Improvement of Laboratory Capabilities for Safeguards Measurements of Nuclear Material

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Abstract

The Directorate-General Energy (ENER) fulfils the safeguards obligations stipulated by the Article 77 of EURATOM treaty for European facilities. For physical verification, samples are taken at different stages of the process and are being analysed either on site or shipped for analysis. The Joint Research Centre (JRC) carries out destructive and non-destructive analyses of nuclear material for ENER on the site of JRC-Karlsruhe and on the site of the reprocessing plant from Orano, at La Hague, France.

Safeguarding of nuclear material strongly depends on state of the art analytical performance. It is crucial for inspectors to receive timely analytical results with lowest achievable uncertainty for facilities handling bulk material. The JRC is operating analytical instruments fit for purpose, ensuring continuity and quality control of the generated analytical results. The quality of these results is monitored through participating in inter-laboratory comparison exercises and by the use of traceable reference materials. The JRC strives for continuous improvement through ambitious R&D projects in order to keep up with global evolving technologies and cutting-edge scientific developments.

The availability of “off the shelf” analytical techniques adapted to the needs of the small nuclear safeguards community is limited. The JRC’s vision is to keep up with the state of the art technology and succeeded to modernise the infrastructure. Two MAT262 mass spectrometers have been upgraded to state of the art electronics and hardware. The newly refurbished mass spectrometers provide net advantages, which will be presented.

Furthermore, new “off the shelf” alpha spectrometry stations have been modified for measurements inside glove boxes, as no other options were available. They replace the outdated equipment making use of modern software and electronics. Maintenance for the foreseeable future is thus guaranteed.

The paper below presents the experience of operating the newly up scaled and recycled instruments.

1 Introduction

Article 77 of the Euratom Treaty (*1*, *2*) assigns the duty to implement nuclear safeguards the European Commission (EC). The EC shall “… satisfy itself that …. special fissile materials are not diverted from their intended uses as declared by the users.” The Directorate-General Energy (ENER) fulfils these safeguards obligations by physical and surveillance verifications. For physical verification, samples are taken at different stages of the process and are being analysed either on site or shipped for analysis. Joint Research Centre (JRC) is a scientific organisation and its values of Integrity, Innovation and Accountability are of great help for the safeguards community. The absolute impartiality of results, the state-of-the-art applied technologies and the reliability of the services provided, are key requirements for the mission of safeguarding nuclear material. The Analytical Service (AS) of the JRC carries out destructive and non-destructive analyses of nuclear material for ENER on the site of JRC-Karlsruhe and on the site of the reprocessing plant from Orano, at La Hague, France in behalf of DG ENERGY. The laboratories analyse yearly about 1000 samples containing the two key elements uranium and plutonium delivering independent results. The high precision of the results is crucial for the drawing of solid safeguards conclusions. The innovation function of the JRC is equally important for the community, as the state of the art can only be maintained by continuous R&D projects in order to keep up with global evolving technologies and cutting-edge scientific developments.

2 Analytical Service mission and activities

The AS has been the strategic operational partner of DG-ENER for the last decades, performing the analytical work required by the different type of samples, and at the same time, providing tailored analytical solutions for emerging requirements. In times of scarce resources the balance between the large service- (i.e. analytical measurements) and the R&D – components must be constantly adjusted to keep up with the technological progress and thus, maintain the relevance of the service.

The AS of JRC extends its function besides the partnership with policy partner, DG-ENER, to collaborations with other international bodies in the field of nuclear safeguards. The collaboration with the International Atomic Energy Agency (IAEA) is one of the most intensive, and consists of analytical work but also in exchange of technological solution and operational practices. Equally important are the partnerships with the United States Department of Energy (U.S. DOE), the French Alternative Energies and Atomic Energy Commission (CEA), the Brazilian Argentine Agency for Accounting and Control (ABACC), The Korean Institute of Nuclear nonproliferation And Control (KINAC) and the Japan Atomic Energy Authorities (JAEA). In the frame of these collaborations, the AS performs sample analyses, provides high quality results and strengthens the technological developments in the field of nuclear safeguards.

