Large-Volume Cadmium Zinc Telluride Modules for Safeguards Verification of Unirradiated Nuclear Material

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1 Abstract

The main techniques to date for the non-destructive assay (NDA) verification of fresh nuclear material to support IAEA safeguards verification requirements are primarily based on various high-, medium- and low-resolution gamma spectrometry systems (e.g., high-purity germanium, lanthanum bromide, sodium iodide). The IAEA's objective is to reduce the miscellany of systems and to provide inspectors with spectrometric tools based on advanced technologies capable of covering multiple verification contexts, while offering a user-friendly interface requiring only limited training.

The Department of Safeguards developed user requirements for a compact instrument operable at room temperature and capable of reaching uncertainty performance targets within a measurement time optimized by a combination of high detection efficiency and superior energy resolution. Thus far, the M400 CZT modules offered by H3D, Inc. (USA) have been the only technical solution able to meet such demanding requirements.

After a thorough evaluation of all the required characteristics and performance of the CZT modules, with emphasis on the stability of their detection efficiency over a large temperature range, the IAEA is introducing the MCA-Touch-based CZT Module (MCCM) as its primary solution for NDA verification of unirradiated nuclear material. The MCCM is a gamma spectrometry system based on the M400 CZT and operated by the MCAT acquisition software developed by GBS Elektronik (Germany) hosted on a miniature PC. Including its specially developed tungsten collimator and external battery, the MCCM weighs less than 2 kg and represents a highly portable instrument providing inspectors with a dramatically improved usability experience compared to current systems based on electrically cooled germanium or lanthanum bromide detectors.

This paper reports on MCCM system performance and the initial efforts to achieve readiness for deployment to inspectors for field use of this new instrument. It details the process that the IAEA undertook to introduce the system, from the initial determination of the requirements, through assessment and verification of actual instrument performance, demonstration of suitability for envisaged safeguards verification methods, and other activities related to its integration into the IAEA safeguards instrumentation environment.

2 Introduction

At present, various instruments based on high-, medium- and low-resolution gamma spectrometry systems (see [1]) are required to address different fresh fuel verification contexts and constraints. This requires the IAEA to maintain several pools of equipment in an operational status, including all their concomitant maintenance, sourcing and training requirements.

In recent years, significant progress has been made in the area of large-volume, medium-resolution detectors operated at room temperature, whose performance suggests that a single instrument may be able to cover a larger range of safeguards applications. As a result, the IAEA's Department of Safeguards has undertaken an initiative to identify possible candidates for the next generation of instrumentation that could be integrated into existing non-destructive assay methods. Advances in the core technology initially developed and evaluated for gamma imaging applications were the starting point for evolving to purely spectrometric applications. Called the MCCM (MCAT-based CZT detector module), the new instrument is intended as the primary tool of inspectors in the field for measurements of unirradiated nuclear material, meaning that a particular focus during its development was on pragmatic considerations such as compact size and light weight, reduced power consumption, fast start-up and reduced training requirements.

The main application foreseen at this stage for the MCCM is the determination of U-235 enrichment level by quantitative spectrometry based on absolute calibration (the enrichment meter principle [2]) for material in different forms (e.g., powders, UF_6, pellets, rods etc.). Other applications include the attribute test and (potentially) determination of U-235 enrichment by the peak-ratio technique and quantitative assay based on gamma spectrometry (through spectrum analysis using computed photopeak detection efficiencies).

3 System Description

The MCCM is a gamma spectrometry system based on the M400 cadmium zinc telluride (CZT) detector from H3D, Inc. (USA), powered by a portable battery pack and operated by the MCAT acquisition software developed by GBS Elektronik (Germany) hosted on a miniature PC. Application-specific mechanical components (such as a collimator or holders to ensure reproducible geometry and minimizing biases for measurements of UF_6 cylinder, pellets, rods) were designed[1] to address the needs of particular verification applications (Figure 1).

Figure 1. MCCM system in various measurement configurations: container/cylinder (left and top center), bulk material (bottom center), pellets (top right), fuel rod (bottom right).

The M400 CZT module (Figure 2) is the core of the MCCM system and comprised mainly of a CZT detector with four crystals, each with active area of about 2 cm x 2 cm and crystal depth of 1cm (a version with a depth of 1.5-cm depth also was tested); internal electronics (ASIC/FPGA) and single board computer. The M400 CZT module has external dimensions of about 10.4 cm (L) x 5.7 cm (W) x 5.7 cm (D), and a weight of approximately 550 g. The instrument functions without temperature-control components (Peltier cooler and/or fan).

