

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



**Research Group for Analytical Science
Nuclear Science and Energy Directorate
Japan Atomic Energy Agency**

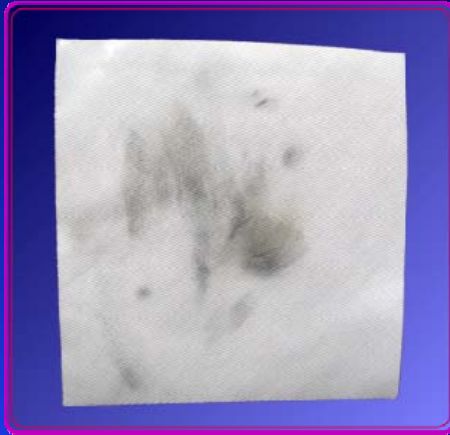
Contents

- ◆ 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

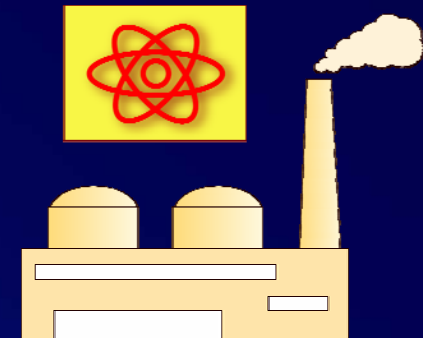
Environmental sample



Cotton cloth
(10x10 cm²)



Cellulose
(2.5cm 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

Laboratory

◆ CLEAR

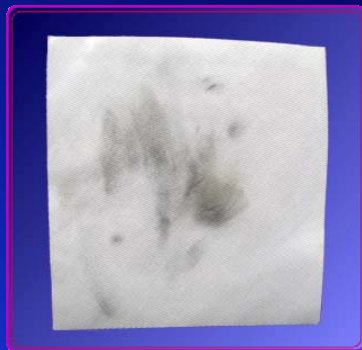
- ◆ Clean room lab.
- ◆ Bulk analysis
- ◆ Particle analysis
- ◆ Non-radioactive sample



◆ NUCEF

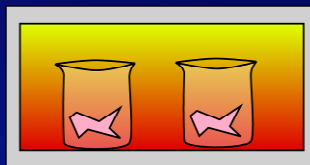
- ◆ Radiochemical lab.
- ◆ Bulk analysis
- ◆ Radioactive sample

Environmental sample analysis



Cotton cloth
(10x10 cm²)

Bulk Analysis

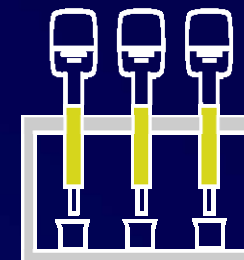


Ashing



Acidic Digestion

DL for Pu : ~10 fg (10⁻¹⁵ g)