Following the JRC value of accountability, the AS succeeds to stay highly relevant by delivering on the commitments and taking responsibility for the outcomes. Being aware of the weight of its mission, to deliver quality results in a timely manner, the AS has been constantly substantiating the analytical performance by employment of methods under accreditation, delivering results traceable to certified reference materials and participating to international inter laboratory comparison exercises.

The AS operates thirteen methods, out of which eleven are accredited by the German accreditation body DAKKS ISO/IEC 17025:2018. Table 1 shows a summary of the methods applied in the AS, with their primary purpose.

Table 1. Analytical Service: Methods and their application

Method	Type	Primary use
Neutron Coincidence Counting (NCC)*	Radiometric	Pu - assay in high burnup fuel
Calorimetry	Radiometric	Pu, Am - assay
High Resolution Gamma Spectrometry (HRGS)	Radiometric	U, Pu - isotopic abundance Detection of fission products, Verification of the presence/absence of contamination with gamma emitting nuclides in samples*
Density measurements	Destructive	Determination of density for liquids
Hybrid K-edge / XRF densitometry (HKED	Radiometric	U, Pu - assay
Combined Procedure for Uranium Concentration and Enrichment Assay (COMPUCEA)	Radiometric	U - assay and isotope abundance
Total Evaporation-Thermal Ionization Mass Spectrometry (TE-TIMS)	Destructive	U, Pu - isotopic abundance
Modified Total Evaporation-Thermal Ionization Mass Spectrometry (MTE-TIMS)	Destructive	U- isotopic abundance of minor isotopes
Isotope Dilution Mass Spectrometry (ID-TIMS)	Destructive	U, Pu - assay
Alpha Spectrometry	Destructive	$^{238}Pu/^{(239+240)}Pu$ isotope abundance
Inductively Coupled Plasma Mass Spectrometry (ICP-MS)	Destructive	determination of: - impurities in Uranium and/or Plutonium matrices - trace level elements in aqueous media - single elements at high precision - isotopic composition of single elements at very low levels
Large Geometry - Secondary Ion Mass Spectrometry (LG-SIMS)	Destructive	Environmental Sample Analysis -search for undeclared nuclear activities
Elemental Analysis (ELANA)*	Destructive	Elemental analysis of carbon, oxygen and nitrogen

* - out of the scope of accreditation

By participating to ILCs the AS compares its results to those of world-class laboratories, and attempts to assess the entire range of methods. The performance delivered during these exercises has been excellent as exemplified by Figure 1, showing the results of the AS right on target.

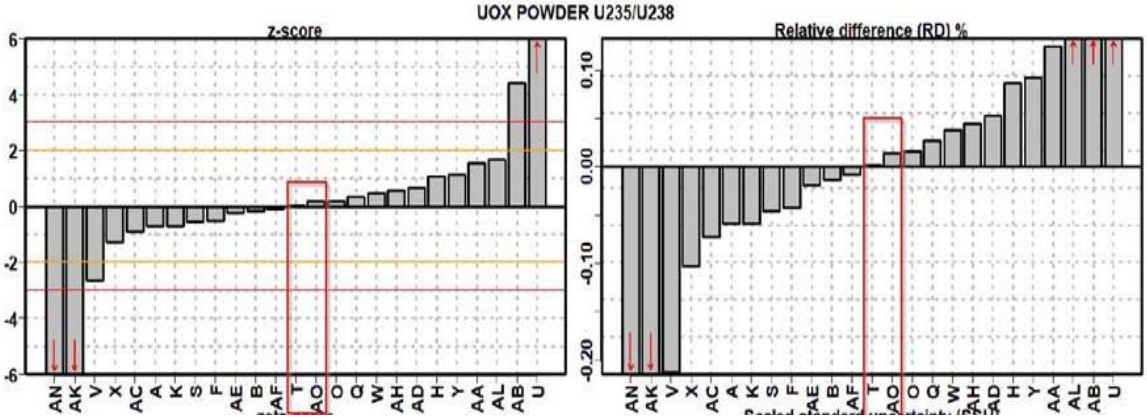

Figure 1. The performance of AS (laboratory codes T –TIMS and AO ID-TIMS) during the Nuclear Material Round Robin (NMRoRo 2022) exercise (preliminary report produced by the IAEA*)*

The exercises help also to identify opportunities for improvement, which are treated with highest consideration by the AS. The methods are investigated and appropriate actions are immediately taken as considered necessary.

