Challenges for development and provision of metrological Quality Control tools in nuclear safeguards, nuclear forensics and nuclear security

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Abstract:

The EU has a particular interest in ensuring that the peaceful uses of nuclear energy take place with the highest standards of nuclear safety, security and non proliferation. The control of civil nuclear material world-wide requires harmonized approaches and reliable (measurement) standards and quality control tools available to safeguards authorities. The same standards and quality control tools are used by the nuclear industry to meet safeguard requirements and by laboratories in the field to demonstrate measurement capabilities to accreditation bodies, regulators, customers, also creating public transparency. Confidence in comparability and reliability of measurement results in nuclear material and environmental sample analysis are established via certified reference materials (CRMs), reference measurements, and interlaboratory comparisons. IRMM is one the few institutes that supplies uranium and plutonium isotopic reference materials worldwide to fulfil the existing requirements for reliable and traceable CRMs in fissile material control and environmental sample analysis. Recently, emphasise is put at IRMM on the developments of new and innovative reference materials and methods for detection of undeclared activities and for nuclear forensics in cooperation with the JRC-ITU, the IAEA, DOE National Laboratories and other major partners in the field. Examples of joint advancements in quality control tools and measurement sciences of international reference and safeguards laboratories are:

- Successful integration of the Modified Total Evaporation technique (MTE) as a new tool for routine thermal ionization mass spectrometry in nuclear safeguards and security
- Research and feasibility studies for the development of new material standards, particularly for nuclear forensics (CRMs for age-dating)
- Quality control tools to support the additional protocol and nuclear security (particle CRMs, NUSIMEP)
- Scientific/technical advice, training and knowledge transfer

The European Safeguards Research and Development Association (ESARDA), the Institute of Nuclear Materials Management (INMM) and the Commission d'ÉTABlisement des Méthodes d'Analyse du Commissariat à l'Energie Atomique (CEA/CETAMA) and the International Atomic Energy Agency (IAEA) Technical Meetings are the platforms to exchange views on the needs and challenges for new Quality Control tools for nuclear safeguards and security. The ESARDA Working Group for Techniques and Standards for Destructive Analysis, is organising dedicated workshops on advancements and applicability of analytical techniques to read signatures in nuclear material and environmental samples with participants beyond the safeguards community. Within the WG DA, currently chaired by IRMM, ESARDA is keeping abreast of methodologies applicable in the fields of nuclear safeguards, security and forensics.

Keywords: isotopic reference materials; interlaboratory comparison, quality control, standardisation, non-proliferation, education and training
1. Introduction

After the natural disaster that hit Japan in March 2011, the EU stated in the EUROPEAN COUNCIL conclusion from 24/25 MARCH 2011 that the highest standards for nuclear safety should be implemented and continuously improved. This applies also to nuclear security, as emphasised by the President of the European Council, Herman van Rompuy, on behalf of the EU at the Nuclear Security Summit ‘the EU, with its large multinational nuclear industry, has a particular interest in ensuring that the peaceful uses of nuclear energy take place with the highest standards of nuclear safety, security and non proliferation’ [1]. The triple concept SSS (Safeguards, Security, and Safety) is converging more and more towards the idea of internationally binding security and safety standards. Reliable and comparable measurement results and the respective quality control tools are at the basis of conclusions to be drawn that can have legal and economic consequences.

Quality Control (QC) is a set of procedures, including technical activities, to ensure that a final product or a performed service adheres to a defined set of quality criteria and meets the customer requirements. Quality Assurance (QA) is defined as a systematic process to check whether a service/product under development is meeting specific requirements before the product or service is completed. Quality Assurance (QA) and Quality Control (QC) in sample analysis for nuclear safeguards measurements are a means to the end of complying with the requirements to provide reliable measurement results for the nuclear safeguards system. QA and QC in analytical measurements comprise different aspects [2]:

- Method validation and instrument calibration
- Traceability and comparability of measurement results
- Uncertainty of measurement results
- External performance evaluation
- Document and material standards
- Deployment of a quality system

IRMM as supplier of uranium and plutonium isotopic certified reference materials aims to fulfil the existing requirements for reliable and traceable CRMs in fissile material control and environmental sample analysis. One new objective recently adopted in the Action Plan of the ESARDA WGDA, currently chaired by IRMM, is to emphasise the technical convergence of nuclear safeguards, nuclear forensics and nuclear security by looking at available and new methodologies including respective quality control tools that serve all three purposes [3].