Chemical Separation

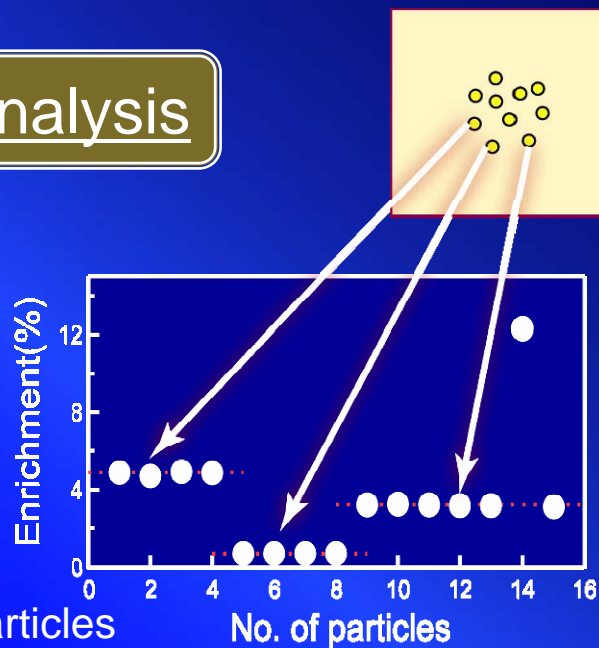
ICP-MS

To remove matrix elements for accurate isotope ratio measurement

Particle Analysis

SIMS

FT-TIMS



Around 1 μm particles



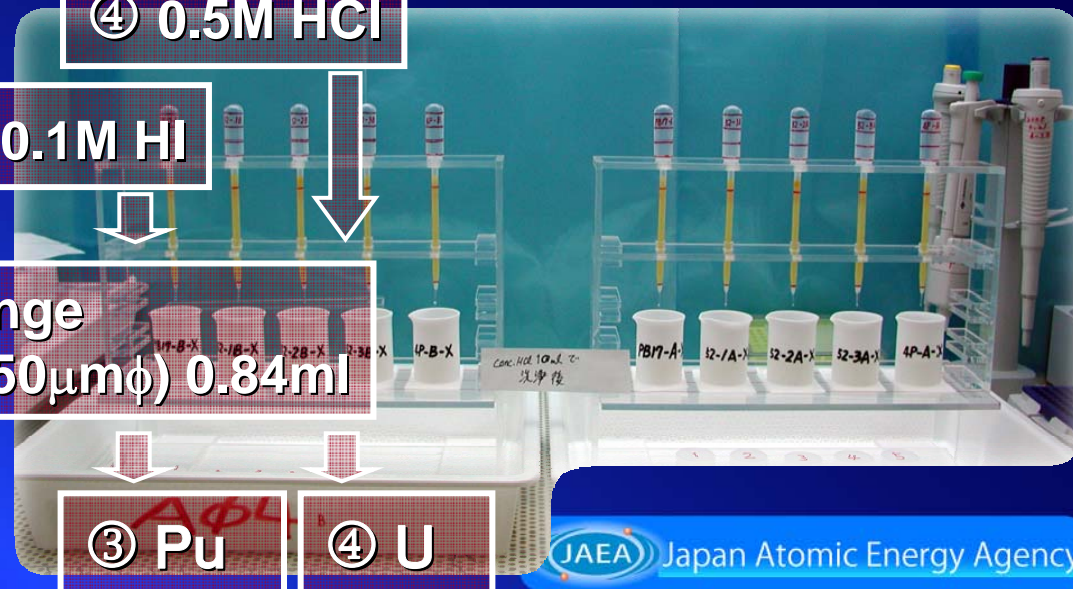
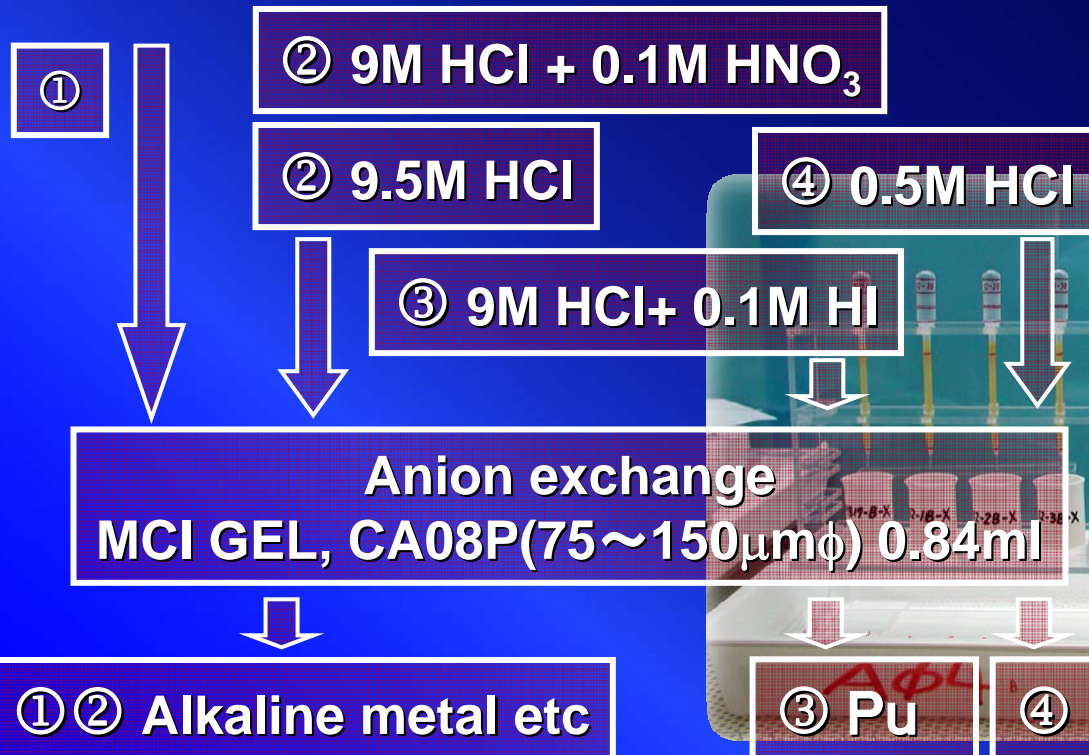
ISO Class 5 Clean room