As further means for maintaining the relevance of results the AS is determined to maintain its methods and instrumentation at state-of-the-art. Continuous improvements are necessary and, where deemed necessary, new instruments are acquired or developed. Due to the highly specialized appliances of the AS, the availability of such instrumentation is rather limited. The instruments are most of the time not off-the-shelf readily available, they require customization for the use in the nuclear field. Mass spectrometers and alpha spectrometers are not available ready to be used in combination with glove boxes. They need modification either at the producers or directly in the laboratories and workshop of the JRC-Karlsruhe.

With the aging of equipment the AS has deemed necessary to revamp the mass and alpha spectrometry equipment used in the laboratories from Karlsruhe first and at a latter stage the LSS –La Hague. The advantages of having a “replicate” strategy for the set-up of the laboratories were recognized from the early stage of concept and design of the laboratories. The laboratories in Karlsruhe profit from a better infrastructure and larger availability of the design office and electro-, mechanical- workshop and they serve for training of personnel before sending them in missions to LSS as well. The research and development work is performed thus in a first step for the equipment in Karlsruhe, following a second stage of implementation of the developments in LSS after the proof of concept and of good functioning have been validated in Karlsruhe.

The developments and the implementation steps of the revamped instruments are described in the following chapters.

3 Alpha spectrometry

Alpha spectrometry has a dual use in the AS of the JRC-Karlsruhe. It serves for the estimation of the concentration of plutonium in solutions prepared for mass spectrometric analyses, but also for the calculation of isotopic composition of plutonium together with the results obtained by mass spectrometric determinations.

Since 1997, the AS has been using in-house made hardware and software combined with commercially available electronics for the analyses. The electronic equipment - Nuclear

Instrumentation Module (NIM) - has altered with the time and replacement parts are no longer available on the market. Similarly, no compatible IT systems can be found in order to sustain the initial software. A general refurbishment of the system has been deemed mandatory in order to assure continuity of this technique.

Only two main competitors providing equipment suitable for the needs of the AS, AMETEK/ORTEC and MIRION/CANBERRA could be considered. The decision was taken to acquire the spectrometers Alpha-DUO Benchtop Spectrometer with the Maestro Software, provided by AMETEK/ORTEC due to its compact size, required by the application in and near glove boxes [3].

The acquisition process was completed in May 2021 with the delivery of three Alpha Duo ensembles, followed by the official hardware and software installation and testing performed by an AMETEK/ORTEC engineer in June 2021. The proper functioning of the system was confirmed. The installation was however, not completed to the level required, as no commercial system is adapted for the work with “external” analysing chambers, as required by the working conditions in the AS. Further work was carried out to adapt the instrument`s use in combination with a the glovebox environment.

3.1 Installation of the Alpha Duo spectrometers

The Alpha Duo ensemble shown in Figure 2 consists of a pair of two internal measurement chambers with the required electronics integrated.

Figure 2. Alpha Duo – a compact, all-in-one, bench-top, dual channel alpha spectrometer

Source: AMETEK/ORTEC

The instrument is, a compact, all-in-one, bench-top, dual channel alpha spectrometer. Not all these features represent an advantage for the intended use. All plutonium bearing samples have to be handled in glove boxes. Therefore, the only option for the direct use of the Alpha Duo instrument, would be to introduce it without further modifications into a glove box. However, due to the acidity present in the glove boxes, the electronics of the spectrometer would be prone to rapid deterioration. The solution opted was to connect the electronics of the Alpha Duo to the existing measurement cells used since 1997. These cells were originally fitted in a glove box and connected with coaxial cable of 50 Ω over S1 LEMO feedthrough to the NIM electronics fitted outside of box. All electronic parts belonging to the previous system have been disconnected from the measurement cells, in preparation for the installation of the Alpha Duo system. The coupling with the Alpha Duo is described in the following chapter.