2. Determination of nuclear signatures

One main focus is the development and improvement of methods for the determination of nuclear signatures in environmental and “special” samples. Isotopic “fingerprinting” is needed for attribution of intercepted materials and for verification of the correctness and completeness of a State’s declarations, particularly in view of the detection of any undeclared material or activities. Recently, under the initiative and coordination of IRMM, four key nuclear mass spectrometry laboratories (IRMM, ITU, IAEA-SGAS and DOE-NBL) published an article on the development of the Modified Total Evaporation technique (MTE) applied for sample analysis in nuclear safeguards, nuclear forensics and other disciplines like geo-and cosmo-chemistry [4]. MTE is a new tool for thermal ionization mass spectrometry in nuclear safeguards and geochemistry developed as a method for accurate measurements in particular for minor isotope ratios of uranium in nuclear material. The MTE method provides a measurement performance which is superior to the present IAEA requirements, enabling more detailed conclusions from source data for source attribution of samples. The performance of the MTE method for the minor uranium ratios \(\frac{n(234U)}{n(238U)}\) and \(\frac{n(236U)}{n(238U)}\) can be seen in Figure 1. In a harmonised approach the MTE method was successfully integrated for routine use at all contributing laboratories and has subsequently been applied on measurements of samples in safeguards and forensics. This is an outstanding example of joint advancements in measurement sciences of international reference and safeguards laboratories that exchange expertise via the ESARDA WGDA platform.
3. Pu certified reference materials for “age dating”

Although the civil nuclear fuel cycle is subject to strict regulatory control schemes, nuclear material (uranium, plutonium) is occasionally discovered outside this regulatory control system. In these cases nuclear forensic investigations are applied in addition to traditional forensics in order to provide clues on the history of the material and to possibly identify the last legal owner of the material. The isotopic and elemental composition, the physical appearance of the material (dimensions of larger objects; particle form and size of powders) and its “age” are determined. The “age” of nuclear material is defined as the time that has passed since the last chemical separation of the daughter nuclides from the mother nuclides (e.g. in-grown uranium and americium from plutonium). For the determination of the age of a plutonium material different pairs of mother and daughter radio-nuclides can be used: $^{241}\text{Pu}/^{241}\text{Am}$, $^{238}\text{Pu}/^{236}\text{U}$, $^{239}\text{Pu}/^{235}\text{U}$, $^{240}\text{Pu}/^{236}\text{U}$, and possibly $^{242}\text{Pu}/^{238}\text{U}$. IRMM is cooperating with the Institute for Transuranium Elements (EC-JRC-ITU) in a feasibility study on the development of Pu reference materials for “age dating” in nuclear forensics [5]. In the course of this work the reference materials NBS SRM 946, 947 and 948 (NBL CRM 136, 137 and 138) will be investigated among others. As a spin off: additional results on isotope ratios for these materials will be available to the community as part of the respective action sheet NBL-IRMM.

4. Pu – “fingerprints” in environmental samples

Advancements in low-level analytical techniques serve nuclear safeguards, nuclear forensics and nuclear security purposes. An example is the successful determination of plutonium isotope ratios in environmental samples from different geographic origins and from Chernobyl, through application of applying state of the art Thermal Ionization Mass Spectrometry (TIMS) in combination with multiple ion counting (MIC) and filament carburization [6]. Due to this developed procedure, data were acquired with lower uncertainties not only for the $^{240}\text{Pu}/^{239}\text{Pu}$ major isotope ratio but also for the $^{241}\text{Pu}/^{239}\text{Pu}$ and $^{242}\text{Pu}/^{239}\text{Pu}$ minor isotope ratios, which is particularly important for fingerprinting of environmental samples. This information reveals different sources of plutonium contamination in the environment and can help to distinguish if the observed contamination in collected samples originates from global fallout or from an accident like in Chernobyl. This technique could also be applied to environmental samples collected around the Fukushima site. Soils samples from different geographic origin would be very promising candidate reference materials for certified for Pu isotope ratios useful in nuclear safeguards and in earth science applications.