Chemical separation

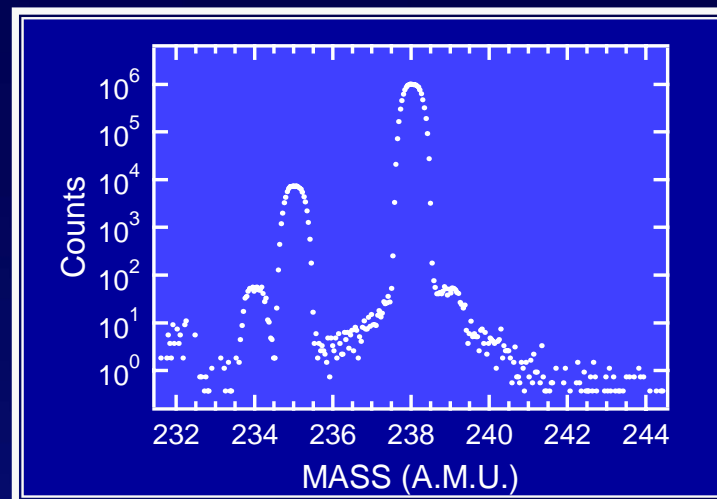
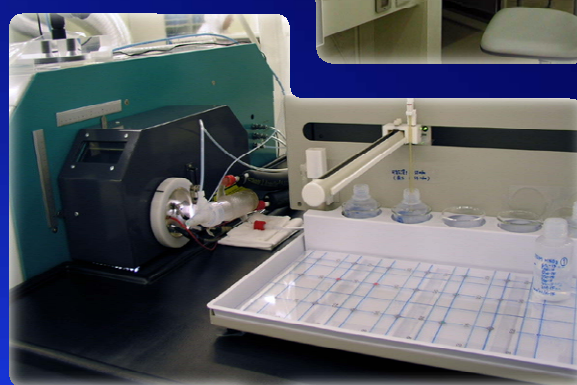
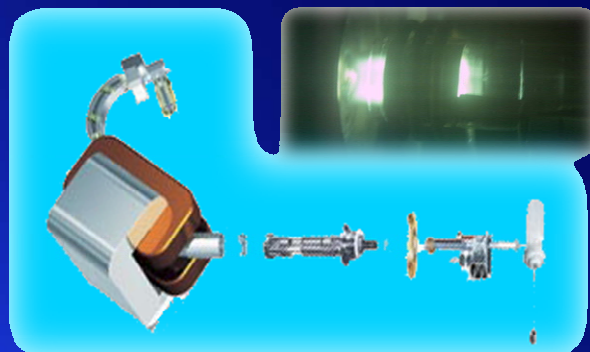
- 15M HNO₃ & Dry up (3 times)
- 9.5M HCl & Dry up
- 1M HCl + 3M NH₂OH·HCl
- Standing for 60 min.
- Dry up slowly
- 9M HCl + 0.2M HNO₃



Clean exhaust hood



Isotope ratio measurement



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

Double focus ICP-MS : ELEMENT1

Ion detector: SEM Resolution (M/ΔM)= 300

Scan mode: E-Scan

Bulk analysis (Radioactive sample)

Ashing & Acid Digestion

Redox

② 9M HCl + 0.1M HNO₃

② 0.7M HNO₃ + 90%CH₃OH

② 7M HNO₃

③ 7M HNO₃

⑤ 0.5M HCl

Anion exchange

MCI GEL, CA08Y (25μmφ), 50ml

①② Alkaline metal etc

③ U

④ Waste

⑤ Pu

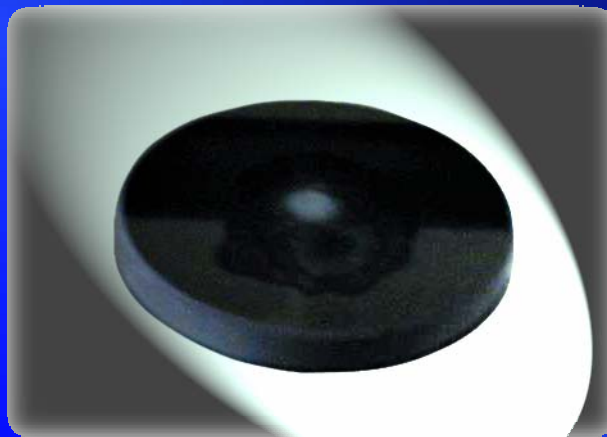
TIMS

Particle analysis

◀ SIMS method (Routinely used)



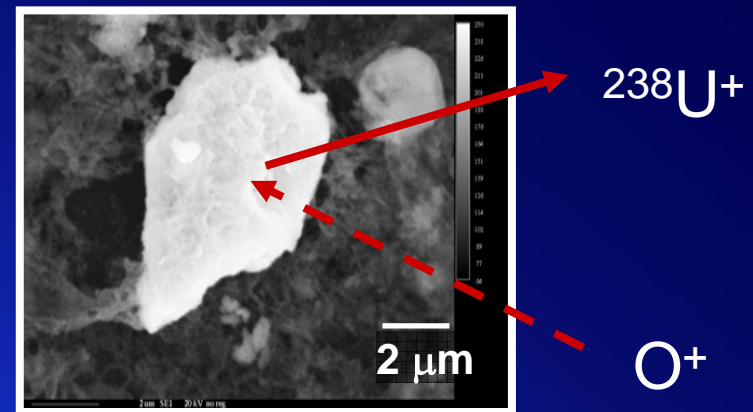
Particle recovery (Vacuum impactor)



