Nuclear Material Measurements at ABACC: Status Update and Future Steps

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Abstract:

The Brazilian-Argentine Agency for Accounting and Control of Nuclear Material (ABACC) is a bilateral organization created in 1991 by Argentina and Brazil to verify the peaceful use of nuclear materials and installations in both countries. Among several responsibilities, ABACC has to perform quantitative verification of the declared nuclear material inventories and transactions by independent measurements that must meet internationally recognized quality levels.

In this paper we present and discuss some of the most relevant measurement results obtained by ABACC inspectors during on-site activities, as well as data resulting from destructive analysis of samples performed by the network of Argentinean and Brazilian analytical laboratories that support ABACC. Data obtained during recent past years are compiled and evaluated according applicable international standards.

Finally, we discuss the future actions that will be essential for ABACC to maintain the capacity to accomplish its mission, in accordance with the agreement established by Argentina and Brazil.

Keywords: nuclear safeguards, non-destructive and destructive analysis, measurements systems.

1. Introduction

Argentina and Brazil decided to establish a common system of accounting and control of all nuclear materials (SCCC) and create ABACC in 1991, as an independent organization responsible for verifying the appropriate implementation of the SCCC in both countries [1]. The countries took another decision, in the scope of the international safeguards regime implemented by the IAEA, with the decision to move from separate and limited bilateral safeguards agreements to a multilateral comprehensive safeguards agreement fully consistent with the international non-proliferation regime [2].

In order to avoid unnecessary duplication of efforts, most of the verification activities are jointly performed by ABACC and IAEA inspectors, but the agencies draw independent conclusions. In most inspections, measurements of uranium are jointly performed by the inspectors using non-destructive systems. The agencies establish joint use procedures and ensure that appropriate measurement capabilities are always available. On the other hand, since destructive analysis techniques are not yet submitted to this scheme, samples are collected in duplicate during inspections for independent analysis at laboratories that provide analytical support to ABACC or the IAEA.

Data obtained during recent past years are compiled and evaluated according applicable international standards [3] and statistical historical values.

2. Non-destructive measurements

Non-destructive (NDA) gamma spectroscopy technique to determine 235U fraction or enrichment can be used with low, medium and high-resolution systems and is capable of providing results on the field after a few minutes of spectrum collection. This feature is very useful for applications where results must be promptly available or sampling for destructive analysis (DA) at a laboratory is unpractical or impossible. On the other hand, the errors associated with NDA are usually higher than the ones achievable with DA techniques. As result, a typical safeguards approach for bulk facilities considers a combination of both NDA and DA measurements for verification of different strata.

There are relevant bulk facilities both in Argentina and Brazil: commercial and R&D fuel fabrication, conversion and enrichment. During inspections at these facilities, depleted (DU), natural (NU) and low enriched (LEU) non-irradiated uranium items are measured by inspectors using portable NDA systems under a joint-use scheme. Currently, typical NDA systems for enrichment measurements are composed by a portable mini multichannel analyzer, a medium resolution Lanthanum Bromide scintillation gamma radiation detector (LaBr3(Ce) - typical size 1.5 x 1.5 in.) or a high resolution and high-purity Germanium detector (HPGe - typical relative efficiency is 15% and FWHM at 122 keV < 600 eV). The use of low resolution detectors (NaI(Tl)) was discontinued in 2016. The software NaIGEM version 2.4 is used for analysis of LaBr3(Ce) spectra and MGAU version 3.2 for HPGe spectra.

Typical uranium items are: UF_6 cylinders, drums containing U_3O_8 or UO_2 and UO_2 pellets in fuel rods. The HPGe detector is selected for UF_6, while $LaBr_3$(Ce) is used for the other items. Counting time (live) is typically five minutes. A portable digital ultrasonic thickness gauge is also used to determine wall thickness with 0.1 mm resolution for attenuation correction. Calibrations are performed with one or more certified standards selected from the set identified as NBS-SRM-969 [4]. In the case of fuel rods, a set of reference rods is available for calibration. The measurement geometry is kept fairly constant using lead collimation and appropriate detector positioning.