3.2 Connection of the Alpha Duo to the measurement cells inside the glove box

The Alpha Duo spectrometer is designed to measure samples using its own internal measurement cell. On the right side of Figure 2 the golden Passivated Implanted Planar Silicon (PIPS) detector is connected inside the measurement cell. The most direct option to connect the Alpha Duo to the

PIPS detectors, inside the glove box, is by using a BNC/SMA adaptor as shown in Figure 3, left. This type of connection was nevertheless, not suitable and the spectra acquired with this setup were not delivering the desired resolution as shown in Figure 2, right. The separation of the three peaks is poor and the electrical noise is unacceptable high.

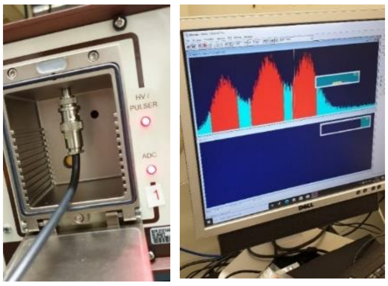

Figure 3. Alpha Duo electronics connected with external detectors, using a BNC/SMA adaptor, left. Spectrum of the AL-2843 Am-241, Cm-244, Pu-239 standard, right.

Source: EC JRC.G.II.8

Figure 4 presents the second attempt to connect the Alpha Duo Electronics to the external detectors. The cables leading to the external PIPS detectors were soldered at the exit point of the Alpha Duo internal cell. The green insulators which can be observed in Figure 4 are part of the original design and expose directly the soldering connection of the internal Alpha Duo own detectors with the electronics. This type of connection delivered also only poor resolution for the spectra and it was abandoned.

Figure 4. Alpha Duo electronics connected with external detectors, with connector cable soldered on the “green insulator”

Source: EC JRC.G.II.8

After several attempts, the source of electronic noise was identified to be generated partly by the quality of the soldering and partly by the insufficient – for this type of connection- electrical grounding of the Alpha Duo housing. The length of the soldering connection should be kept at the minimum technical achievable in order to have a clean signal transmission from the external PIPS detectors to the electronics of the Alpha Duo. One indication for the high level of electronic noise, is high “detector dead time” values during measurements. Under good operation conditions these values should not exceed 7 %. The length of the connector cables did not seem to influence the quality of the spectra acquired.

Figure 5 shows the final setup of the system with the cables connected as described above. It can be observed that the original cables have been detached from the green insulators. This had a positive effect on the electronic signal quality. As announced above, it was determined that the grounding of the external case of the Alpha Duos plays also a very important role in reducing the electrical noise in the setup as described above. The grounding must be done using individual copper cables of no less than 6 mm^2. The potential equalization is done such that all three cables are connected to a common ground socket, at one end, and to the backside of each Alpha Duo at the other end.

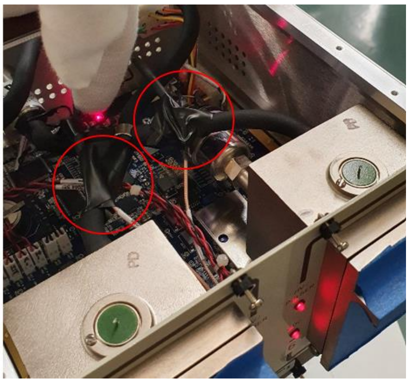

Figure 5. Connection of the cables inside of the Alpha Duo

Source: EC JRC.G.II.8

With the setup described above the spectra obtained from measurements of an unsealed radioactive source Am-241, Cm-244, Pu-239 from Eckert&Ziegler have shown very good resolution and a baseline separation. Figure 6 shows typical spectra obtained for measurement of the standard (above) and a sample prepared by the operators in the glove box under normal conditions. Both spectra show a *baseline separation* of the peaks, which indicate a clean measurement and data processing.

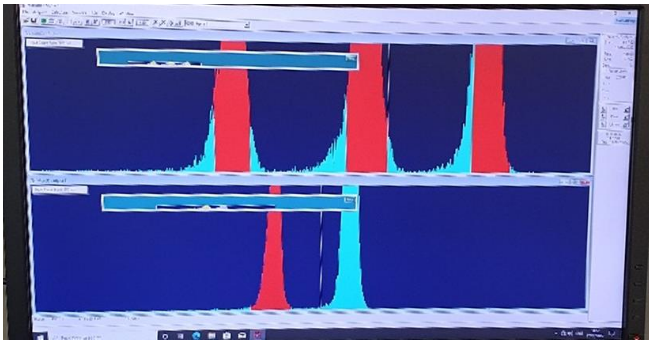

Figure 6. Spectra of the Am-241, Cm-244, Pu-239 standard on top and of a routine sample on the lower part

Source: EC JRC.G.II.8

With the assurance that the system is operating at its full potential, the last step of validating the data acquisition and processing was performed on the calibration source and three different LRMs.

