Joint Research Centre (JRC)

Advances in Nuclear and Environmental Analysis for Safeguards Purposes.

Magnus Hedberg, Herbert Ottmar, Piet van Belle, Said Abousahl, Sten Littman, Carin Kastelyn, Lilly Duinslaeger, Ylva Ranebo, Jaroslava Pravdova, Nicole Erdmann, Klaus Luetzenkirchen, Christine Vincent, Noelle Albert

Nuclear Safeguards and Security Unit - ITU

Introduction

This presentation will cover topics related to European and International Safeguards:

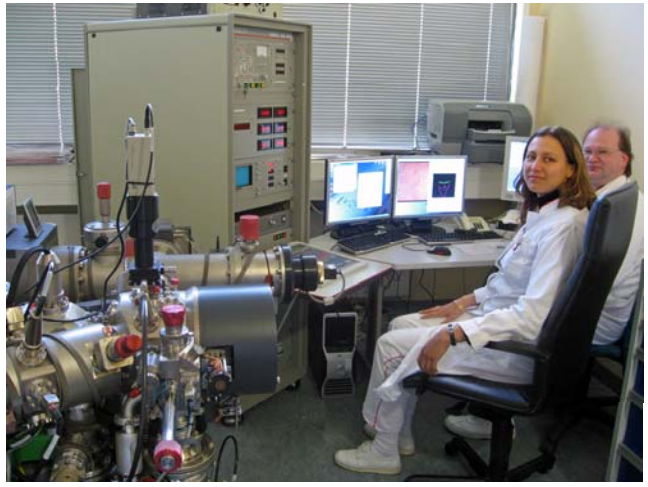
- Analysis of trace amounts of U and Pu from dust samples containing aerosol particles released by nuclear material handling.
- Traditional analysis for Nuclear Material Accountancy (NMA) using DA and NDA methods.

Environmental sampling

Sampling of dust at a nuclear facility using a cotton swipe.

Samples, often in duplicates, are sent to laboratories for Fission Track / TIMS and SIMS analysis

FT/TIMS



SIMS

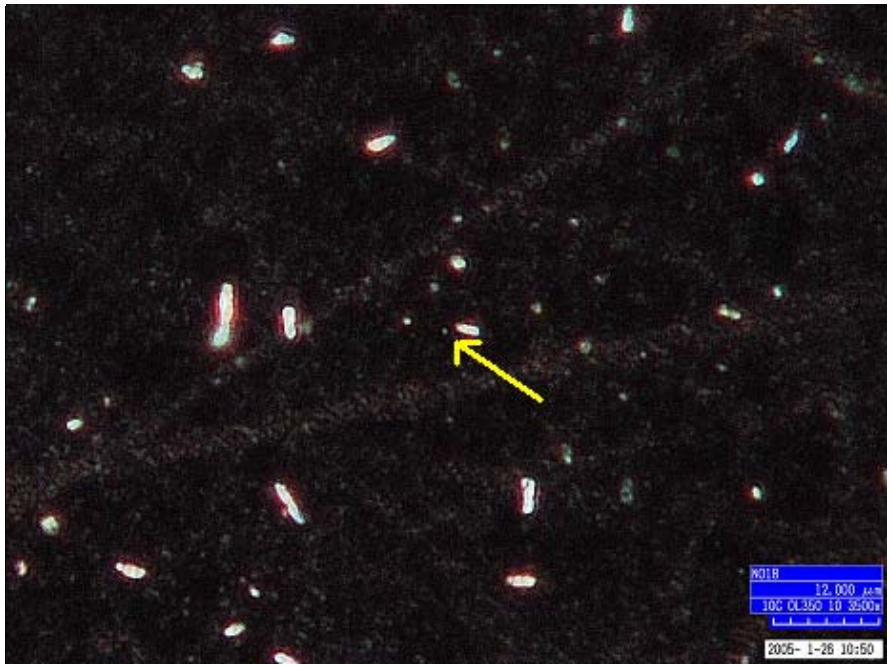
New results from UHS-SIMS evaluation

- A detailed evaluation of the UHS-SIMS method to compare with the current SIMS technique is just being finalised with a very positive result for the new method. (This work is part of a PhD thesis, Y.Ranebo)
- New tools are being developed that will significantly improve the capabilities in particle analysis by SIMS.
- ITU have started a small scale operation in analysing real inspection sample by UHS-SIMS. A first set of samples have already been reported.