Results obtained are shown in Figures 1 and 2, where the relative deviations between declared and measured values are plotted. Since declared values for LEU UF_6 cylinders are obtained with DA techniques and for NU the nominal value is used, the uncertainties on the declared values are assumed to be negligible when NDA techniques are used for verification.

Figure 1 shows results of enrichment measurements for NU powders using medium resolution systems. Because there is not yet an established ITV for this technique, the dashed lines indicating a possible acceptance interval (±3*ITV) are based on the ITV for low resolution.

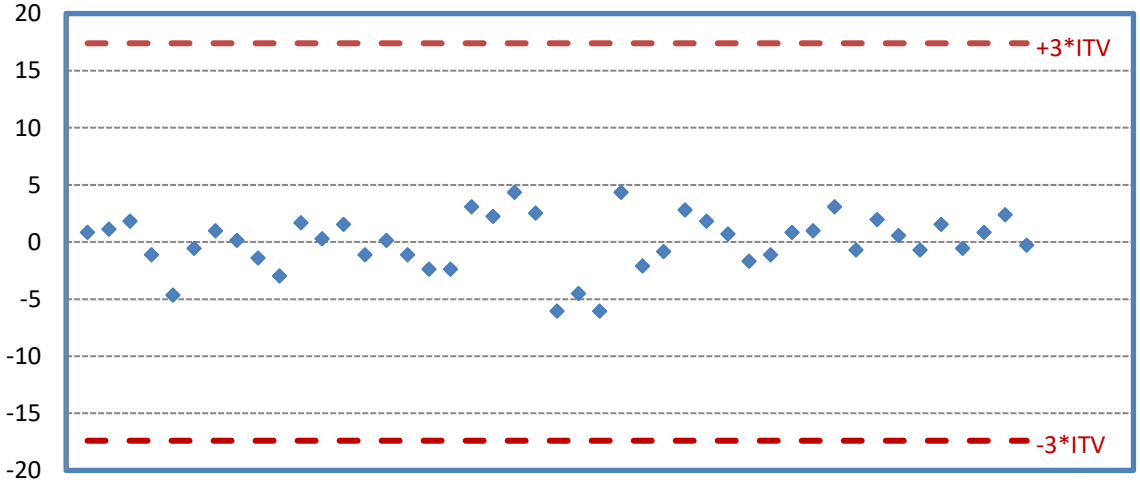

Figure 1: Declared to Measured % deviation in enrichment measurements of NU powders with medium resolution gamma spectrometry.

The relative standard deviation of the data plotted in Fig. 1 is 2.4%, which is significantly lower than the ITV for low resolution (5.8%). This indicates that medium resolution systems have been able to perform much better than low resolution and then a specific ITV should be available.

Results obtained from verification of LEU UF_6 cylinders are plotted in Figure 2.

