $^{229}\text{Th}$ in HNO$_3$

Dilute H$_2$SO$_4$

Anion Exchange

$^{233}\text{U}$ $\rightarrow$ $^{229}\text{Th}$

30-50 g Th Spallation target

Cation Exchange

$^{229}\text{Th}$ $\rightarrow$ $^{225}\text{Ac}/^{225}\text{Ra}$

5M HNO$_3$

75 mL Column 300 mL 5M HNO$_3$
Dissolution of Th in H$_2$SO$_4$/HF

H$_2$SO$_4$
HF
H$_2$O

Heat
H$_2$O
Low Solubility??!!
Solubility curves of the hydrates of thorium sulphate.
Dissolution of Th in $\text{H}_2\text{SO}_4$/HF

- $\text{H}_2\text{SO}_4$ 0.6 M
- HF 0.03 M
- Th 0.1 M
- pH 0.8 – 1.0

Heat

H$_2$O

Low Solubility??!!

Cool. Mix.
- 30-50 grams of Th as Th(SO$_4$)$_2$, Th(SO$_4$)$_3^{2-}$, Th(SO$_4$)$_4^{4-}$ rejected

- Ra$^{2+}$ and Ac$^{3+}$ uptake decreases above 0.4-0.5M sulfate at pH = 2.0

- >99% recovery Ra
- 93-98% recovery Ac

Precipitation $\text{Ba}^{(225}\text{Ra})\text{SO}_4$

Dissolve $\text{BaSO}_4$ in $\text{NaOH/EDTA}$

Adjust to 1-2M $\text{HNO}_3$

Separate Ac on DGA (>95% Ac recovery)

$\text{>95% Ra on 1-2 mg/L Ba(SO}_4)$

Only 10-15% $^{225}\text{Ac}$ released.
- UTEVA (Phosphonate) removes remaining Th, Pa, U

- DGA retains Ac$^{3+}$, rare earths, Ca

- Ra$^{2+}$ passes both columns (Fe, Ba, Al, many fission products)

- UTEVA and DGA can have small traces of Ca impurity (precondition separately)

- DGA can be used to separate Ca and rare earths from Ac fraction.

$^{225}\text{Ac}$ Purity (Direct Actinium Product)

FWHM
40-60 keV

Ac-227  Ac-225  Fr-221  At-217
$^{225}$Ac Purity (Actinium from $^{225}$Ra)

FWHM
40-60 keV
FWHM
200-340 keV

$^{225}\text{Ac}$ Purity

$^{227}\text{Ac}$ $^{225}\text{Ac}$ $^{221}\text{Fr}$ $^{217}\text{At}$

Counts

Energy (keV)
$^{225}\text{Ac}$ Purity (Rare Earth Removal)

FWHM
200-340 keV

$^{225}\text{Ac}$

$^{221}\text{Fr}$

$^{217}\text{At}$

$^{227}\text{Ac}$
$^{225}\text{Ac Purity (Ca Removal)}$

FWHM $\frac{60-80}{\text{keV}}$

$^{227}\text{Ac}$

$^{225}\text{Ac}$

$^{221}\text{Fr}$

$^{217}\text{At}$
Ac Separation from Calcium

$k'$ on DGA, Normal Resin vs HNO$_3$

$k'$ on DGA, Normal Resin vs HCl
$^{225}$Ac Elution (breakthrough in calcium removal)

50-100 $\mu$m DGA, Normal Resin

Column
4.2 cm length
0.7 cm diameter

Cartridge
2.7 cm length
0.9 cm diameter

4M HNO$_3$
9M HCl
0.1M HCl

90% Ac
95% Ac

5 mL column
5 mL cartridge
$^{225}$Ac Elution (breakthrough in calcium removal)

50-100 $\mu$m DGA, Normal Resin

Column
4.2 cm length
0.7 cm diameter

Cartridge
2.7 cm length
0.9 cm diameter

- 2 mL column
- 2 mL cartridge
- 8M HNO$_3$

90% Ac
95% Ac
>99% Ac
0.1M HCl
**$^{225}$Ac Separation (8M HNO$_3$)**

- **50-100 µm DGA, Normal Resin**
- **Column**
  - 4.2 cm length
  - 0.7 cm diameter
- **Complete removal of Fe, Ba(Ra), Pb, Sr, Bi**
- **>95% removal of Ca**
- **La co-elutes with Ac**

![Graph showing separation of various elements using different nitric acid concentrations and HCl.]
$^{225}$Ac Separation (9M HCl)

50-100 µm DGA, Normal Resin

Column
4.2 cm length
0.7 cm diameter

- Complete removal of Fe, Ba(Ra), Pb, Sr
- 99.5% removal of Ca
- 99.0% removal of Bi
- La co-elutes with Ac
Ac Polishing Scheme

#1 UTEVA + DGA (preconditioned separately)
   Load sample in 4M HNO3 (Fe, Al, Th, Pa, U, TRU removal)
   Rinse 4M HNO3 (Remove UTEVA)
   Rinse DGA only with 8M HNO3 (Ca, Sr, Ba, Pb)
   Recover Ac in 10M HNO3 (REE stay on DGA column)
   Dilute Ac to <5M HNO3

#2 UTEVA + DGA (preconditioned separately)
   Load sample/Rinse/Remove UTEVA (Concentrate Ac, additional purification)
   Rinse DGA only with 1M HNO3 (Reduce HNO3)
   Rinse 9M HCl (3-4 bed volumes, convert to HCl)
   Recover Ac in 0.1-2M HCl (3-4 bed volumes)
Future Work

Optimization of target dissolution for massive thorium

Test Optimized elution conditions on irradiated target

Behavior of spallation byproducts in cation exchange/sulfate system

$^{213}\text{Bi}$ from $^{225}\text{Ac}(^{227}\text{Ac})$