Particles collected on planchet



SIMS



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

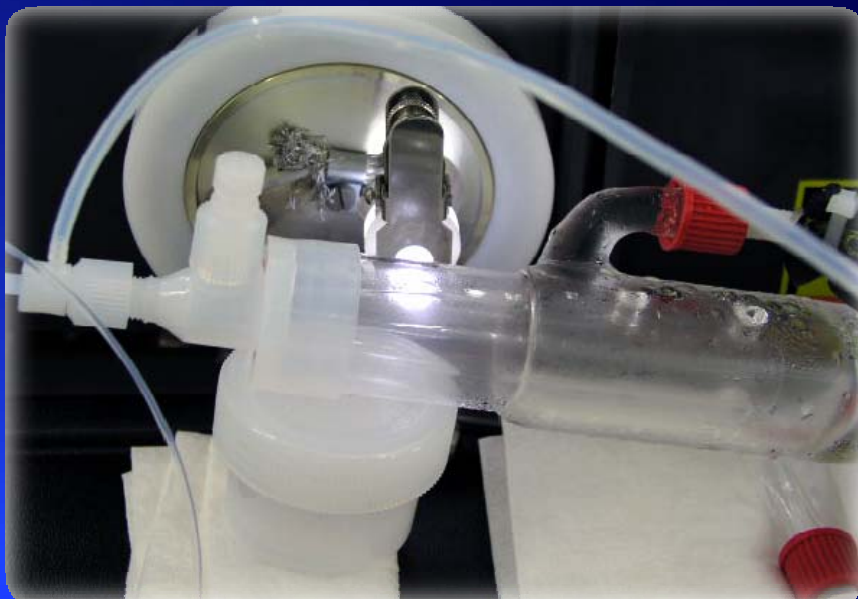
◆ 2004 February First analysis of IAEA samples

Sample analysis

Bulk: 28 Swipes/y (Average 2005-2006)

Particle: 33 Swipes/y (Average 2004-2006)

Desolvation 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



<http://www.elementalscientific.com/products/apex.asp>

Pick-up SIMS method

◀ Particles pick up



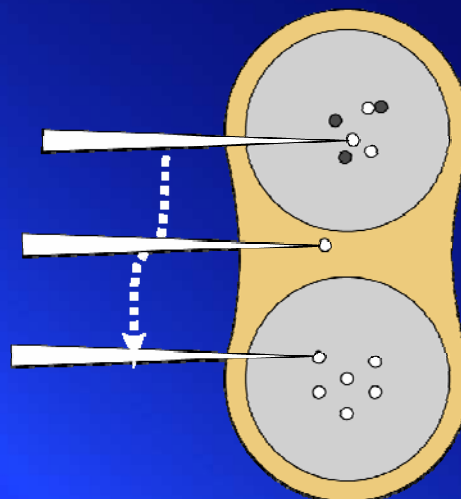
Particle recovery (Vacuum impactor)



Scanning electron microscopy (SEM)