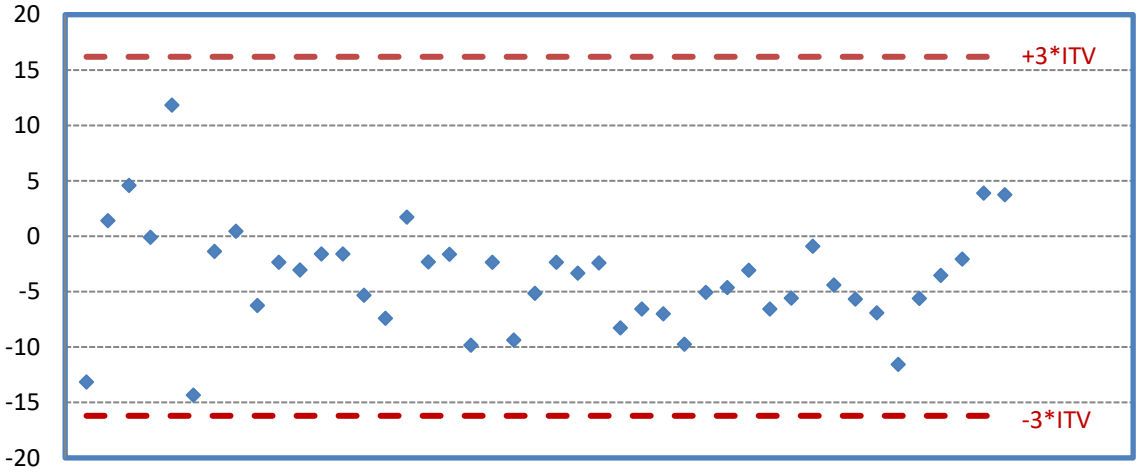

Figure 2: Declared to Measured % deviation in enrichment measurements of LEU UF6 cylinders with high resolution gamma spectrometry.

The relative standard deviation of the data plotted in Fig. 2 is 4.9%, which is consistent with the corresponding ITV (5.4%). Although all data points are well within the indicated limits, the plot indicates a negative bias of about 3.7%. Since most data points refer to measurements of 30B cylinders, such a bias may be due to inaccurate correction of the attenuation caused the thick and dense wall of the cylinder (0.5 in. carbon steel). Improvements on it could be implemented for example by minimizing wall attenuation variations between calibration and item measurements, i.e. material type and thickness.

Low enriched fresh fuel assemblies produced at two commercial fuel fabrication plants are verified by active and passive neutron measurements using the uranium neutron coincidence collar (UNCL). A simple hand-held gamma monitor is used to determine the active length of the fuel assembly, which is used as one of the input values for the analysis software. Figure 3 shows results jointly obtained by ABACC and the IAEA during the last five years.

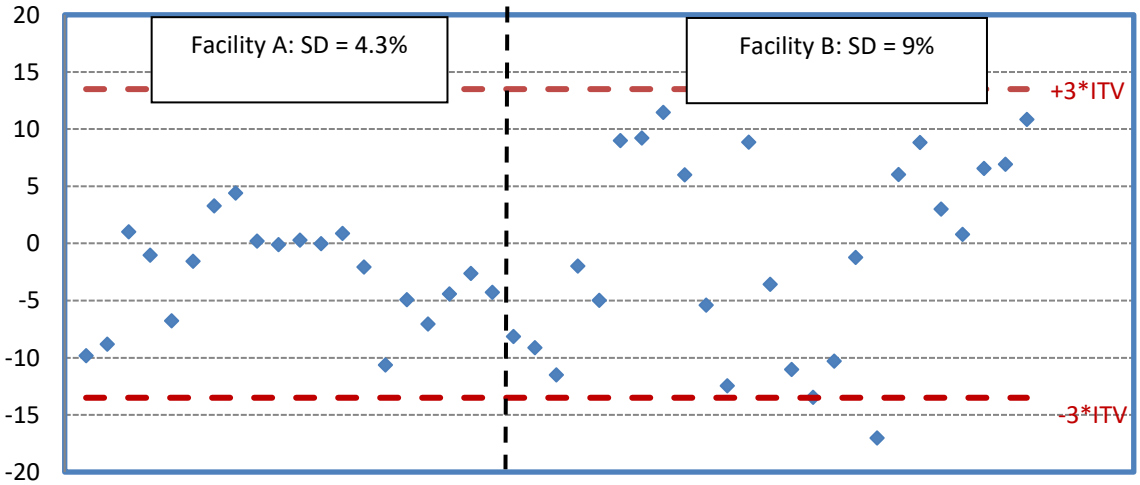

Figure 3: Declared to Measured % deviation in 235U mass content resulting from verification of fresh fuel with the UNCL system.