The Task

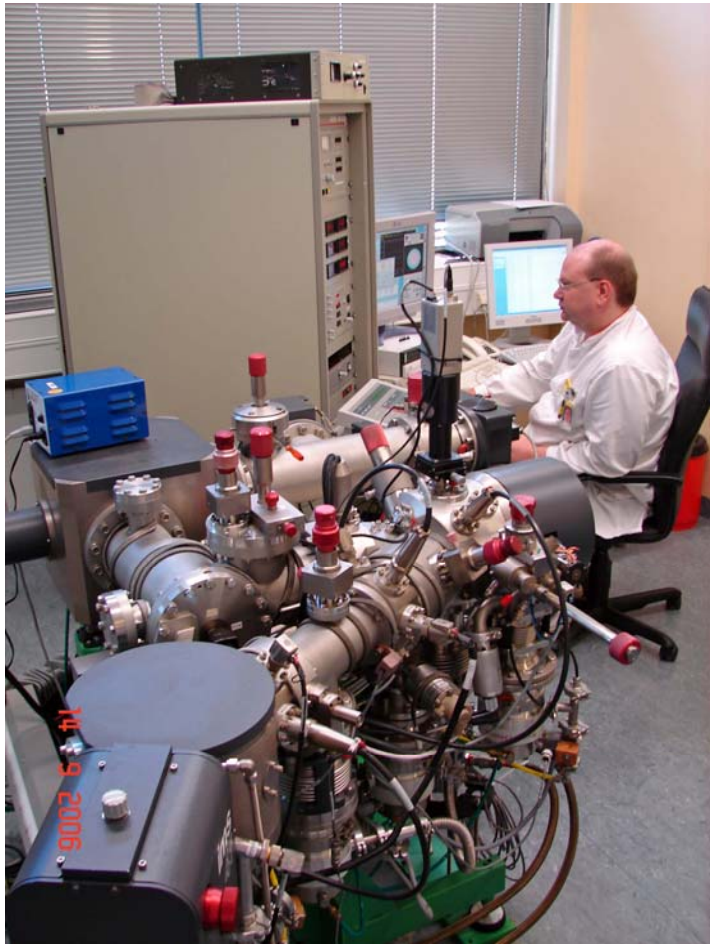
The main objectives in particle analysis:

- To search through millions of particles to find the particles of interest. A classic “needle in the haystack” problem performed under strict time pressure.
- To make precise and accurate measurements of both major and minor isotopes.



Particle distribution. (Resolution 1050x and 3500x).

Secondary Ion Mass Spectrometry (SIMS)



Cameca 4F at ITU.

+ Timeliness and throughput.

+ Ability to locate U particles in a matrix of other materials.

+ Distribution of enrichments in rich samples. (If automated imaging and data processing is available).

- Accuracy and precision in samples with background interferences.

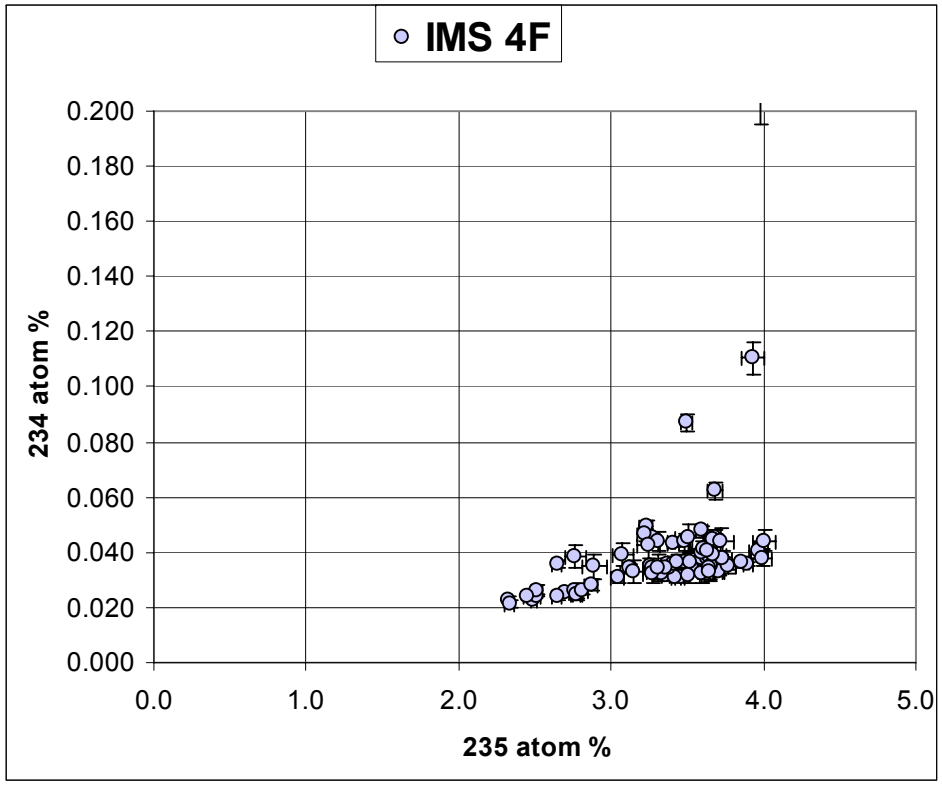
An alternative to SIMS: UHS – SIMS Tested at NORDSIM, Stockholm