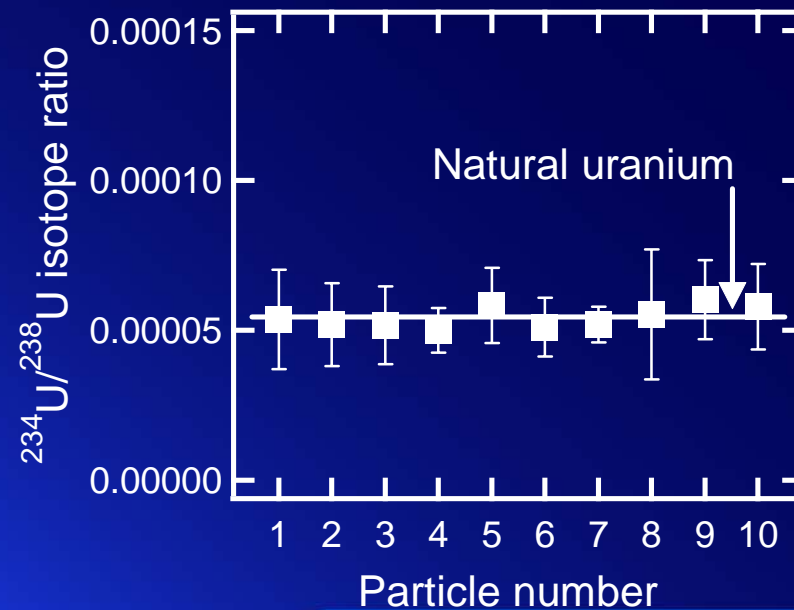
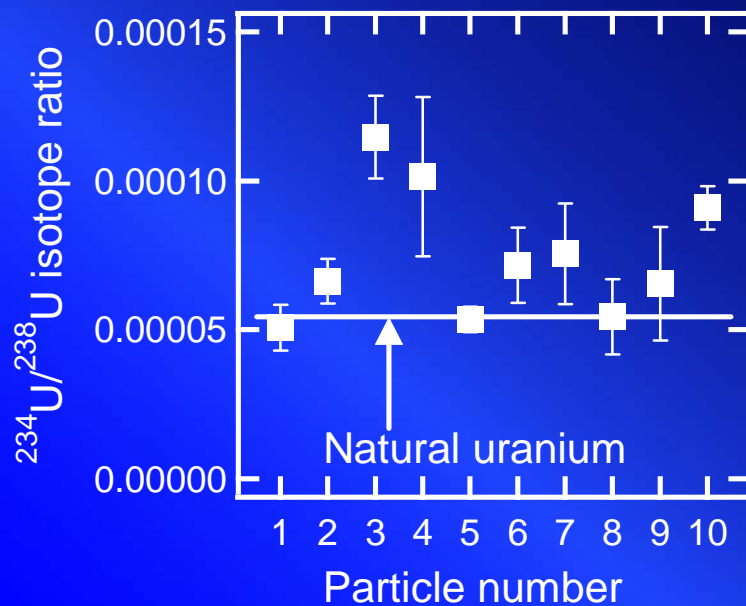
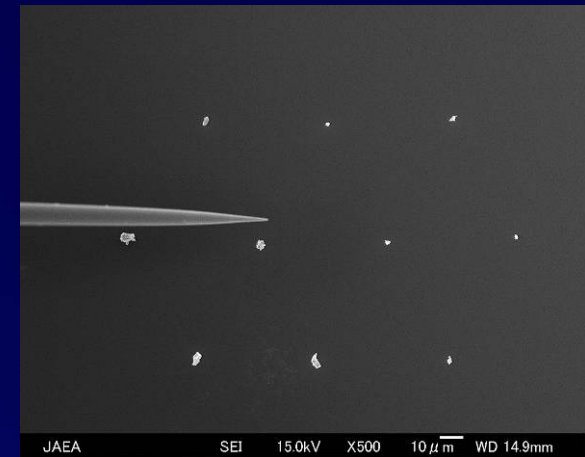
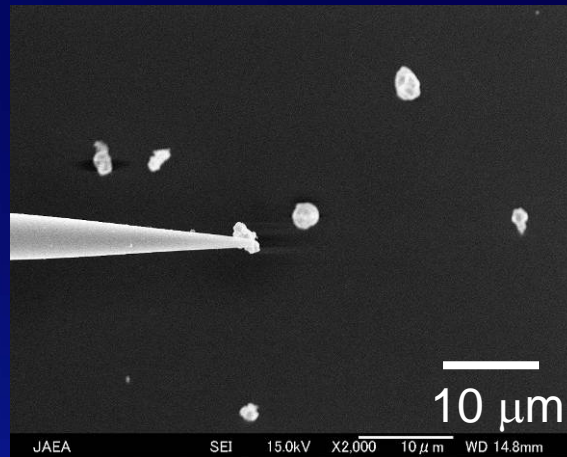
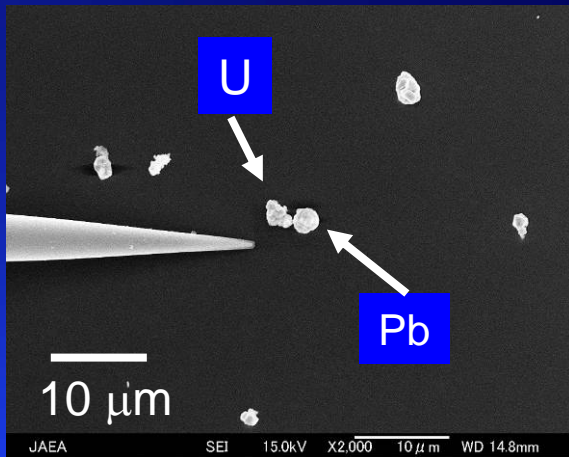