At facility A (left side of Figure 3), the measured fuel assemblies are all of the same design, same enrichment and without burnable poison. The observed RSD is 4.3%, which is in good agreement with the corresponding ITV (4.5%). At facility B (right side), fuel assemblies of different designs are measured: geometry, enrichment and burnable poison content. This explains why the observed RSD (9%) is significantly higher. Although in both cases the results are well within ±3*ITV (only one is out), the enlarged data variation for this facility indicates that the measurement technique is very sensitive to the fuel design. It should be noted that the currently available ITV does not consider the influence of burnable poison.

3. Destructive measurements

Destructive assay techniques can provide improved measurement accuracy and precision in comparison with NDA. They may be used for the evaluation of other NDA or DA measurement techniques used by facility operators or detection of small discrepancies on declared values. Differently from NDA, uranium samples are always taken by ABACC and IAEA inspectors in duplicate, allowing for independent elemental and/or isotopic analysis. Due to the higher strategic value, samples are more frequently taken from LEU materials than DU and NU. The number of samples is limited to the minimum necessary due to the high associated cost and large time associated with the process: sample collection at the facility, international transportation arrangements, sample treatment and analysis, and final issue of the analysis report. The process may take approximately 3 months to be completed. In case of UF_6 samples, this time can be longer due to more severe transportation requirements.

The uranium materials submitted to sampling are typically the same as for NDA. Pure nitrate solutions found in NU conversion plants is one of the exceptions, but with reduced sampling frequency. The analysis techniques used by the laboratories are Davies & Gray titration for element fraction and thermal ionization mass spectrometry (TIMS) for isotopic analysis. Traceable uranium reference materials are used for quality control and calibration of the analytical instruments.

Results obtained during the last five years are shown in the next Figures. Figure 4 shows the relative deviations between declared and measured uranium concentration values for powders. The dashed lines indicate three times the combined ITV resulting from propagation of sampling and D&G measurements performed by both operator and ABACC. The plot indicates good consistence between actual performance (RSD = 0.39%) and the propagated ITV (0.35%).

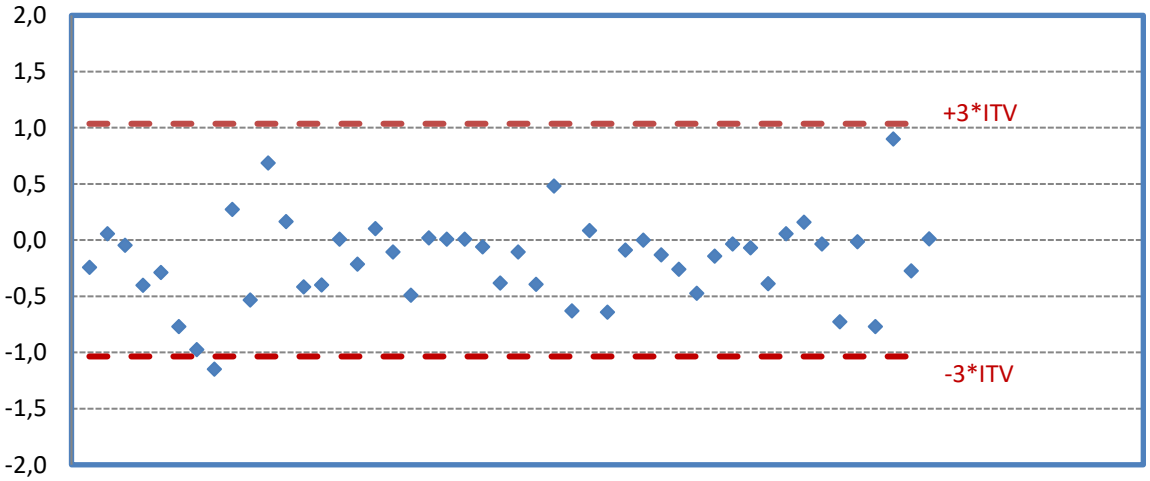

Figure 4: Declared to Measured % deviation in uranium concentration measurements of powders by D&G Titration.