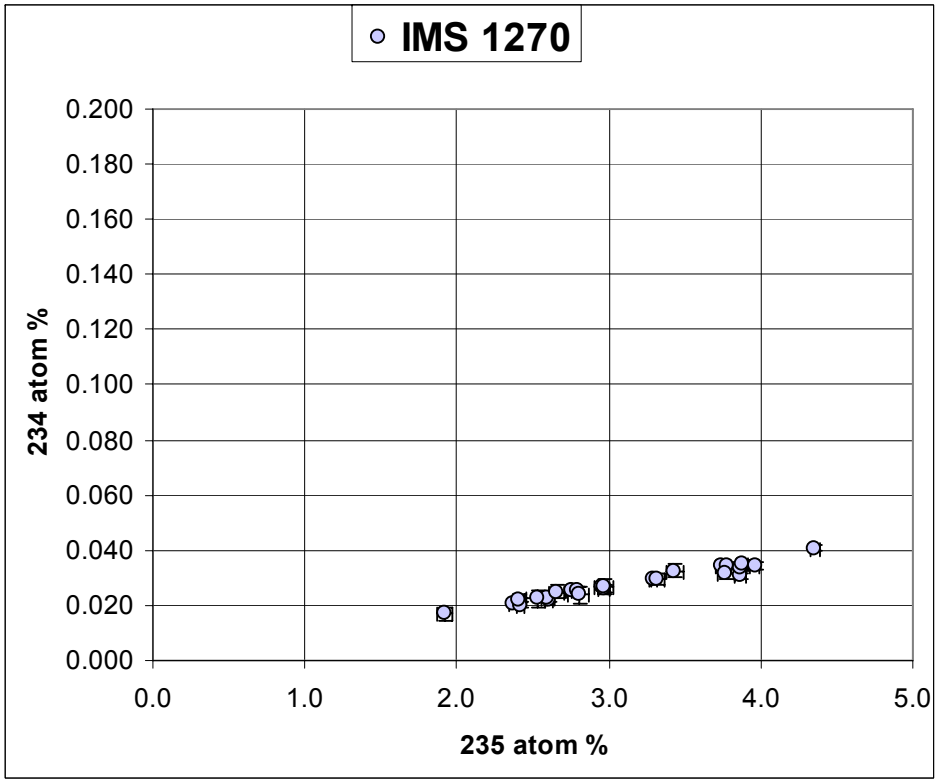
NORDSIM control room for IMS 1270.

IMS 1270 laboratory.