Figure 1 Performance of the Modified Total Evaporation method for minor uranium ratios $n(^{234}\text{U})/n(^{238}\text{U})$ and $n(^{236}\text{U})/n(^{238}\text{U})$ [Erreur ! Signet non défini.]
5. Isotopic “fingerprinting” of U-particles

Information accessible via isotopic “fingerprinting” of uranium particles can provide additional information on undeclared nuclear material or activities, on attribution of intercepted materials, and on response to theft, illegal transfer of nuclear materials. IRMM set up cooperation with ITU, with CEA/DAM and DOE-LLNL (action sheet) for development, characterisation and production of uranium reference particles. High-quality isotopic data on single U-reference particles were obtained using an improved thermal ionisation mass spectrometry method based on in situ SEM micromanipulation, filament carburization and MIC detection [7]. The improved method has been successfully applied for detection of U-signatures in real-life particles collected at a nuclear facility [8]. Currently research is ongoing towards production of U-oxide reference particles certified for isotopic abundances and U amount content per particle to be used for evaluating sensitivity in MS techniques (SIMS, TIMS, LA-ICP-MS).

6. NUSIMEP-7 Interlaboratory Comparisons for U particle analysis

The REIMEP - Regular European Interlaboratory Measurement Evaluation Programme REIMEP was started by IRMM in 1982 for carrying out external control of the quality of the measurements of the nuclear fuel cycle materials. REIMEP samples are matching materials analysed routinely in the nuclear industry and controlled by safeguards authorities (UF6, synthetic input solution, synthetic MOX fuel solution, U and Pu nitrate). The Nuclear Signatures Interlaboratory Measurement Evaluation Programme (NUSIMEP) was established in 1996 to support the growing need to trace and measure the isotopic abundances of elements characteristic for the nuclear fuel cycle present in trace amounts in the environment. REIMEP and NUSIMEP are external Quality Control tools for laboratories from nuclear safeguards, nuclear industry but also from the environmental, geochemistry field and academia. The laboratories receive well-characterised samples with undisclosed values. They can compare their results with an independent reference value, independent of the participants’ results. Laboratory performance evaluation is according to ISO guidelines on performance evaluation in proficiency testing by interlaboratory comparison.

The most recent ILC is NUSIMEP-7 organised by IRMM for laboratories dealing with uranium particle analysis, in particular for the IAEA network of analytical laboratories (NWAL) for environmental sampling to further improve their detection and analysis capability. Participating laboratories in NUSIMEP-7 received two test samples of uranium particles on a graphite planchet with undisclosed isotope amount ratio values \( n^{(234)}U/n^{(238)}U \), \( n^{(235)}U/n^{(238)}U \) and \( n^{(236)}U/n^{(238)}U \). One sample had a single isotopic deposition and the other sample consisted of particles with two different isotopic compositions. For both samples, the uranium isotope amount ratios had to be measured by participating laboratories on a prescribed number of particles using their routine analytical procedures. Performance criteria have been set according to safeguards requirements. Preliminary results are presented in this paper since the final report is still in preparation. It will be accessible via the IRMM web-site [9].
. Figure 2 NUSIMEP-7 participants results for \( \frac{n(^{235}U)}{n(^{238}U)} \) single deposition – preliminary graph

7. Exchange with experts in the field

Needs for Quality Control tools and development of (reference methods) are discussed with safeguards authorities, reference materials producer and expert laboratories in consultant / expert group meetings using different platforms:

- Exchange of expertise using the ESARDA platform
- Exchange of expertise using the INMM annual meetings (NBL-SME)
- Exchange of expertise using the CETAMA WGs
- IAEA Technical Meetings:
  - Particle Analysis of Environmental Samples for Safeguards
  - Bulk Analysis of Environmental Samples for Safeguards
  - Nuclear Reference Materials for Safeguards Verification Measurements by Destructive Analysis (DA)

Particularly, the WG DA organises dedicated workshops on advancement and applicability of analytical techniques to read signatures in nuclear material and environmental samples, providing a platform for exchange of expertise from safeguards, nuclear forensics, nuclear security, earth sciences, geochemistry and industry. The reports of all those dedicated workshops are published in the ESARDA Bulletin
7. Training & Education on QC tools

