Development of Analytical Techniques for Safeguards Environmental Samples at JAEA as IAEA Network Analytical Laboratory

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Introduction of safeguards
environmental sample analysis (ESA)
  Objective of ESA in safeguards
  Technique of ESA
    Bulk analysis
    Particle analysis (SIMS method)
    Particle analysis (FT-TIMS method)

Milestone

Introduction of CLEAR facility
Introduction

- Suspicion for North Korea and Iraq in early 1990s
- 93+2 Programme to strengthen the international safeguards system
  - Introduction of environmental sample analysis
    - Undeclared nuclear activities
    - Undeclared nuclear materials
Environmental sample

Cotton cloth (10x10 cm²)

Cellulose (2.5 cm φ)

Wipe wall and floor in a nuclear facility

For example, looking for:
- High enrichment uranium in enrichment plant
- Weapon grade plutonium in R&D facility

Isotope ratio of U & Pu
CLEAR
- Clean room lab.
- Bulk analysis
- Particle analysis
- Non-radioactive sample

NUCEF
- Radiochemical lab.
- Bulk analysis
- Radioactive sample
Environmental sample analysis

Bulk Analysis
- Ashing
- Acidic Digestion
- Chemical Separation

ICP-MS
To remove matrix elements for accurate isotope ratio measurement

Particle Analysis
- SIMS
- FT-TIMS

Cotton cloth (10x10 cm²)

Around 1 μm particles

DL for Pu: ~10 fg (10⁻¹⁵ g)

ISO Class 5 Clean room
Chemical separation

- 15M HNO₃ & Dry up (3 times)
- 9.5M HCl & Dry up
- 1M HCl + 3M NH₂OH•HCl
- Standing for 60 min.
- Dry up slowly
- 9M HCl + 0.2M HNO₃

Steps:
1. 9M HCl + 0.1M HNO₃
2. 9.5M HCl
3. 9M HCl + 0.1M HI
4. 0.5M HCl

Anion exchange:
MCI GEL, CA08P(75~150μm) 0.84ml

Clean exhaust hood
Isotope ratio measurement

Double focus ICP-MS: ELEMENT1

Ion detector: SEM  Resolution (M/ΔM) = 300
Scan mode: E-Scan

0.32M HNO₃ Solution
1,800 cps / (pg/ml)
NU: 5 ~ 800 pg/ml (ppt)
Bulk analysis (Radioactive sample)

Ashing & Acid Digestion

Redox

1. 7M HNO₃
2. 0.7M HNO₃ + 90% CH₃OH
3. 9M HCl + 0.1M HNO₃

Anion exchange
MCI GEL, CA08Y (25μmφ), 50ml

1. 2. Alkaline metal etc
2. U
3. Waste
4. Pu

TIMS
Particle analysis

SIMS method (Routinely used)

Particle recovery (Vacuum impactor)

Particles collected on planchet

SIMS

$^{238}\text{U}^+$

$^{16}\text{O}^+$

2 μm
Milestones

- **2002 November Review at CLEAR**
  - Two IAEA experts visited JAEA
  - Analytical results
  - Facility performance (CLEAR/NUCEF)
  - QA/QC

- **2003 January** Qualification to IAEA NWAL

- **2003 February** First analysis of domestic samples

- **2004 January** NWAL contract with IAEA

- **2004 February** First analysis of IAEA samples

Sample analysis

- Bulk: 28 Swipes/y (Average 2005-2006)
- Particle: 33 Swipes/y (Average 2004-2006)
Desolavation module

Scott-type spray chamber
Sensitivity: 1.8 Mcps / ppb $^{238}$U

Bulk analysis

Over 7 times improvement !!

Apex
High-efficiency inlet system
Sensitivity: 13 Mcps / ppb $^{238}$U

Pick-up SIMS method

Particles pick up

Particle recovery (Vacuum impactor)

Particles pick up and transfer

Scanning electron microscopy (SEM)

SIMS
Results of pick-up SIMS

F. Esaka et al., Talanta 71 (2007) 1011.
FT-TIMS method

Collect particles on filter

Prepare irradiation sample

Neutron irradiation

Etching

Accurate method
A lot of time to measure
XRF for screening

X-ray tube
Detector
Swipe sample

Uranium amount
Impurity measurement
Pb, Fe etc.
Results of XRF measurement

X-ray spectrum

Semi-quantitative measurement

Elemental distribution

Uranium

5 x 7 mm

Intensity (x100 cps)

Energy (keV)

U-Lα  U-Lβ  U-Lγ

Results of XRF measurement

U, Pb, Fe etc.
Cleanness class is defined as maximum number of particles (> 0.5 μm) in 1 ft³.

No cleanness-controlled rooms, e.g. normal office rooms, sometimes contain the particle more than one million.
Air flow and pressure control

Pressure:
- +30 Pa
- +20 Pa
- +10 Pa
- -10 to 0 Pa

Air flow:
- Supply
- Exhaust
- Circulation
- Transfer

Service area

Supply plenum chamber

FFU

Raise floor

Return plenum chamber

Return duct

Class 100

Class 1,000

Fan-filter unit (including HEPA filter)

Clean hood