Example of background problem

Sample 1, 234/235



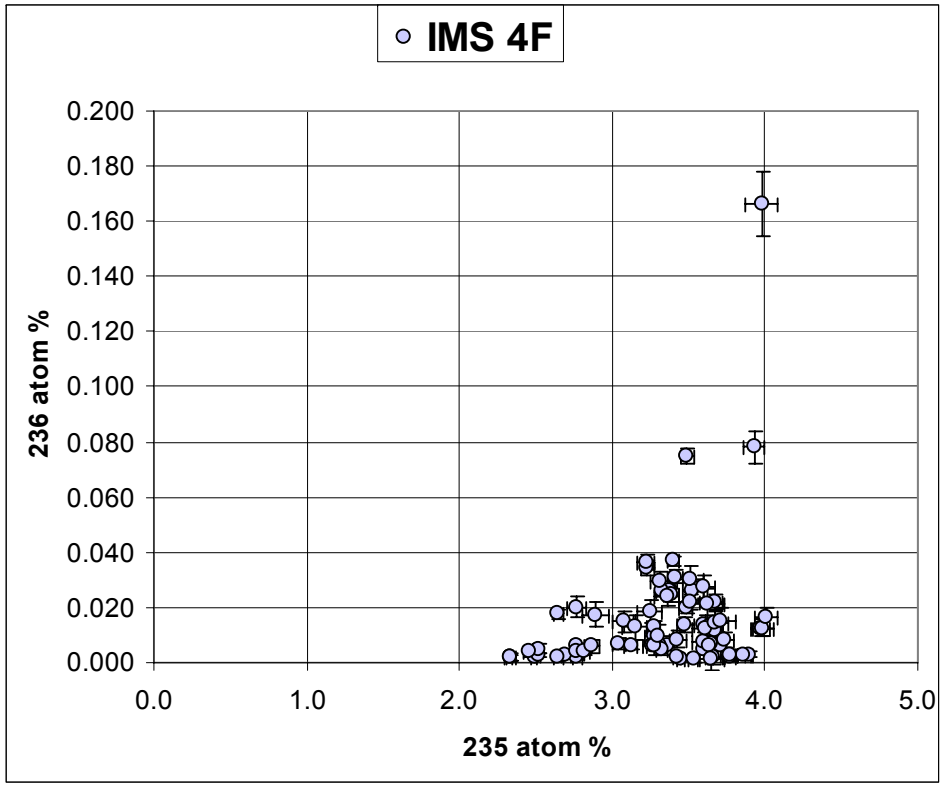


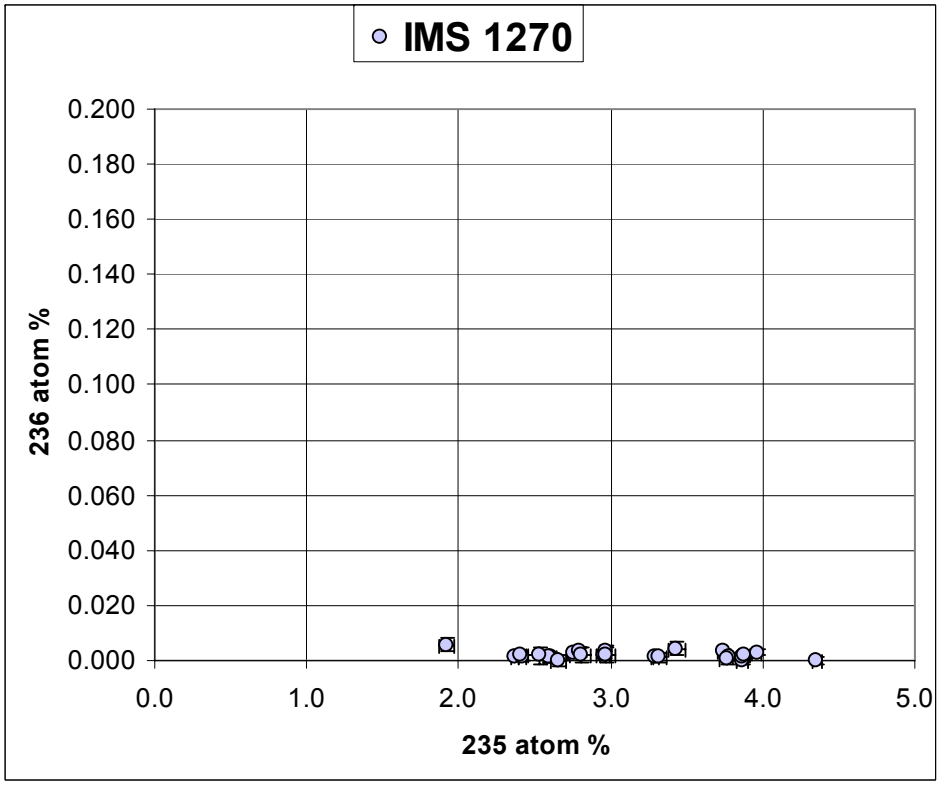
234/235 micro beam measurements from a sample taken at an enrichment facility.