The technology is based on pixelated anode arrays and depth sensing technique [3], making it possible to determine the 3D position of each interaction within the detector crystal. Precise spatial characterization of each pixelated anode (array of 11x11 pixels per crystal, as described in [4]) performed in the manufacturer's factory allows specific properties (e.g., charge collection non-uniformities) to be compensated locally and the response of each individual region to be calibrated. This ultimately significantly improves the overall performance of the detector, by obtaining a good energy resolution and increased efficiency due to the large crystal volume. By processing the signals induced on pixels that neighbour a charge-collecting pixel, the position in the anode plane is determined to the level of subpixel resolution, while the depth position is determined by the relative contribution of the anode and cathode signals.

Figure 2. Photographs of the M400 module.

Running on a commercial off-the-shelf miniature laptop with Windows 10, MCAT (MCA Touch, whose main interface is visible on miniature PC screens shown in Figure 1) is the standard data acquisition software used by IAEA inspectors [5][6]. Developed for ease of use and robust operation in field conditions, its modular design facilitates integration of the requisite data analysis packages. Under the auspices of the German support program, the standard MCAT application was extended to add a data acquisition interface to the H3D M400 detector and algorithms are being updated to address the particular response characteristics of the detector in gamma enrichment measurement (GEM) analysis methods [7].

4 Performance requirements and evaluation

The initial steps prior to the introduction of the new MCCM instrument consisted of specifying and evaluating performance of the core system component, the CZT module. Specifications and other associated requirements were developed for a general-purpose spectrometer, taking into consideration the intended applications (from whence performance parameters were derived), user needs and the state-of-the-art technologies available at that time. The aim was to cover as large a number of uses as possible, with a focus on the major applications and deployment contexts, thus, specific emphasis was placed on performance in the 185-keV region of interest (ROI) of the uranium gamma spectrum (efficiency, resolution, stability), usability (reduced form factor and weight, short start-up time, possibility to adapt to standard user interface, absence of need for external cooling), versatility (for potential integration into different NDA systems) and particular design features (low power consumption, absence of a fan).

The next sections describe the evaluation results for the basic spectrometric parameters of the core M400 component with the 10-mm-deep version of the CZT crystal (unless otherwise stated) and for the application-specific performance of the complete MCCM system.

4.1 Basic spectrometric and general performance of the M400 module

During the development process, M400 component performance was extensively tested, characterized and optimized through standard and customized testing procedures, as well as a series of iterative design evaluations and improvements. Five CZT modules of the latest version available at the time were used for testing; excerpts of the test results for key parameters are provided herein.

Energy resolution was evaluated at a range of energies covering most of the required range of 50 keV to 3 MeV. Measured FWHM[2] is shown in Figure 3; its typical value at 185 keV is about 3.2 keV. For reference, typical FWHM values at 185 keV for the lanthanum bromide (LaBr) and electrically cooled HPGe detectors (ECGS) that are currently in use by the IAEA are on the order of 10 keV and 1.2 keV, respectively. FWHM dependence on energy was found to be approximately linear up to 1 MeV (Figure 3); above 1 MeV it can be described by a power law. Peak shape for CZT detectors is known to deviate significantly from a theoretical Gaussian shape; in addition, the technology applied in H3D’s spectrometric instruments (e.g., precise characterization of each pixelated anode, combination of the spectra from the four individual CZT crystals included in the module) is different from that of conventional CZT detectors, resulting in a singular overall response. Peak shape was characterized empirically mostly through visual examination, extraction of the FWTM[3]/FWHM ratio and application of various peak models. In general, the peak shape can be approximated as being comprised of a Gaussian component; low and high energy tails stemming from non-uniformities in the charge collection (in particular, lower performance closer to the anode); and particular features ascribable to individual pixel calibration, which are to some extent detector-specific. Measured FWTM/FWHM values also are shown in Figure 3. Visual comparison of the peak shape for some uranium and arbitrary Cs-137 measurements taken with a few of the M400 modules with LaBr and ECGS detectors are shown in Figure 4. Deviation from a Gaussian shape increases significantly with energy. Evaluations performed mostly at 662 keV demonstrated that peak shape was independent of count rate, at least in the studied input range of 10–75 kcps.