Chapter I of the EURATOM treaty concerning the promotion of research and the European Council conclusion on *the need for skills in the nuclear field* set out the legal framework for putting efforts in maintaining and transferring the expertise in the nuclear field from one generation of engineers and researchers to the next [12]. The WG DA strongly supports ESARDA in fulfilling an educational role in the nuclear field and also in reaching the general public. Technical sheets on analytical techniques, reference materials and quality control tools are accessible to the public via the ESARDA library website [13]. The important role of QC tools is part of the module on destructive analysis regularly presented at the ESARDA academic course on Nuclear Safeguards and Non-Proliferation at on BNEN advanced course on safeguards in Belgium (SCK, IRMM). IRMM regularly provides a training course on the *Use of Reference Materials and the Estimation of Measurement Uncertainty*. Since this course provides participants with the theoretical basis for the estimation of measurement uncertainty, establishment of traceability, and the proper selection and use of reference materials it is applicable to all measurement fields. The aim is to use reference materials to achieve true traceability of measurements, proving accuracy of methods and demonstrating proficiency of laboratories. The course strongly emphasises practical application of the theoretical concepts presented during the lectures. Dedicated exercises are undertaken by the participants in small groups with support from a trainer. Although the course layout was originally set for laboratory managers, practitioners in analytical laboratories and ISO 17025 auditors from the non-nuclear field, it is now also very much appreciated by nuclear inspectors from Euratom Safeguards coming from the field of non-destructive analysis. [14].
8. Conclusions

The European Union (EU) has affirmed that it will support international cooperation on technological infrastructure and networks necessary to verify the non-diversion of declared nuclear material but also the absence of illicit nuclear material and activities. Nuclear safeguards is about the complete, correct and comprehensive picture of a State's or a region's nuclear activities; nuclear forensics about the identification of the origin and intended use disclosing additional information inherent to nuclear material; and nuclear security about prevention/detection/response of theft, sabotage, unauthorized access, illegal transfer of nuclear/radioactive materials and associated facilities. Verification, detection, conformity and response are based on reliable measurement results with appropriate quality control tools as prerequisite and on advanced measurement techniques and instrumentation [3]. Therefore emphasise needs to be put on Quality Control tools that serves the technical convergence of nuclear safeguards, nuclear forensics, and nuclear security (e.g. CRMs and ILCs for bulk and particle analysis of environmental samples and age dating of nuclear material). The exchange of expertise using the ESARDA, INMM, CETAMA and IAEA working groups and technical meetings as platforms is a prerequisite for reference materials producers such as IRMM to meet the needs of the measurement community for new and tailor-made quality control tools. Training and education in the correct use of reference methods, reference materials and in the benefit of external quality control tools are of major importance to researchers, engineers, scientists, and (young) professionals from all three fields. The EU envisages supporting the ongoing efforts to strengthen IAEA's analytical capabilities with a contribution from the Instrument for Stability (IfS) to the expansion and modernisation of the IAEA Safeguards Analytical Laboratories (SAL) under the project of “Enhancing Capabilities of the Safeguards Analytical Services” (ECAS). On requests of EuropeAid Development and Cooperation (DG DEVCO) and the European External Action Service (EEAS) the JRC provides via the EC-SP technical/scientific advice for the EU donation for the new IAEA Nuclear Material Laboratory [15].

Table 1 attempts to give an overview on how nuclear safeguards, nuclear forensics and nuclear security are linked on the technical level and the main role QC tools have in all three fields [3].
Table 1: Technical convergence of nuclear safeguards, nuclear forensics and nuclear security

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<th>Scientific Disciplines: Chemistry, Physics, Material science</th>
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<td><strong>Analytical and technical tools</strong></td>
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<td>Verification of non-diversion</td>
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<td>Environmental sample analysis</td>
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<td>Detection of undeclared activities</td>
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<td>Seized/collected material analysis</td>
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<td>Consistency, Coherence, Conformity of information, materials and processes</td>
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<td>Metrological quality control tools</td>
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<td>Method development, method validation, QC/QA</td>
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<td>Other sources of information</td>
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| Amount content, isotopes                                      |
| Bulk and particle analysis; isotopic fingerprint               |
| Bulk and particle analysis; isotopic fingerprint; anionic and metallic impurities, microstructure, "age" – last separation date |
| Certified Reference Materials (CRMs)                          |
| Interlaboratory Comparisons (ILCs)                            |
| Close cooperation with data analysts, police, governments,… |

**Nuclear Safeguards**
Complete, correct and comprehensive picture of a State’s nuclear activities

**Nuclear Forensics**
Identifying origin and intended use information inherent to the (nuclear) material

**Nuclear Security**
Prevention/Detection/Response of theft, sabotage, unauthorized access, illegal transfer of nuclear/radioactive materials and associated facilities

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<th>Nuclear Safeguards</th>
<th>Nuclear Forensics</th>
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5. Acknowledgements
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6. References


[12] European Council Council conclusions on the need for skills in the nuclear field 15406/08; 16577/08