Institute for Transuranium Elements

Example of background problem

Sample 1, 236/235



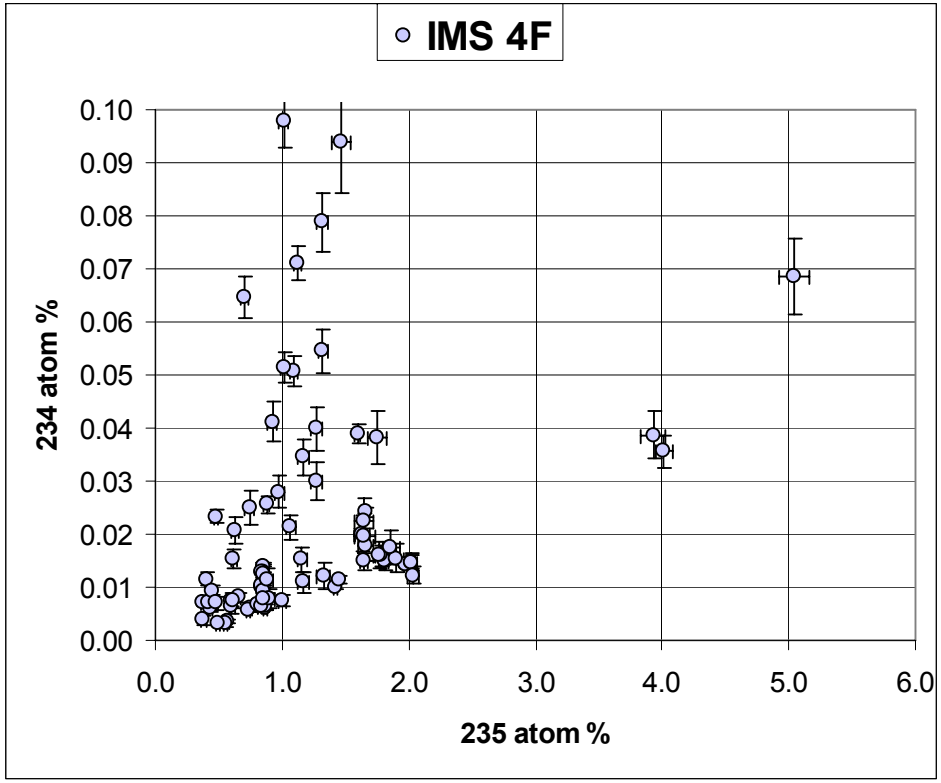


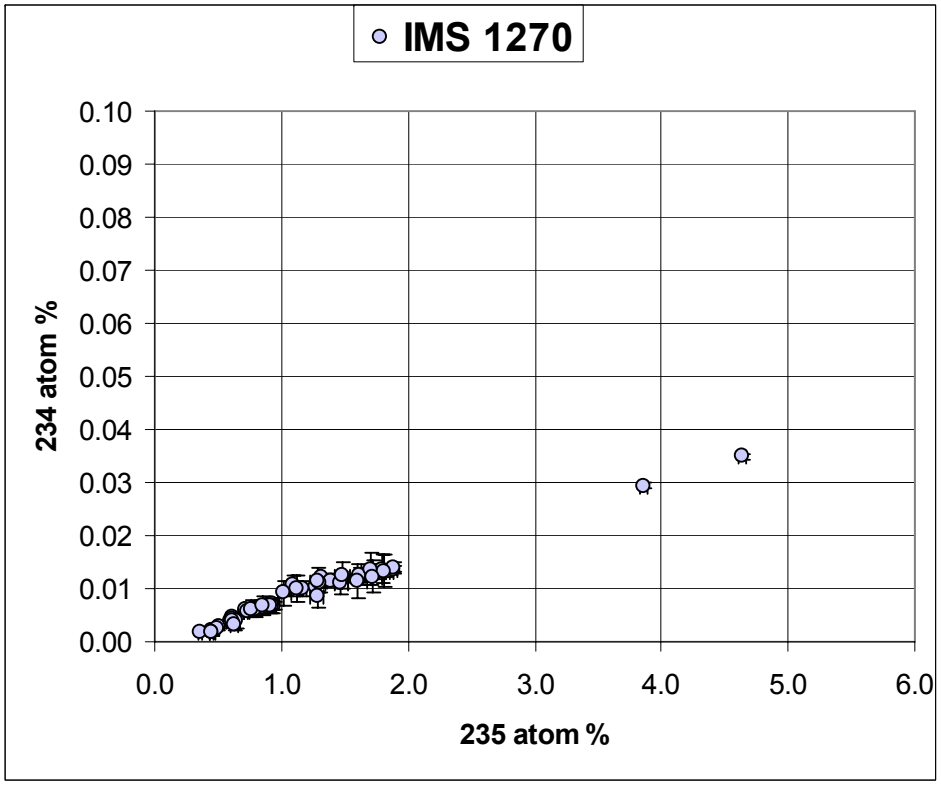
236/235 micro beam measurements from a sample taken at an enrichment facility.

Institute for Transuranium Elements

Example of background problem

Sample 2, 234/235



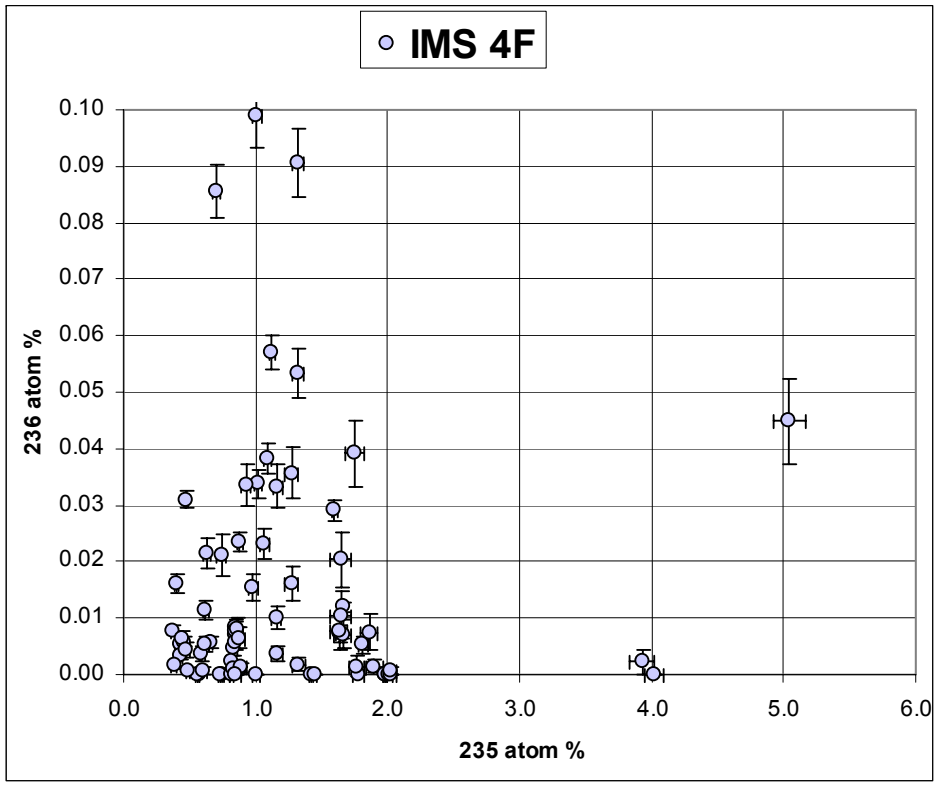


234/235 micro beam measurements from a sample taken at an enrichment facility.