In regards to UO_2 sintered pellets, as shown in Figure 5, actual RSD is 0.11%. This is because pellets are more stable and less susceptible to the presence of undesirable components (i.e. humidity) than powders.

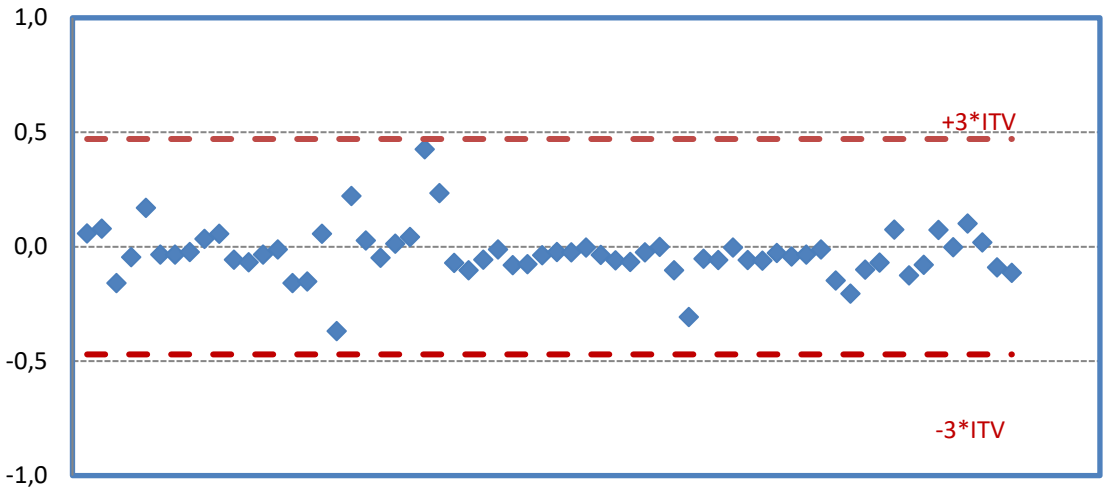

Figure 5: Declared to Measured % deviation in uranium concentration measurements of UO2 pellets by D&G Titration.

Determination of 235U/238U ratio is performed by the laboratories using TIMS for all samples. For NU and LEU materials with enrichment below 1%, as shown in Figure 6 and 7, the actual performance data is quite consistent with the propagated ITV (0.41%) considering two independent measurements.

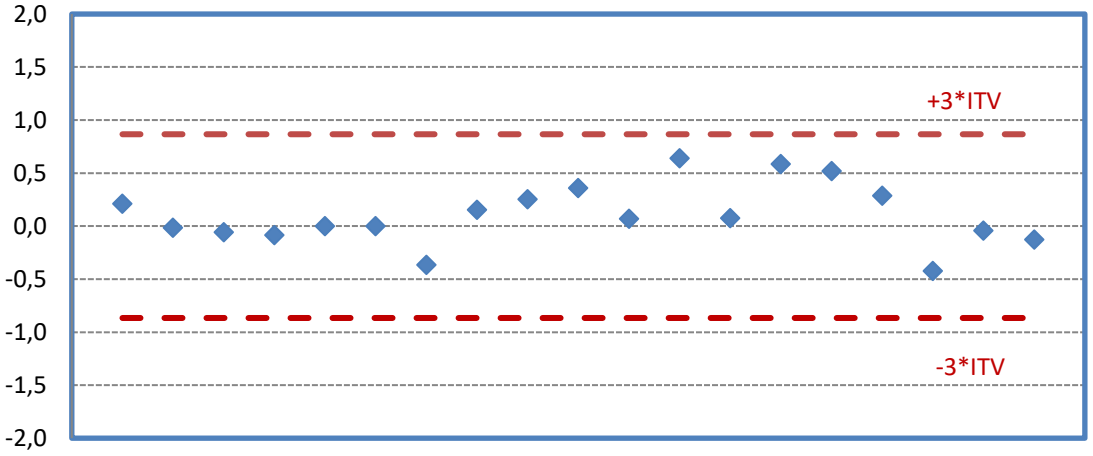

Figure 6: Declared to Measured % deviation in 235U/238U ratio of natural UO2 pellets by TIMS