Particles pick up and transfer



SIMS

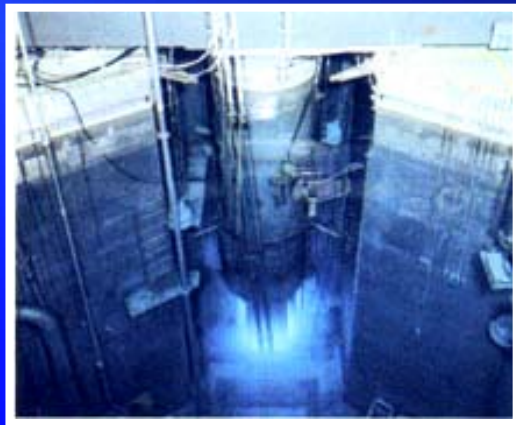
Results of pick-up SIMS



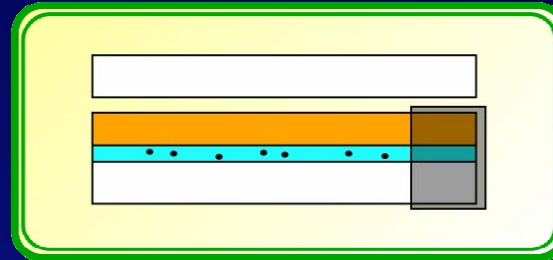
FT-TIMS method



Collect particles on filter



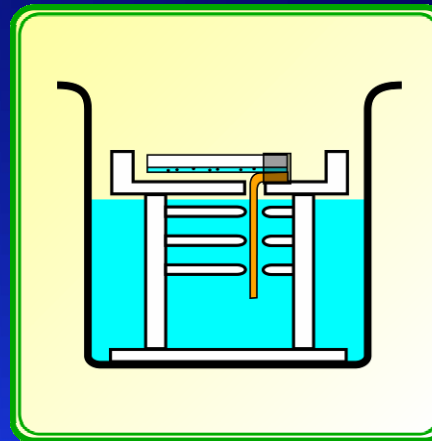
Neutron irradiation



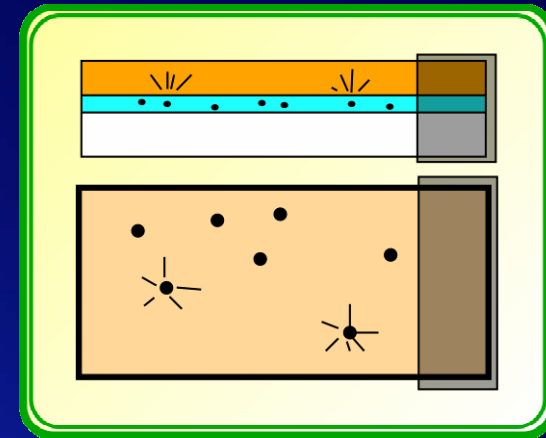
Prepare irradiation sample



TIMS