For the calibration of the instrument, the above described source Am-241, Cm-244, Pu-239 from Eckert&Ziegler was used. The parameters monitored for the quality control (QC) have been the ratio between the activity registered on the decay energy of the Pu-238 against Pu-239+Pu240. Figure 7 presents a chart with the ratio measurements obtained by the old system starting from January 2020 and by the Alpha-DUO starting from February 2022. There is no significant difference between the means of the two systems and for the Alpha-DUO the standard deviation is slightly reduced.

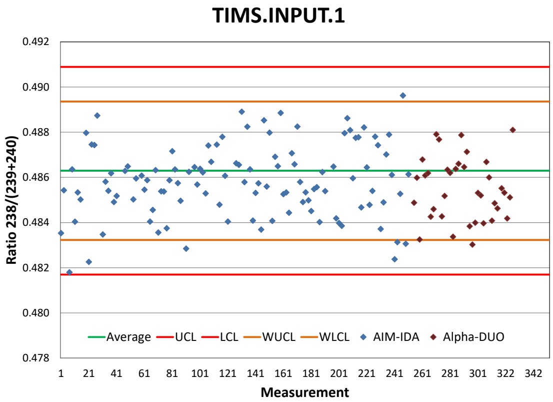

Figure 7. Control charts for the reference material TIMS.INPUT.1

Source: EC JRC.G.II.8

The values obtained with the new system for validation are fitting well in the QC limits defined in the previous system. All parameters relevant for measurements at the AS of the JRC-Karlsruhe have been assessed and the performance has been deemed satisfactory. Thus, the system has been validated and in use at the AS in Karlsruhe since July 2022. The validation report is restricted to JRC use and cannot be distributed.

4 Mass Spectrometry

Two Thermal Ionisation Mass Spectrometry (TIMS) instruments, type MAT26x were installed at the JRC Karlsruhe (1982) and at the Laboratoire Sur Site (LSS) La Hague (1999) for high precision U and Pu isotope ratio measurements of safeguards samples. The instruments are more than 20 years old, so original spare parts were becoming increasingly difficult to obtain. For the unit at JRC Karlsruhe a complete set of new electronics were considered. However, for the LSS spectrometer, only renewal of software, computers, and ion source head with accompanying high voltage (HV) supply unit have been considered sufficient. The similar strategy as announced at the previous chapter has been adapted, namely the progressive refurbishment would start in the laboratories in Karlsruhe and later the experience such gathered would be used for the refurbishment of the mass spectrometer in LSS following the points below:

a) Installation of the TI-Box in JRC Karlsruhe

b) Training the analysts in JRC Karlsruhe

c) Installation of the upgrade in the LSS La Hague

A complete upgrade of the electronics (TI-Box) and the renewal of the ion-source head was purchased from the company Spectromat for the mass spectrometer in Karlsruhe. By far the biggest challenge encountered during the refurbishment was the modifications necessary for the work in a glove box.

Figure 8 shows the spectrometer from Karlsruhe before and after the refurbishment.

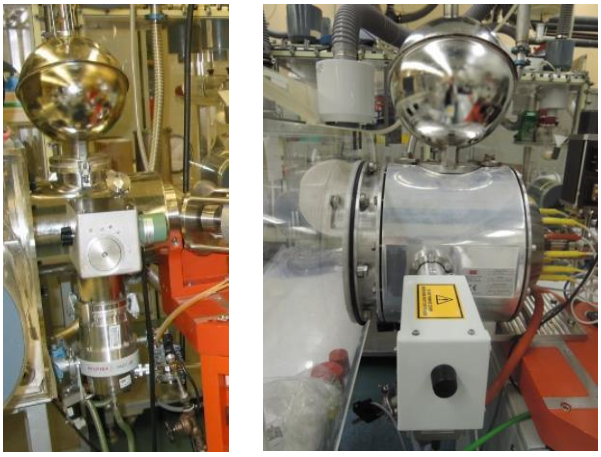

Figure 8, Ion source head, Mat262 left, TI-Box right.