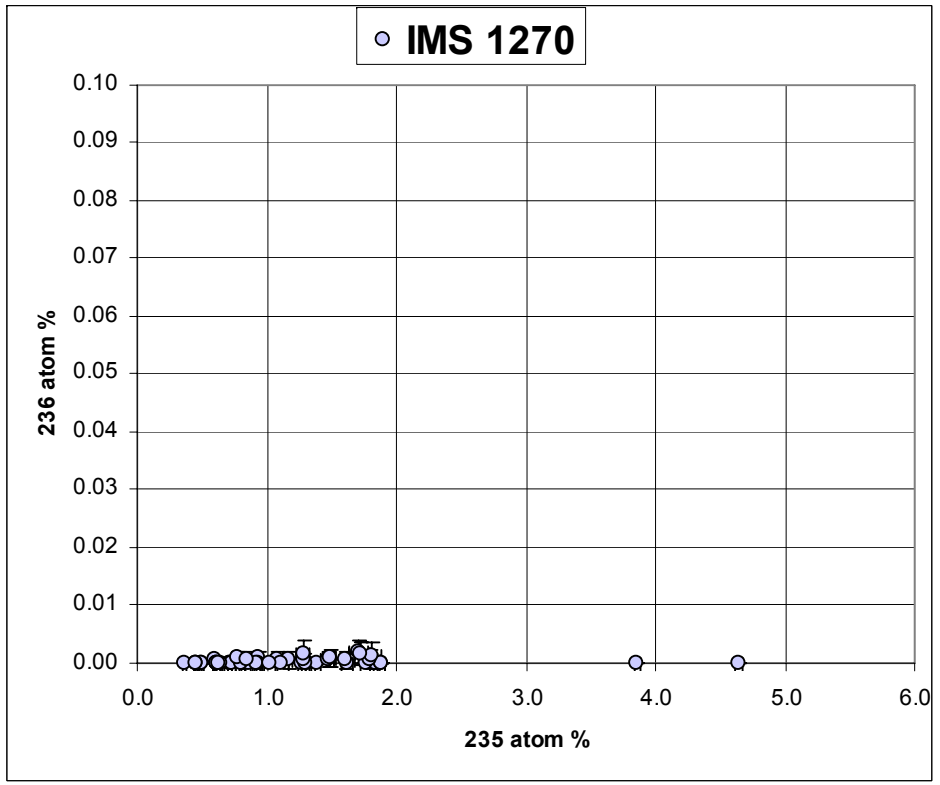
Institute for Transuranium Elements

Example of background problem

Sample 2, 236/235



236/235 data from 4 days of measurements on the IMS 4F.
Are the 236 results correct?



No, the 4F results are mainly background! It took 1 day to correctly characterise the same sample with the IMS 1270.

Institute for Transuranium Elements

Evaluation SIMS versus UHS-SIMS

SIMS:

• Good sample preparation can help in reducing the background problem.

• The correctness in the data can be significantly improved by removal (non reporting) of measurements with identified background interferences. Methods: Removal of data with high 234/235 ratios, do not report data from small particles, make mass scans to examine the background.

• There is however no guarantees that faulty data is still not reported when there is matrix materials in the SIMS sample.

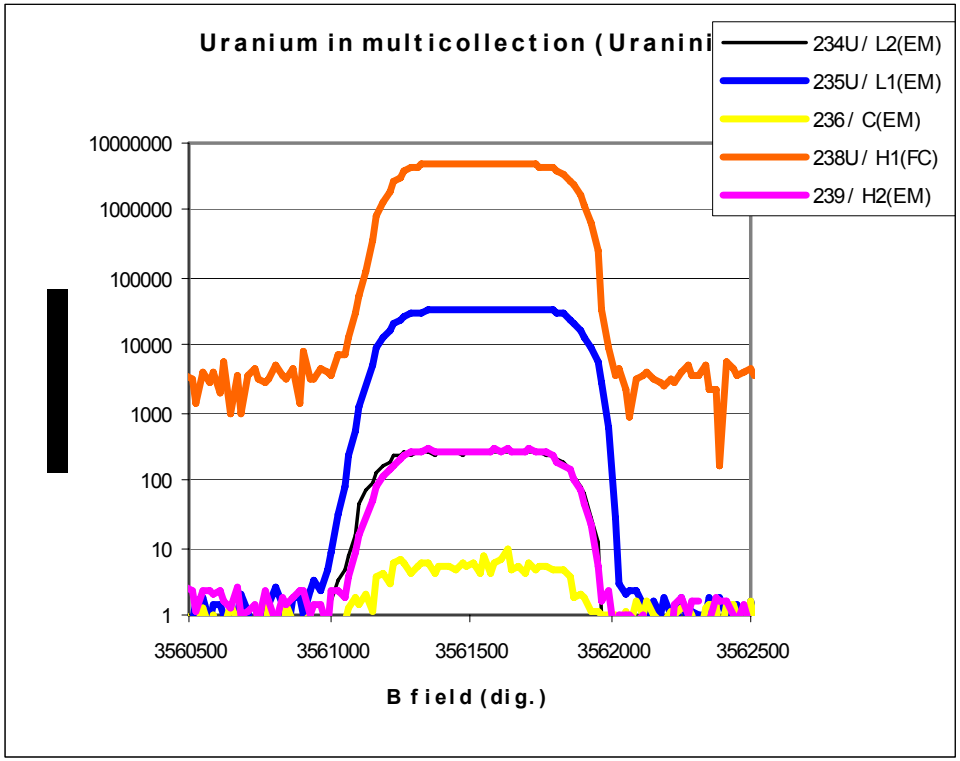
UHS-SIMS:

• The main problem with the SIMS measurements, the background, is removed effortless.

Evaluation SIMS versus UHS-SIMS

Further enhancements in UHS-SIMS:

- Multi ion counting and imaging using a multi ion detector system.
- New software tools dedicated for particle analysis that will significantly enhance the analytical performance.

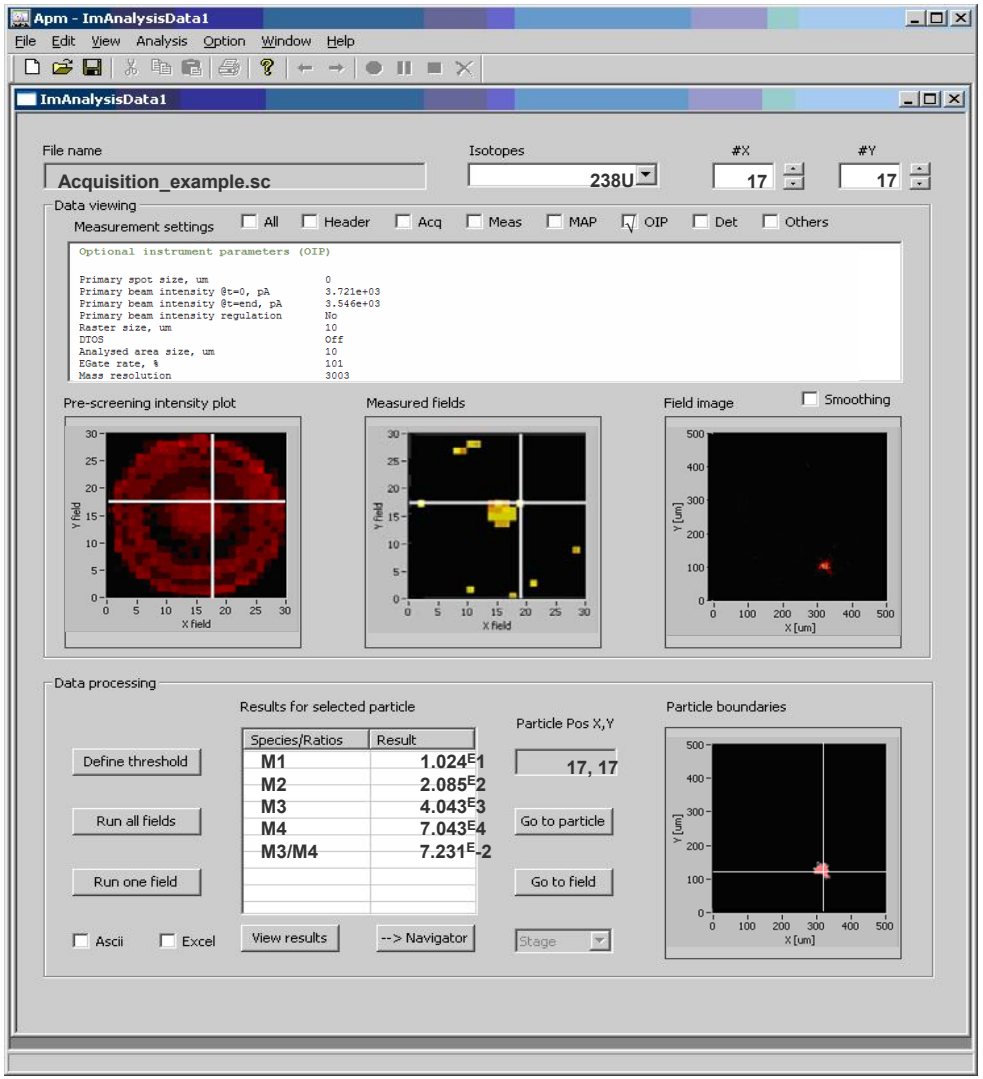


Mass scan over the U isotopes using a mixed FC/EM multi collector configuration on a IMS 1280. (Plot made by P. Peres Cameca)

New developments in SIMS: APM software

Automatic Particle Measurement (APM) Software

- Automatic two dimensional scan over the sample: defined ion images are recorded if the intensity for a given signal is high enough.
- Image data visualization
- Image data processing to automatically determine the particle boundaries, location and its isotopic composition by overlaying images of different isotopes.
- Automated chained analysis with micro beam measurements for best isotopic analysis. Particle locations can be imported from Image data processing.