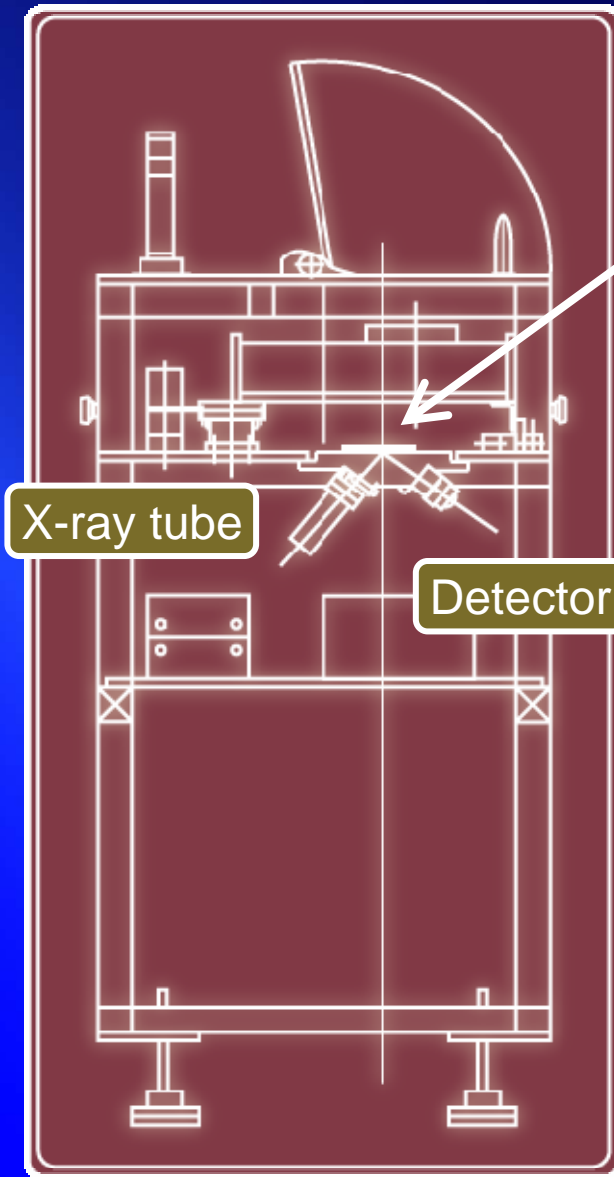


Etching



Accurate method
A lot of time to measure

XRF for screening



Swipe sample

X-ray tube

Detector

Uranium amount

Impurity measurement

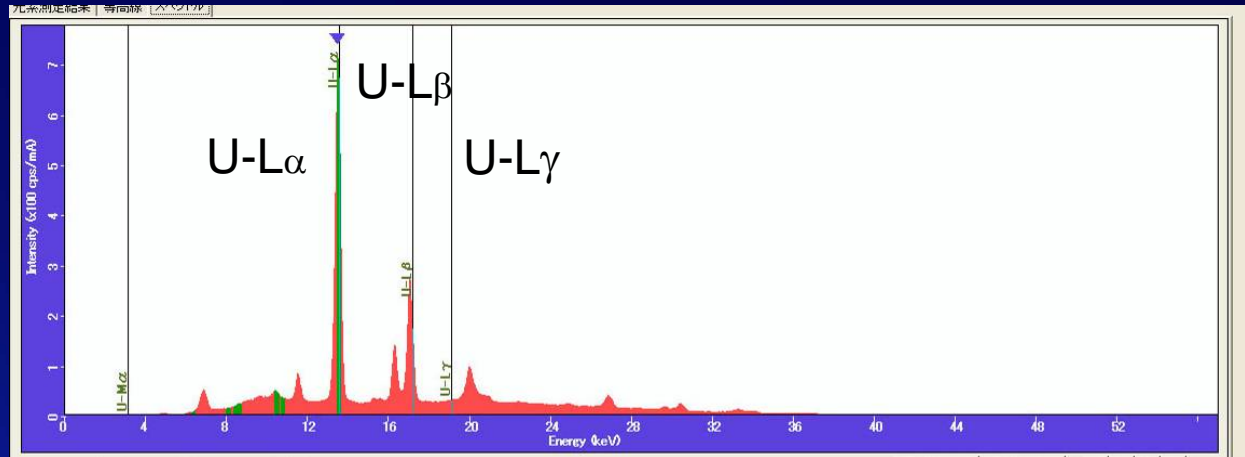
Pb, Fe etc.



Results of XRF measurement

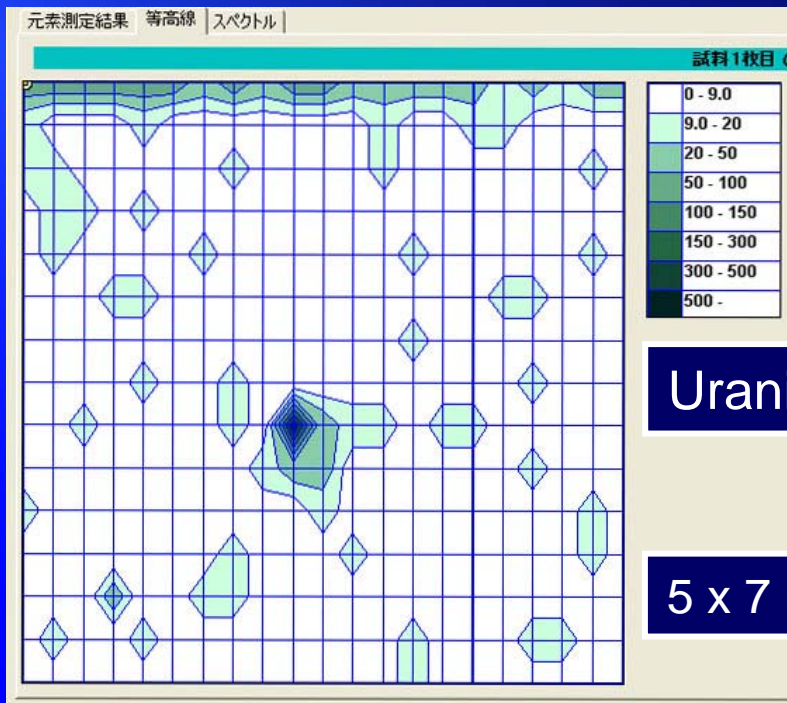


Intensity (x100 cps)



Energy (keV)

