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Title:	241Am (n,gamma) isomer ratio measurement
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Los Alamos
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Form 836 (7/06)

^{241}Am (n,γ) Isomer Ratio Measurement

Evelyn M. Bond, David J. Vieira, W. Allen Moody, Alice K. Slemmons

The objective of this project is to improve the accuracy of the ^{242}Cm/^{241}Am radiochemistry ratio. We have performed an activation experiment to measure the ^{241}Am(n,γ) cross section leading to either the ground state of ^{242g}Am ($t_{1/2}$=16 hr) which decays to ^{242}Cm ($t_{1/2}$=163 d) or the long-lived isomer ^{242m}Am ($t_{1/2}$=141 yr). This experiment will develop a new set of americium cross section evaluations that can be used with a measured ^{242}Cm/^{241}Am radiochemical measurement for nuclear forensic purposes. This measurement is necessary to interpret the ^{242}Cm/^{241}Am ratio because a good measurement of this neutron capture isomer ratio for ^{241}Am does not exist.

The targets were prepared in 2007 from ^{241}Am purified from LANL stocks. Gold was added to the purified ^{241}Am as an internal neutron fluence monitor. These targets were placed into a holder, packaged, and shipped to Forschungszentrum Karlsruhe, where they were irradiated at their Van de Graff facility in February 2008. One target was irradiated with ~25 keV quasi-monoenergetic neutrons produced by the ^{7}Li(p,n) reaction for 3 days and a second target was also irradiated for 3 days with ~500 keV neutrons.

Because it will be necessary to separate the ^{242}Cm from the ^{241}Am in order to measure the amount of ^{242}Cm by alpha spectrometry, research into methods for americium/curium separations were conducted concurrently. We found that anion exchange chromatography in methanol/nitric acid solutions produced good separations that could be completed in one day resulting in a sample with no residue.

The samples were returned from Germany in July 2009 and were counted by gamma spectrometry. Chemical separations have commenced on the blank sample. Each sample will be spiked with ^{244}Cm, dissolved and digested in nitric acid solutions. One third of each sample will be processed at a time. First, the gold will be removed by anion exchange chromatography. Then the ^{242}Cm will be separated from the ^{241}Am using the methanol/nitric acid anion exchange method. When a sufficient separation has been achieved, a deposit will be prepared and the ^{242}Cm will be counted by alpha spectrometry. The purified ^{241}Am fraction containing the long lived ^{242m}Am will be allowed to decay into ^{242}Cm for a period of ~6 months. After this time, the americium/curium separations will be repeated and the ^{242}Cm that has grown in will be counted by alpha spectrometry. At the conclusion of the experiment, we will have cross section measurements for ^{241}Am (n,γ) ^{242g}Am and ^{241}Am (n,γ) ^{242m}Am at two energies.

^{241}Am (n,γ) Isomer Ratio Measurement

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David J. Vieira, Todd Bredeweg, Walter Allen Moody, Alice Slemmons

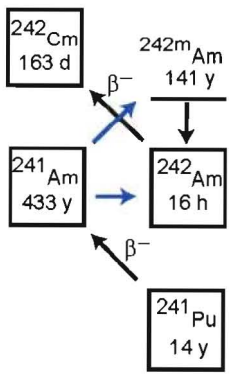
FY11 Program Review

NA-22 Nuclear Forensics R&D Program

Jan 11, 2011

Objective

- The objective of this project is to improve the accuracy of the ^{242}Cm/^{241}Am radiochemistry ratio.



^{241}Am

- Present in Pu due to the β-decay of ^{241}Pu ($t_{1/2}$=14.4 yr)
- Can be used as an internal tracer
- Neutron-induced reaction products can provide valuable nuclear forensics information.