Efficiency was evaluated across the energy range, with a focus on the 185-keV area. The key parameter under consideration was the combination of the absorption in the detector window and the actual intrinsic efficiency of the CZT crystals, which represents the probability that incident gamma rays on the detector external surface in the solid angle subtended by the detector active surface at the source will interact and give a pulse in the full energy peak. The efficiency parameter can therefore be expressed as follows:

$$\varepsilon = \frac{N}{A.BR}\frac{4\pi}{\Omega(\mathrm{d},\mathrm{D})}.100\%$$

where N is the net peak area in cps; A is the activity of the source in Bq; BR is the gamma emission probability; and **Ω** is the solid angle represented by the detector from the point source position, which is a function of the distance between the source and the detector surface d and of the detector active area D. Results (see Figure 5) show that the efficiency parameter for the M400 detector at 185 keV is typically about 53%. For reference, the efficiency parameter measured in similar conditions with LaBr and ECGS detectors at 185 keV has values of 62% and 57%, respectively. It should be noted that the LaBr and ECGS detectors used by the IAEA have larger active areas of about 20.2 cm^2 and 19.6 cm^2, respectively, making the absolute efficiency difference larger. In the course of the evaluation, a 15-mm-deep version of the CZT crystal also was tested, and showed that efficiency increased by a factor on the order of 30% compared to the standard 10-mm-deep version, i.e., the absolute efficiency of the 15-mm-deep version is approximately equivalent to that of the ECGS.

Dead time, its correction and pileup rejection were evaluated by means of several parameters. Overall dead time magnitude as a function of input count rate was measured across a wide range and is shown in Figure 6. For reference, dead time magnitude at a 10 kcps input count rate is about 2.5% and 12% for LaBr and ECGS detectors, respectively, vs. 5% for the M400. Dead time correction precision with pileup rejection implementation was evaluated two ways: 1) using the two-source method and 2) measuring the change in the count rate in the region of interest produced by a source in a fixed position in a configuration with the source alone and in configurations with additional sources of lower energies. The first method used Cs-137, whereby performance is characterized by the relative deviation R of the net area count rate in the presence of two sources (CR_{12}) to the sum of the net area count rates for each source (CR_1, CR_2):

$$R = \left(\frac{CR_{12}}{CR_1 + CR_2} - 1\right).100\%$$

The R ratio was found to be within ±1% up to an input count rate of about 70 kcps with the two sources.

The second method was performed either with uranium in a fixed reference position and Am-241 at various positions or with Co-60 in fixed reference position and Cs-137 at various positions. The measurements were processed to extract the relative deviation D of the peak count rate in the presence of both sources (CR_B) to the peak count rate in the presence of reference source alone (CR_A):

$$D = \left(\frac{CR_B}{CR_A} - 1\right).100\%$$

D values were found to remain below ±1% up to about 30–35 kcps. Above this count rate, the deviation was found to rise to about 4% at 60 kcps, which can be expected to introduce significant bias. It is worth noting, however, that the vast majority of IAEA fresh fuel measurements are not expected to exceed 30 kcps.

Time stability was evaluated through repeated measurements performed with a Ho-166m source and M400 modules in a fixed geometry for about 65 hours of continuous operation. The measurements were processed to assess the stability of several spectrometric parameters, again focusing on the 185-keV region. Table 1 shows the results in terms of the relative standard deviation (RSD) of each parameter over all measurements. Very good stability is observed for the count rates in the region of interest (efficiency), with uncertainty (i.e., measured RSD) on the order of the random component, due to the counting statistics for the particular setup used. Peak centroid position and total count rate in the spectrum are also very stable. Other peak parameters (FHWM, FWTM/FHWM ratio) show more fluctuation, yet remain mostly within 1% RSD. Stability across power cycles also was evaluated, with a focus on efficiency-related parameters. For short power cycles of a few minutes, no significant effect on stability was observed by powering the device off/on. However, for longer power cycles of a few hours, a definite trend could be seen for about half an hour after powering on, with an offset kept to below one percent on the first 1-hour spectrum.

The influence of external temperature on performance was thoroughly studied, as it is an important parameter for field operations. Initial observations showed that efficiency is significantly dependent on temperature. (Other parameters also showed some temperature-dependence, but these parameters are not critical for the intended applications.) The magnitude of this efficiency dependency was progressively and significantly reduced throughout the various design iterations, addressing mainly the primary issue at high temperatures.[4] The influence of temperature on detector performance was evaluated in experiments involving a source and M400 modules in a fixed geometry placed inside a thermal chamber, where the temperature was adjusted within the operating range of the instrument (-10 to +35 °C across typical temperature cycles of 24–72 hours duration) and the instruments set to repeated measurement acquisition (similar to the time stability measurements). The measurements were processed to assess the temperature-dependence of several spectrometric parameters in the 185-keV area. The results in Figure 7 show that temperature has a definite effect on the instrument response parameters, with net count rates varying within an overall relative range of up to about 1.5%, total spectrum count rate up to about 4%, FWHM up to about 20% and the FWTM/FWHM ratio up to about 10%. The peak centroid position shows a minimal variation of 0.3%. While efforts are ongoing to address the effect of temperature on detector efficiency (which is the critical parameter for field applications), it should be noted that above 0 °C, which is typical for most use cases, the overall efficiency variation was below 1% for all M400 devices.