Source: EC JRC.G.II.8

The new ion source head is easier to maintain and the exchange of the sample turret is also a much simpler task, as it slides on a rail into the MS rather than been screwed in [*4*] as can be seen in Figure 9.

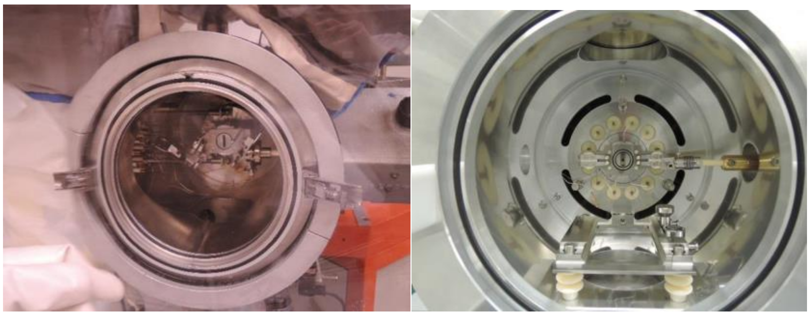

Figure 9 Ion source head - Mat262 left, Spectromat right

Source: EC JRC.G.II.8

The method has also been validated and been in use by the AS. Validation measurement for both, uranium and plutonium isotope composition show good agreement with measurements performed before upgrades as well as with external quality control comparisons. Figure 10 shows the results obtained using the old and the new system as comparison.

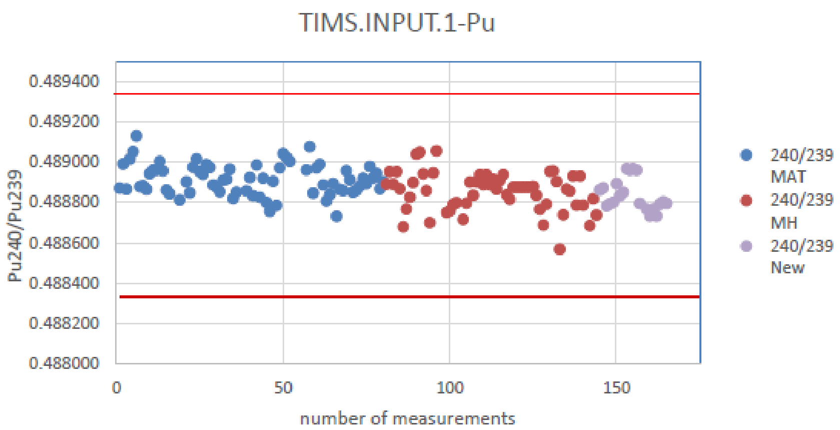

Figure 10 Results of the Pu-240/Pu-239 ratio comparison for LRM TIMS.INPUT1

Source: EC JRC.G.II.8

Following the successful installation of the TI-Box in Karlsruhe a training was given to the analysts and the scene was set for the upgrade in LSS to follow. A new ion source head was installed replacing the original one in LSS. This operation required also the change of the ion source HV supply unit. A new QHV2 ion source HV supply unit from Spectromat was fitted. Both hardware installations were performed within a week followed by software debugging caused by the fact that the TI-Box ion source head and the ion source HV supply unit have been attached to the old MAT 262 electronics. This made the software debugging much more difficult than the debugging process performed in JRC Karlsruhe. The lack of experience with the software and additionally problems with the old hardware made it difficult to identify and verify the various issues. Finally, in December 2018, a new software release RunIt26X V5.6 b-10025 was installed and the validation measurements completed.

5 Conclusion

The setup of the Alpha Duo and of the TI-Box systems in Karlsruhe and LSS- La Hague described above constitute solid tools for the measurements of Pu isotopic ratios by alpha and mass spectrometry respectively. The hardware and software are from a newer generation and are a necessary modernization of the existing systems. The actual systems are state of the art and replace the outdated system used by the AS and the LSS since the installation of the laboratories in the 1980s and 2000s respectively.

6 References

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