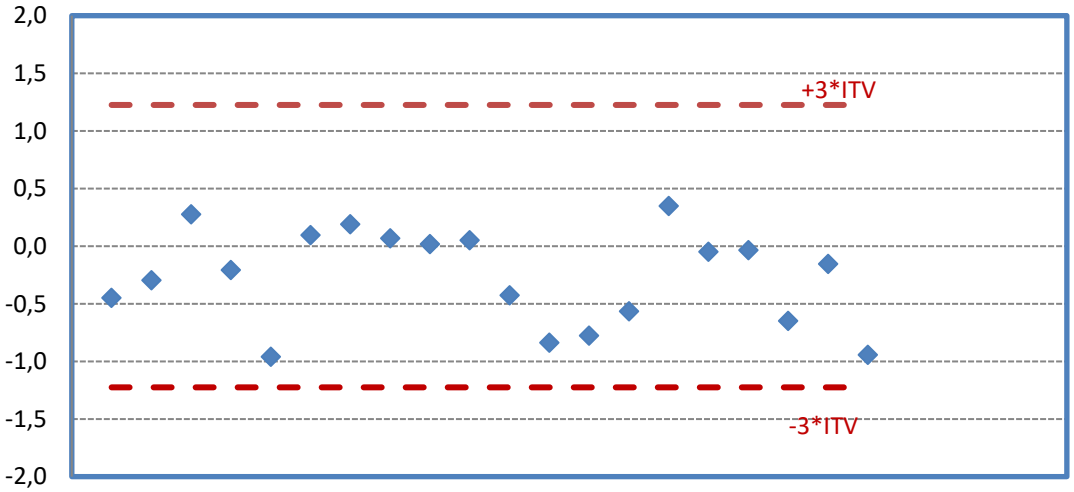

Figure 7: Declared to Measured % deviation in 235U/238U ratio of LEU (< 1%) materials by TIMS

However, for LEU samples enriched above 1%, as presented in the Figure 8, the actual performance (RSD = 0.46%) is affected by important sampling uncertainties associated with sampling of gaseous UF6 from feed and withdraw lines at enrichment plants.

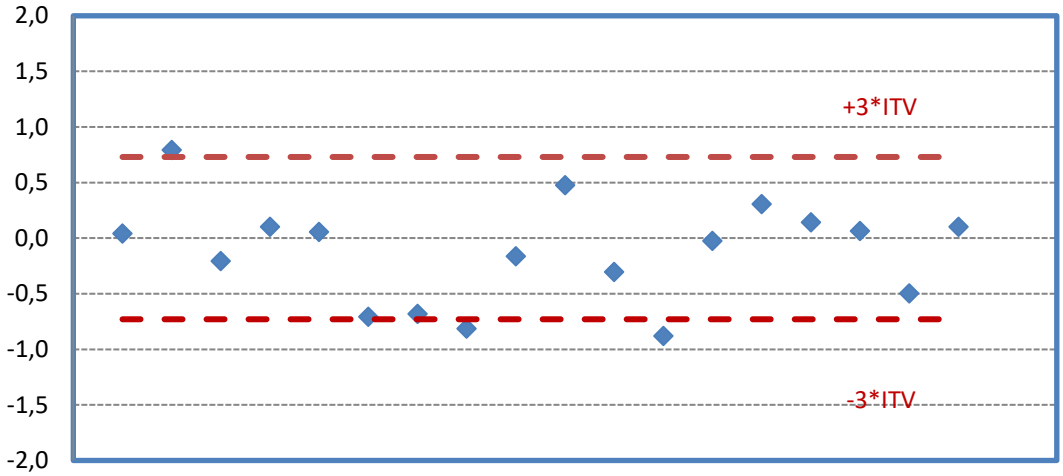

Figure 8: Declared to Measured % deviation in 235U/238U ratio of LEU (> 1%) materials by TIMS

