Advances in non-proliferation and nuclear forensics analysis at CEA/DIF

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Analytical laboratories at CEA/DAM are part of the NWAL (Network of Analytical Laboratories of the International Atomic Energy Agency – IAEA) for the analysis of environmental samples since 2001 for both bulk and particle analysis.

Bulk analysis gives an average composition of the samples. Uranium and plutonium are separated on ion exchange resins and isotopic analyses are performed on Inductively Coupled Plasma Mass Spectrometer (ICP-MS). For particle analysis the isotopic composition of individual particle is determined in order to check consistency of declaration and to identify possible undeclared activity inside inspected nuclear facility. Isotopic composition of particles can either be measured with Fission Tracks Thermal Ionization Mass Spectrometer (FT-TIMS) or Secondary Ionization Mass Spectrometer (SIMS) techniques. The developments carried out at CEA for Safeguards are focused on the reliability and the precision of the measurements.

Developments for Safeguards

Our main objective for bulk analysis is to have more robust measurements and avoid false Pu detection. There is a high background level at Pu masses (Fig. 1). 20% of this background is due to polyatomic interferences that are combinations of one heavy element like Pb, Hg, Ir, etc with some of the most abundant atoms in the plasma (O, N, H, Cl, Ar).

Two methods have been developed in order to avoid overestimation of Pu. The first method consists in the estimation and the correction of these interferences in each sample [1, 2]. It is then necessary to determine each day the formation rates of polyatomic interferences. It has been applied for several years now and proves that in many cases it avoids false detection of plutonium at the femtogram level. Nevertheless this method has two main issues: (i) it degrades the detection limit of plutonium and (ii) it is time consuming. Another method has then been implemented in our laboratory. It combines the use of medium mass resolution of the ICP-MS and a desolvation introduction system [3].

Regarding particle analysis, our main concern is the detection of minor isotopes ($^{234}$U and $^{236}$U) in single particles. SIMS analysis of minor isotopes may be biased by polyatomic
interferences and TIMS lacks of sensitivity for measuring minor isotopes in particles smaller than 1 µm (frequently encountered in environmental samples). All our efforts are concentrated on the improvement of the minor isotopes detection limit. A new TIMS Triton (Thermo, Bremen) equipped with an array of 5 channeltrons allows the simultaneous measurement of all U isotopes. Figure 2 shows the increase in the precision of $^{234}\text{U}/^{238}\text{U}$ isotopic ratios on particles from a real sample.

![Figure 2: Comparison of the $^{234}\text{U}/^{238}\text{U}$ relative standard deviation on uranium particle analysis using single collector old generation VG TIMS and multiple collector new generation Thermo “Triton” TIMS.](image)

We also studied the feasibility of Laser Ablation - ICP-MS (LA-ICP-MS) for the isotopic analysis of single sub-micrometer uranium particles [4]. This technique allows rapid measurements compared to the other techniques traditionally used, as soon as particles have been located beforehand. For this we used either Scanning Electron Microscope (SEM) or fission tracks. It has been applied to a sample from the “NUSIMEP 6” interlaboratory exercise, organized by IRMM (Geel, Belgium) (Figure 3). Values are in good agreement with the certified value.
Capability development for nuclear forensics analysis

All the expertise inherited from environmental analysis is now used to develop capabilities in nuclear forensics analysis. It concerns the age dating of U material and the geolocation of U-ores. These two projects are currently under development at CEA/DIF. For the first one, we have established a procedure to date small quantities of uranium (below 1 µg) with the radioactive couple \( ^{234}\text{U}/^{230}\text{Th} \). As our equipments are dedicated to trace analysis, only micro-quantities of nuclear materials can be handled in the laboratory in order to avoid contamination. We have then used micro-columns of 100 µL chromatographic resins to separate thorium from uranium. Measurements were performed on ICP-MS for Th and TIMS for U. The detection limit (DL) for \(^{230}\text{Th} \) determination is 1 fg. If we extrapolate this DL to particle matter, we would be able to date a 15 years natural particle of UO\(_2\) with a diameter of 40µm, and a 15 years highly enriched (93%) particle of UO\(_2\) with a diameter of 8µm. Another age dating procedure is being studied using the couple \(^{235}\text{U}/^{231}\text{Pa} \). Chemical procedure should be written within this year.

Concerning the geolocation, the project has just started at CEA/DIF. The objective is to establish the origin or production site of unknown materials.

Conclusion

Analytical laboratories at CEA/DIF are specialised in the isotopic analysis of trace quantities of uranium (typically nanograms) and plutonium (at femtogram level). The development carried out for Safeguards are focused on the reliability and the precision of the measurements. New analytical demands in the field of nuclear forensics drive the laboratory to develop capabilities in age dating of uranium material and geolocation of uranium ores. The ultimate goal would be to be able to determine the age of uranium micro-particle!

References