X-ray spectrum



Uranium

5 x 7 mm

Semi-quantitative measurement

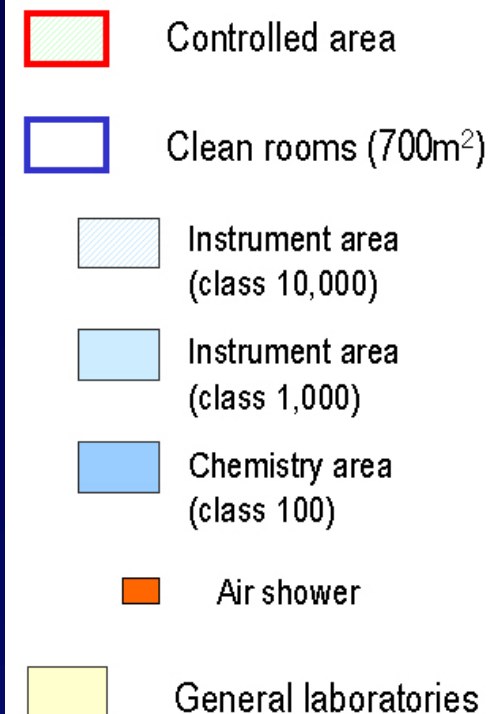
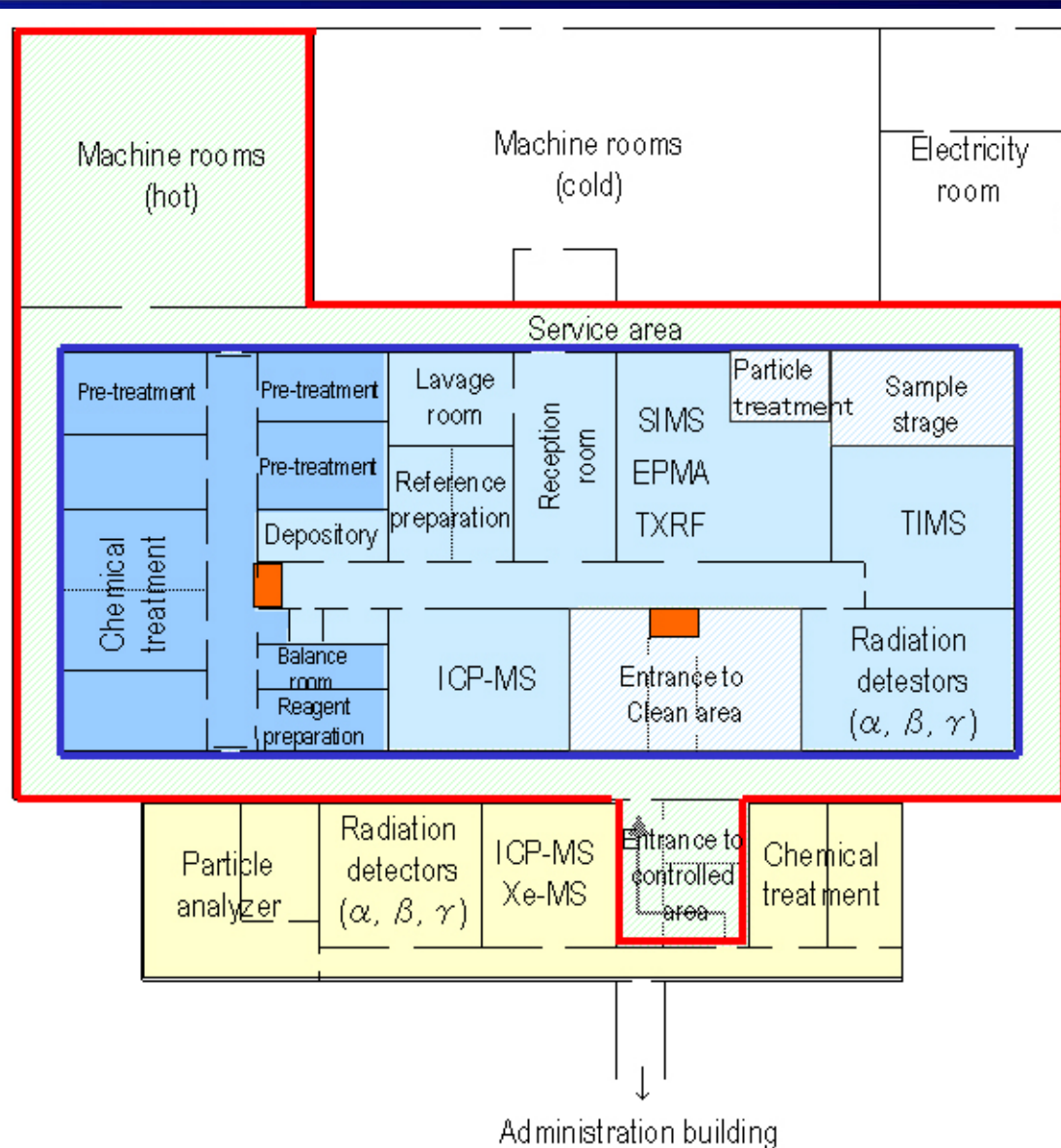
Elemental distribution

U, Pb, Fe etc.

CLEAR



CLEAR – Floor plan



Cleanness class is defined as maximum number of particles ($> 0.5 \mu m$) 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