Office of Nonproliferation & Verification R&D (NA-22) Unclassified

Objective

$^{242}Cm/^{241}Am$

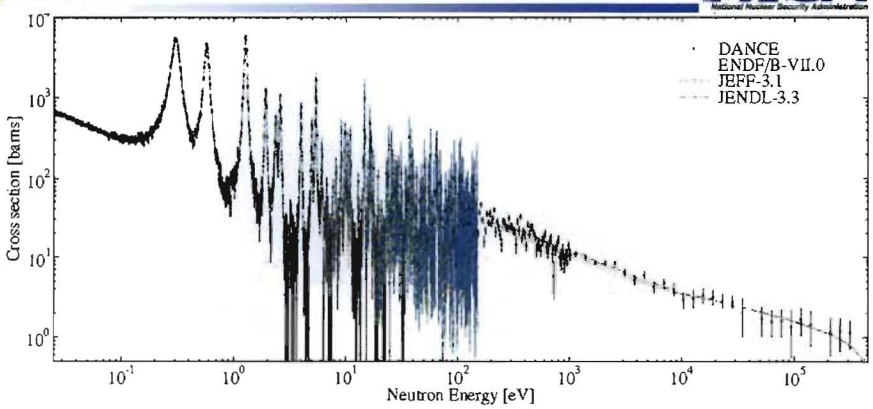
- Produced by (n,γ) neutron capture reaction

$$^{242m}Am \xrightarrow[141\ years]{IT} {}^{242g}Am \xrightarrow[16\ hours]{\beta^-} {}^{242}Cm \xrightarrow[163\ days]{\alpha}$$

- Provides information on low-energy neutrons (down-scattered and thermalized fission neutrons)

In order to correctly interpret $^{242}Cm/^{241}Am$, we need to measure the (n, γ) reaction leading to ^{242}Am and ^{242m}Am

Objective



The $^{241}Am(n\gamma)$ cross section has been measured at DANCE up to 300 keV. But, this information is for total cross section, i.e $^{242g}Am + {}^{242m}Am$.

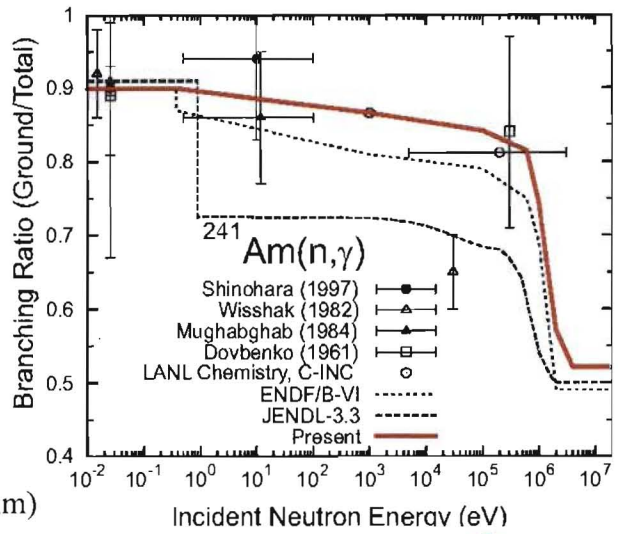
Jandel, M. et al. Phys. Rev. C, 78 (2008) 034609.

Objective

Current isomer ratio data for $^{241}Am(n,\gamma)$ have large error bars

More accurate isomer ratio data are required

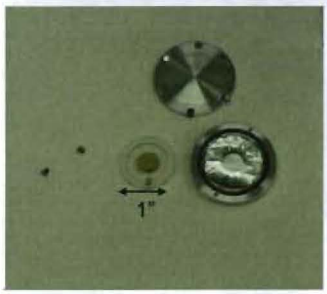
Branching ratio = $^{242g}Am/(^{242g}Am + ^{242m}Am)$



Outline of Experimental Method

1. Prepare ^{241}Am targets
2. Ship targets to Karlsruhe, Germany
3. Irradiate targets at Karlsruhe
4. Develop Am/Cm separation chemistry
5. Ship targets back to LANL
6. ^{242g}Am determination
   a) Am/Cm separation
   b) Plate and count ^{242}Cm
7. ^{242m}Am determination
   a) Am/Cm separation
   b) Plate and count ^{242}Cm
8. Summary