4. Measurement Evaluation Programs

The routine assessment of the quality of the analytical measurements is very important for the laboratories, in particular when a formal QA/QC program is in place. In this context, ABACC supports the participation of the laboratories of its analytical network in safeguards Measurement Evaluation Programs (MEP) promoted by internationally recognized organizations such as the New Brunswick Program Office (NBL) in the USA and the Institute for Reference Materials and Measurements (IRMM) in Europe.

ABACC coordinated the participation of its network laboratories in the Nuclear Material Round Robin (NMRoRo) promoted by the IAEA as a MEP in 2017, which offered the following sample for analysis: one gram-sized low enriched UO2 pellets for determination of atom ratios (n235/n238, n234/n238 and n236/n238) and/or uranium mass fraction. The following laboratories from Argentina and Brazil participated in this MEP:

- Development of Uranium Compounds Laboratory (LADCU/CNEA – Argentina)
- Physical and Chemical Control Laboratory (CNEA – Argentina)
- Mass Spectrometry Laboratory (LEM/CNEA – Argentina)
- Uranium Characterization Laboratory (LCU/CTMSP – Brazil)
- Safeguards Laboratory (LASAL/CNEN – Brazil)

ITV values were also used as reference in establishing acceptance levels.

Uranium mass fraction: four laboratories (two from each country) provided results obtained by Davies & Gray titration. All reported results were within expected performance levels.
Uranium atom ratios: two laboratories (one from each country) provided results obtained by total evaporation thermal ionization mass spectrometry (TE-TIMS). All n235/n238 reported results were consistent with the expected performance levels.
Although minor isotope ratios n234/n238 and n236/n238 have no available ITV, reported results were consistent with the corresponding reference values at 95% (two-sigma) confidence level.

5. Conclusions

The performance for the most relevant NDA and DA measurements routinely performed by ABACC has been summarized. In the area of NDA, the full migration from low to medium resolution gamma spectrometry systems based on LaBr3(Ce) scintillation detectors represented a remarkable step for improved enrichment measurements of powders in particular. The need for a specific ITV for this technique seems to be evident and the experience of ABACC may be useful

in this regard. Enrichment measurements of UF6 cylinders by high resolution gamma spectrometry is still a valuable tool, but special attention to the calibration procedure is required since the size and wall thickness of the measured cylinders may vary significantly.

Total 235U mass measurements in modern PWR fresh fuel assemblies using the UNCL system may be subject to enlarged uncertainties, in particular for fuel designs that include different enrichment layers in the same fuel rod and/or gadolinium as burnable poison. These additional uncertainties are probably of systematic nature, caused by limitations on the calibration procedure. As an attempt to mitigate those limitations, advanced neutron counting systems and calibration techniques are under development at the international level. In this regard, ABACC, Brazil and the IAEA are working together to test a new NDA system based on fast neutron coincidence counting. Since there is currently a single ITV available for fresh fuel measurements, a future revision should consider the possibility to differentiate uniform and single enrichment from complex fuel designs.

In regards to DA analysis of the samples collected during inspections by D&G titration for uranium concentration determination, the operator versus ABACC data comparison has indicated good agreement with international practices. As for enrichment measurements, uncertainties associated with sampling of powders and gaseous UF6 may be relevant for the evaluation process.

The performance achieved in the 2017-NMRORO program indicates that laboratories currently supporting ABACC are performing reliable uranium mass fraction and isotopic determinations. ABACC continues to support the participation of the laboratories in different MEPs that are currently being promoted by the IAEA and NBL during the biennium 2018/2019.

6. References

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[4] P. Matussek, *Accurate Determination of the ^{235}U Isotope Abundance by Gamma Spectrometry*, KfK 3752 (1985).