New CAMECA APM software that will be available for: IMS 6f-PC, 7f, 7f-Geo, 1280

Institute for Transuranium Elements

Evaluation SIMS versus UHS-SIMS

Main conclusion:

- **A change from SIMS to UHS-SIMS will significantly improve the quality and reliability of the particle analysis and reduces the risk of incorrect reports of sample results.**
- **As the work of analysing safeguards samples is greatly simplified it also improves the timeliness.**

A program has been initiated at ITU to build up the experience in analysing inspection samples by UHS-SIMS. A first set of samples have been successfully analysed in Oct. 2007. The aim for 2008 is to analyse 10-15 more samples by UHS-SIMS.

Future directions at the JRC in particle analysis

Main topics in the work on particle analysis at the JRC:

- Work has been initiated by ITU and IRMM to establish a joint R&D and analytical UHS-SIMS laboratory to replace the older IMS 4F currently used.
- Collaboration between IRMM and ITU in producing certified U and Pu particle materials.
- Continue and strengthen the programs for method development in the field of particle analysis.

Further development of particle analysis for Safeguards purposes is a priority.

Traditional Nuclear Safeguards: On-Site Laboratories

LSS La Hague. (Since 2000) Hotcells for handling dissolved spent fuel samples.

OSL Sellafield. (Since 1999) Robotic system for small spiking, separation chemistry and alpha counting

Traditional Nuclear Safeguards: On-Site Laboratories

Analytical techniques:

- IDMS for Pu and U concentration (primary reference method)
- TIMS for U and Pu isotopics (primary reference method)
- K-edge densitometry / X-ray absorption (Pu and U concentration)
- X-ray fluorescence (U/Pu ratio, absolute low Pu and U)
- Gamma spectrometry (Pu isotopics, Am/Pu ratio)

Improving the primary methods IDMS and TIMS.

- **IDMS**: One fundamental in the IDMS measurements is related to the certification and QC of the LSD spikes used. (Has been the topic of a separate meeting at this workshop).
- **TIMS**: Main issue is in how to improve the accuracy in the performed total evaporation measurements and how to improve the uncertainty estimations on individual measurements.

Combined CAlorimetry, NEutron coincidence counting and GAmma spectrometry system.

- Overcome the ^{242}Pu problem in NDA by combining analytical methods
- Improve the Pu-mass determination through improved Pu isotopics.
- Enhance measurement confidence and reliability through complementary and redundant information.
- Evaluate the possibility to combine the different metods into one instrument.

Small sample calorimeter

Water bath calorimeter equipped with up to 4 independent twin calorimeter units.

COMPUCEA 2nd generation for U measurements

(Combined Procedure for Uranium Concentration and Enrichment Assay)

Purpose:

In-field analysis of uranium samples (powders, pellets) during Physical Inventory Verification (PIV) in LEU fuel fabrication plants (France, Spain, Belgium, Sweden) The method have now been used during PIV campaigns for more than a decade

Measurement System:

- X-ray absorption L-edge densiometry for U concentration.
- HRGS with a $LaBr_3$ detector for ^{235}U enrichment determination.

❑ Destructive method as it requires sample dissolution prior to the radiometric measurements.

❑ The COMPUCEA equipment is transportable and travels between sites.

❑ Performance within International Target Values: (U cons. RSD<0.25%. U enrichment RSD<0.45%)

COMPUCEA procedure

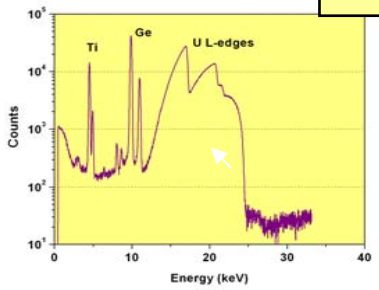
Sample weighing

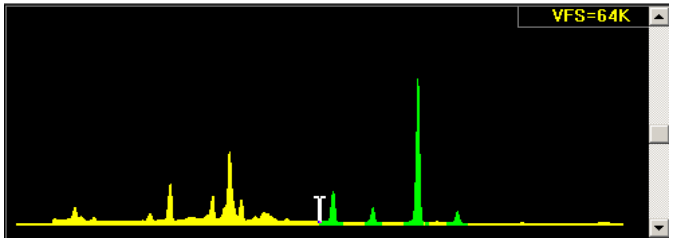
Dissolution (final solution: 3M HNO_3)

Solution density and temperature

L-edge measurement
→ U concentration

Aliquot weighing + γ-counting
→ U enrichment





Data evaluation

Results for inspectors

Thank you for your time. Questions?