1. Prepare ^{241}Am targets



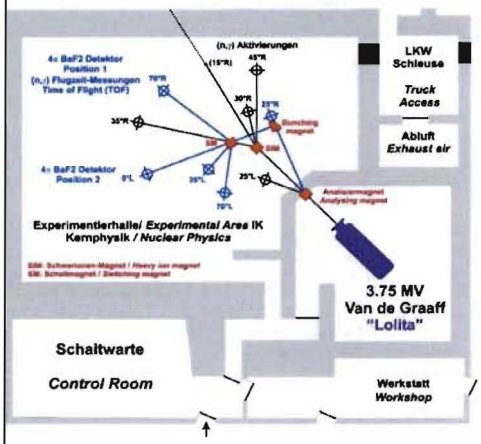
- Purified 100 mg of ^{241}Am
- Encapsulated three 15 mg ^{241}Am targets in Ti.
- Gold was added to each target to monitor neutron fluence.
- Blank targets also prepared
- Glovebox and open front hood work performed at CMR at LANL
- Work performed in FY2007

2. Ship targets to Karlsruhe

- The samples were shipped to Karlsruhe, Germany in February 2008
- Shipment required ~ 2 months to obtain required permissions and packaging
- We were aided by specially trained shippers at TA-55 at LANL

Croft 2799E Package

3. Irradiate targets at Karlsruhe



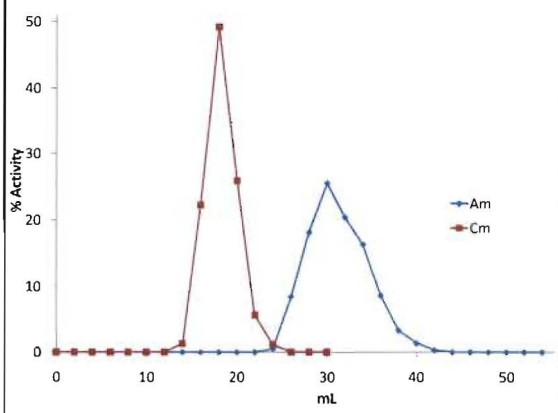
- Quasi-mono-energetic neutrons at 25 keV and 500 keV
- Produced by ^{7}Li(p,n) reaction
- 3 days for each target
- Flux of 10^{9} to 10^{10} n/cm^{2}-s
- ^{198}Au activation measured with Ge detector at Karlsruhe to determine integrated neutron flux.
- Lead attenuator used to reduce strong 59.5 keV γ-ray emanating from ^{241}Am target.

This was the last experiment at "Lolita", it is now shut down.

Karlsruhe staff were great hosts: Franz Käppeler, Stephan Walter, Iris Dillman

Thank you!

4. Develop Am/Cm separation chemistry

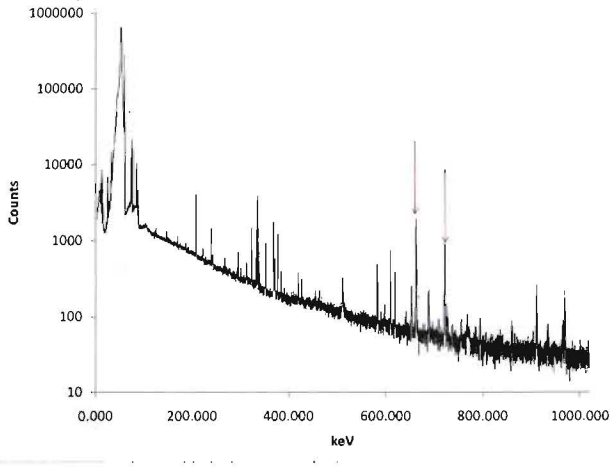


- Separations chemistry developed concurrently with target prep, shipment, and irradiation
- Developed anion exchange method using 60% methanol /nitric acid mixtures
- Several separations may be needed to obtained needed separation to obtain ^{242}Cm alpha spectra

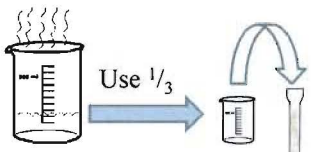
5. Ship targets back to LANL

The three targets were shipped from Germany to CMR in July 2009

Each target was assayed by gamma spectroscopy by C-AAC personnel (Donivan Porterfield, Patrick Martinez)



