The Measurement of Cs Isotopes by Accelerator Mass Spectrometry

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Caesium Radioisotopes

- Anthropogenic
- Product of nuclear fission
- Age dating from nuclear weapons testing
- Identifying reactor products

Thermal Neutron Fission of U-235



Why AMS?

- Cs 137 has a relatively short half life (30a)
- Most has decayed since the late 50s
- Cs 135 much longer (about 2 Ma)
- The Ratios of Cs 135/137 quite useful

135Cs Half Life

- N. Sugarman (1949)
 - Xe-135 $\xrightarrow{\beta}$ Cs-135 + e⁻ + \overline{V}_e
 - 1.85 → 2.3 Ma
- Oak Ridge (1949)
 - 2.95 ± 0.3 Ma

Proposed Experiment

- Use <u>liquid scintillation</u> <u>counting</u> and <u>accelerator</u> <u>mass spectrometry</u>
- LSC: Find A_o
- AMS: find N_o





Challenges

- Purity of Cs135 and yield tracers
- Optimization of beam current
- Isobaric interferences
- Development of AMS Procedure
- Cs memory

Isotope Production

- Require sources of Cs with minimal amounts of isotope contamination
- Xenon Decay
- Neutron Capture



Creation of Cs135: ¹³⁵Xe Beta Decay

Bq = Becquerel = Decay/Second

- 1 Bq of ¹³⁵Cs requires the decay of 2 GBq of ¹³⁵Xe
- Chalk River Labs supplied a section of pipe that had Xe running through it.



LSC

- $t_{1/2} = \frac{\ln 2}{A_o} N_o$
- Mix aliquot of solution with scintillating agent and count the beta's
- Small Cs¹³⁷ peak.
- Can't use 137 for tracer
- Use of surrogate beta emitter for efficiency



Neutron Capture

$$N_{A+1} = N_A \sigma_A \phi t$$

$$N_{A+2} = \frac{1}{2} N_A \sigma_A \sigma_{A+1} \phi^2 t^2$$

$$N_{A+3} = \frac{1}{6} N_A \sigma_A \sigma_{A+1} \sigma_{A+2} \phi^3 t^3$$

 $\bullet = \sigma$ $\Sigma \bullet = N$

Creation of Cs134: Cs133 (n, γ) Cs134





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Cs Current Optimization: The Study of Ion "Sourcery"

- Choosing anion
 - CsF_2^-
- Homogenous mixing
 - Chemical vs Mechanical





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Isobaric Interferences

- Barium
 - Masses 134→138
- Zn Dimers
 - Masses 128→140

Isobar Separator for Anions



Isobar Separator for Anions





Zn Dimer Interference

- Noticed counts at mass 136 and 138
- No counts at 137 so not Barium
- Measure mass 136 and increase stripper pressure





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Development of AMS Procedure

- Isotope scanning
 - Monitor isotopes during analysis
- Yield Tracer
 - Ensure no fractionation

Masses Monitored During Analysis

- Require # of Cs134 atoms as yield tracer (134 amu)
- Require # of Cs135 atoms as analyte (135 amu)
- Ensure no Barium contamination (136 amu)
- Check mass 137 for Barium/Caesium (137 amu)

Barium Natural Abundance





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RadioCs Measurement



Cs Memory



Predicted vs Observed



| | Measured Half Life (Ma) | Uncertainty (%) |
|---------------------|-------------------------|-----------------|
| Sugarmann (1949) | 1.85 | _ |
| | 2.3 | _ |
| ORNL (1949) | 2.95 | 10% |
| C. MacDonald (2014) | 0.69 | 42% |
| | 0.63 | 45% |

In Summation



- Eliminated isobaric interferences
- Demonstrated the effectiveness of ISA-AMS routine
- Successfully measured rare Cs isotopes by AMS

Acknowledgments

Department of Physics, University of Ottawa

- C.R.J. Charles
- X.L. Zhao
- L. Kieser

Department of Earth Science, University of Ottawa

• J. Cornett

IsoTrace Laboratory, University of Toronto

• A.E. Litherland

Isobarex