Accelerator Mass Spectrometry for long-lived radionuclides

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AMS in Zürich

1960s, 6 MV
AMS since 1979

15 m

$^{10}\text{Be}$, $^{36}\text{Cl}$
AMS in Zürich

1960s, 6 MV
AMS since 1979

15 m

$^{10}\text{Be}$, $^{36}\text{Cl}$

1990s, 600 kV

2.5 m

$^{10}\text{Be}$, $^{129}\text{I}$, $^{239}/^{240}\text{Pu}$
AMS in Zürich

10Be, 36Cl

1960s, 6 MV
AMS since 1979

10Be, 129I, 239/240Pu

1990s, 600 kV

14C

2000s, 200 kV

15 m

2.5 m

1 m
Content

Introduction to Accelerator Mass Spectrometry (AMS)

Detection limits

Applications of AMS for radioecology and nuclear waste management

Summary
What is AMS?

The most sensitive isotope selective detection method

- Single ion detection capabilities
- Huge dynamic range
- Extreme sensitivity
- High accuracy

Sample:
long-lived radioisotopes ($^{10}$Be, $^{14}$C, $^{36}$Cl, $^{129}$I, $^{239/240}$Pu,...)

Isotope ratios:
$10^{-8}$ - $10^{-15}$ (for example $^{36}$Cl/Cl)

Typical size:
1 mg ≈ 1 million atoms of long-lived radionuclide
Long-lived Radionuclides

- **$^{14}$C**: 5730 yrs
  - Archaeology, Art History, Oceanography, Earth & Environmental Sciences

- **$^{10}$Be**: 1.5 Myrs
  - Oceanography, Geology, Geophysics, Earth & Environmental Sciences...

- **$^{26}$Al**: 716 krys
  - Geology, Geophysics, Life sciences, Biomedical Applications...

- **$^{36}$Cl**: 301 krys
  - Polar Ice Cores, Geology, Solar Activity, Hydrology

- **$^{41}$Ca**: 103 krys
  - Life Sciences, Biomedical Applications

- **$^{129}$I**: 16 Myrs
  - Earth & Environmental Sciences, Radioecology, Nuclear safeguards...

- Actinides (Pu, U, Pa)
  - Radioecology, Nuclear Safeguards, Oceanography
Limits of Mass Spectrometry

Measurement of radioisotopes with long half-life

Problems:
- Molecular interferences
- Isobaric interferences

Solution:
- Stripping process (Breaking up bindings)
- Negative ion formation
- Particle identification

AMS
Overview

2. filter mass analysis (low-energy end)

3. filter molecule destruction

4. filter mass analysis (high-energy end)

5. filter particle identification dE/dx

1. filter negative ion formation

stripping process charge exchange

mass spectrometer

electrostatic deflector

$^{13}\text{C} / ^{12}\text{C}$

$^{14}\text{C} / ^{12}\text{C}$

$^{12}\text{C}$

$^{13}\text{C}$
Ion source

- Cs-sputtering
- Solid samples
- High yield neg. ions

Samples at the large machine
Sample wheel at Tandy
Ø 2.5 • 0.5 mm
Ion Beam Physics

ETH

Stripper

negative ions

incoming beam

(molecular break-up caused by ion gas collisions)

Argon gas

positive ions

outgoing beam

- no molecular background

\[
\begin{align*}
\text{incoming beam:} & \quad 238\text{U}^{16}\text{OH}^- \\
& \quad 238\text{U}^{17}\text{O}^- \\
& \quad 239\text{Pu}^{16}\text{O}^- \\
\text{molecular break-up:} & \quad \text{ion gas collisions} \\
\text{outgoing beam:} & \quad 239\text{Pu}^{n+} \\
& \quad 238\text{U}^{n+} \\
& \quad \text{H}^+ \\
& \quad \text{O}^{n+}
\end{align*}
\]
Detector

Ionization
Charge collection
Charge sensitive Pre-Amplifier
Energy loss measurements

Si$_3$N$_3$: revolution in low energy particle detection
Isobar Separation in Detector

\[ \frac{dE}{dx} \text{(a.U.)} \]

- boron
- beryllium

\[ \Delta E \text{ (30 mm)} \]

Path (a.U.)
$^{10}\text{Be}$ Spectrum

\[ \Delta E \]

\[ E_{\text{res}} \]
Detection limits

- $^{36}\text{Cl} \sim 3 \times 10^5$ atoms (0.02 fg)
- $^{129}\text{I} \sim 10^6$ atoms (0.2 fg)
- $^{239}\text{Pu} \sim 2 \times 10^5$ atoms (0.1 fg)
Why AMS?