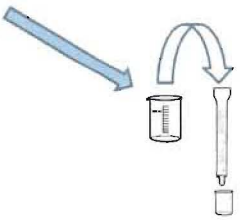
^{242g}Am determination



Use $^1/_3$

1. Dissolve targets, add ^{244}Cm tracer

2. Separate Au from sample with anion exchange chromatography



3. Separate Cm from Am using anion exchange from methanol/nitric acid

Repeat separation as needed for clean sample

^{242g}Am determination

Electroplate and count samples by alpha spectroscopy. **This will tell us how much ^{242g}Am was produced by the irradiation.**

Branching ratio = ^{242g}Am/(^{242g}Am + ^{242m}Am)

^{242g}Am determination

Chemistry Status (FY 2009)

Started chemical separations with blank target to determine ^{242}Cm background (if any)

Chemical dissolution

- Epoxy dissolved with acetone
- Deposit dissolved with nitric acid
- Sample spiked with ^{244}Cm

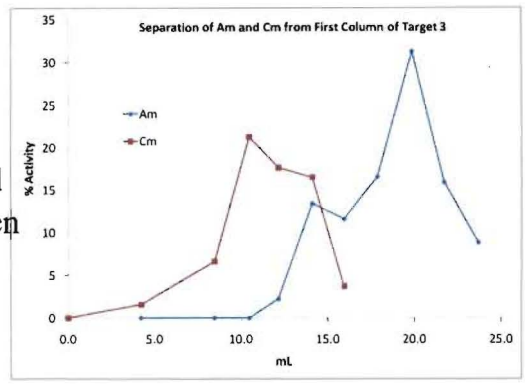
Gold contamination removed with anion exchange chromatography.

^{242g}Am determination

Chemistry Status (FY 2009)

First Am/Cm separation

- Fall 2009
- Elution was slower than anticipated
  - The methanol evaporated
  - Column dried out between samples
  - Am/Cm separation
- Second Am/Cm separation started FY 2010
- Much faster
- Results pending



^{242m}Am determination

- The ^{242}Cm must be removed from the samples with a decontamination factor of 10^{14}
  - ❖ this will require 4-7 separations!
- Wait 6-9 months to allow ^{242}Cm to grow in
- Am/Cm separations performed as before
- Electrodeposition
- Measurement of ^{242}Cm by alpha spectrometry

This second measurement will tell us how much ^{242m}Am was produced by the irradiation.
We will then be able to calculate the branching ratio

$$\text{Branching ratio} = {}^{242g}\text{Am}/({}^{242g}\text{Am} + {}^{242m}\text{Am})$$

Future Research

Complete Target 3 Separations
January –February 2010

Complete Target 1 &2 Separations
By Fall 2010

Ingrowth of ^{242}Cm
Fall 2010 to Spring 2011

Complete Second series of Am/Cm separations
Fall 2011

Calculate Branching Ratio
Fall 2011

Summary

- ^{241}Am Targets have been prepared, irradiated, and returned
- Separation of targets for ^{242g}Am determination is proceeding
- Am/Cm has been developed and can be used for routine separations at LANL
- Technician training
  - Allen Moody hired and trained
  - Helped develop chemistry
- Results will be published in open literature

Summary

Talks

"Americium Curium Separations for Nuclear Chemistry Experiments" A. K. Slemmons E. M. Bond, W. A. Moody, D. J. Vieira, J. R. FitzPatrick, R. Sudowe, 235th ACS National Meeting & Exposition, April 6-10, 2008 (Oral Presentation)

"Progress in Americium and Curium Separations for the ^{241}Am(n, gamma) Measurement", E. M. Bond, W. A. Moody, A. K. Slemmons, D. J. Vieira, ILWOG 42, May 24-27, 2010. (Oral Presentation)

"The Separation of Americium and Curium for Nuclear Chemistry Experiments", E. M. Bond, W. A. Moody, D. J. Vieira, A. K. Slemmons, 239th ACS National Meeting and Exposition, March 21-25, 2010. (Invited Oral Presentation)

"Nuclear Chemistry Experiments with Americium and Curium", E. M. Bond, D. J. Vieira, A. K. Slemmons, W. A. Moody, F. Käppeler, S. Walter, I. Dillman, ILWOG 41, September 8-12, 2008. (Oral Presentation).

Collaborators

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