- no need to wait for decay
- no molecular background
- effective suppression of isobars for $Z < 20$
- huge dynamic range (up to $10^6$)
- small sample sizes
Applications of AMS for radioecology and nuclear waste management
129I from Sellafield and La Hague

- >95% from Sellafield and La Hague
- Point-like location, well-known release function
- Conservative tracer
Disequilibrium in $^{129}$I system

+ Speciation!
+ Biophilic!
$^{129}\text{I}$ in oceanography & ecology

- Water mass labeling (all European outflows!)
- Transport times, ventilation rates
- Benchmark for global models

Alfimov et al., 2004
$^{129}\text{I}$ as a benchmark for ocean circulation models

Orre et al, 2009
$^{129}$I as a benchmark for ocean circulation models

Orre et al, 2009
$^{129}\text{I}$ for retrospective dosimetry

Map of Chernobyl release

$^{137}\text{Cs}$ deposition density

- $185-555 \text{ kBq m}^{-2}$
- $>555 \text{ kBq m}^{-2}$

Michel et al, 2005
$^{129}$I for retrospective dosimetry

Map of Chernobyl release

Michel et al, 2005
\textbf{\textsuperscript{36}Cl in U-rich environments}

- Subsurface n-flux
- Tracing U & Th-rich deposits

\textsuperscript{36}Cl/C\textsubscript{0} vs. U-equivalent [ppm]

\begin{align*}
y &= 3.9143x + 1.1 \\
R^2 &= 0.9999
\end{align*}

\textbf{Balderer et al, 2009}
$^{36}\text{Cl}$ and $^{63}\text{Ni}$ in Hiroshima: recovery of fast and thermal neutron fluxes

Calculation

Measured $^{63}\text{Ni}$

Straume et al, 2003
$^{36}\text{Cl}$ and $^{63}\text{Ni}$ in Hiroshima: recovery of fast and thermal neutron fluxes

Nagashima et al, 2004
Dating groundwater with meteoritic $^{36}\text{Cl}$
$^{36}\text{Cl}$ in Dye-3 and Berkner Island

Year / AD

$^{36}\text{Cl} / g$
„Bomb-pulse“ $^{36}$Cl for nuclear waste storage facilities

Recent measurements of $^{36}$Cl in Yucca Mountain rock, soil and seepage

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Samples of rock, soil and seepage were collected from Yucca Mountain, USA, and analyzed for $^{36}$Cl/Cl ratios by accelerator mass spectrometry (AMS). Rock excavated from the Drill Hole Wash fault at repository horizon depths produced a ratio suggesting that small amounts of water with “bomb-pulse” $^{36}$Cl had percolated to that site over the past 50 years. Rock from four other sites within the exploratory studies facility did not yield bomb-pulse ratios. Ratios in the soil varied depending on depth and location, with some samples producing bomb-pulse signatures. Ratios for seep water were slightly elevated above the present cosmogenic background value. This paper also discusses results from a column study mimicking the passage of $^{36}$Cl through volcanic rock and from an experiment using bromide instead of chloride as a carrier in sample preparation for AMS.

Rock... at repository horizon depths produced a ratio suggesting that small amount of water with „bomb-pulse“ $^{36}$Cl had percolated to that site over the past 50 years.
„Bomb-pulse“ $^{36}\text{Cl}$ for underground water flows

Infiltration of the gravel aquifer from the Danube river, western Hungary

Balderer et al, 2004
Collaborations with
- ZSR Hannover
- Universidad de Sevilla

240Pu/239Pu for source determination

- Gas Cooled
- Boiling Water
- Pressurised Water
- Advanced Gas Cooled

Power reactors

Average weapon test

Weapon grade

Weapon grade (pre 1960)

Taylor et al. 2001
Accident with nuclear weapons (1966)

Collaboration with University of Seville
$^{240}\text{Pu}/^{239}\text{Pu}$ in Soil samples

Chamizo et al., 2006

Collaboration with Universidad de Sevilla
$^{240}$Pu/$^{239}$Pu in the North Sea

- 0.18 - 0.19
- 0.22 - 0.25
- 0.31 - 0.37
North Sea 1995

Tanja Bisinger PhD-Thesis, 2009

Wasserproben

Isotopenverhältnis $\frac{^{240}\text{Pu}}{^{239}\text{Pu}}$
Summary

- AMS is perfect for long-lived radionuclides \(^{10}\text{Be}, \, ^{14}\text{C}, \, ^{26}\text{Al}, \, ^{36}\text{Cl}, \, ^{129}\text{I}, \, ^{240}/^{239}\text{Pu}\)
- \(^{36}\text{Cl}\) is useful to study hydrology near nuclear storage facilities, to investigate U-rich rocks.
- \(^{129}\text{I}\) traces the marine and airborne releases of European wastes and calibrates the global circulation models
- \(^{240}/^{239}\text{Pu}\) shows sources of plutonium
Co-workers, who contributed to this talk:

- Marcus Christl, Lukas Wacker, Hans-Arno Synal

Thank you!