Some specific tests on the influence of a constant magnetic field were performed, in particular because one of the options for field use of the instruments relies on a holder secured to the cylinder with magnets during the measurement (Figure 1). The M400 was tested in a fixed source-to-detector geometry, with measurements performed under 1) no external magnetic field except the natural one, and 2) an additional magnetic field of about 350 µT with a constant intensity and direction, wherein the orientation of the detector-source set varied. An assembly consisting of a Helmholtz coil (0.9 x 0.9 x 0.5 m) connected to a standard power supply was used to generate the magnetic field. The effect on all parameters of interest was observed to be minimal and stayed within the amount of variation expected under the baseline condition of no external magnetic field. Power consumption was determined by assessing the battery life of a typical, commercial off-the-shelf battery pack. Battery capacity of 67 Wh provided an operating time of about 15 hours.

Overall, the performance of the M400 module was gauged to be appropriate for the intended purpose and field context. A few particular characteristics that limit applicability in certain cases were observed; these include:

- Increased pileup contributions/dead time correction inaccuracy above 30–35 kcps;
- Efficiency bias slightly exceeding 1% after a long shutdown (power off);
- Temperature effect on detector efficiency, with basic mechanisms still not fully understood and potentially exceeding 1% at temperatures below 0 °C;

– Possible effects of the specific signal processing approaches, e.g., changes in FWHM due to automated pixel gain stability correction; a particular peak shape resulting in large spread of peak counts, especially at high energies.

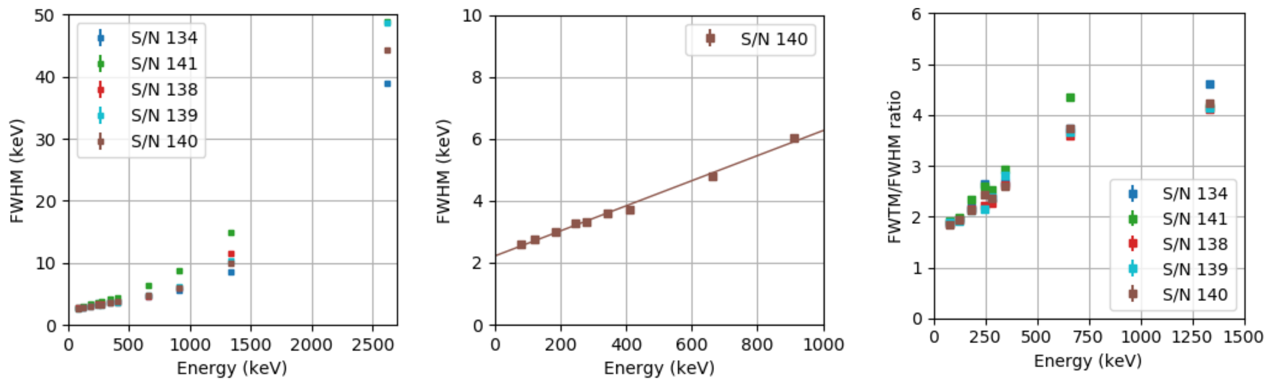

Figure 3. FWHM as a function of energy in the overall energy range for several detectors (left), of the linear fit of the FWHM in the sub-MeV energy range for one detector (middle) and of the FWTM/FWHM ratio as a function of energy (right).

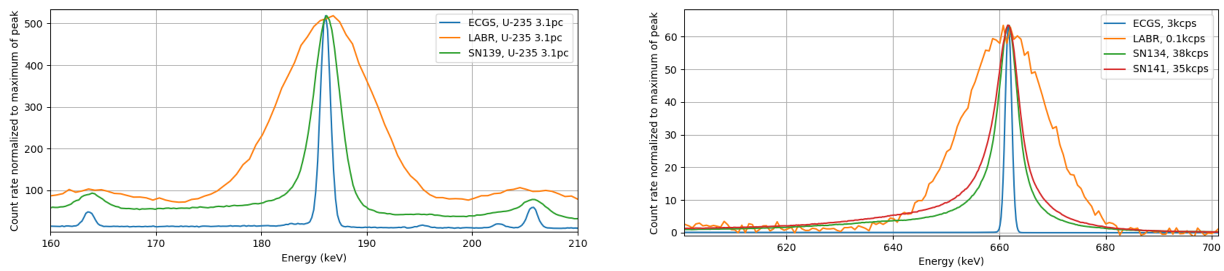

Figure 4. Visual comparison of peak shape for some uranium and arbitrary Cs-137 measurements taken with a few of the M400 modules and an LaBr and an ECGS detector (left: 185-keV region, right: 662-keV region). For visual comparison, counts were scaled to the maximum of the peak and peak positions aligned.

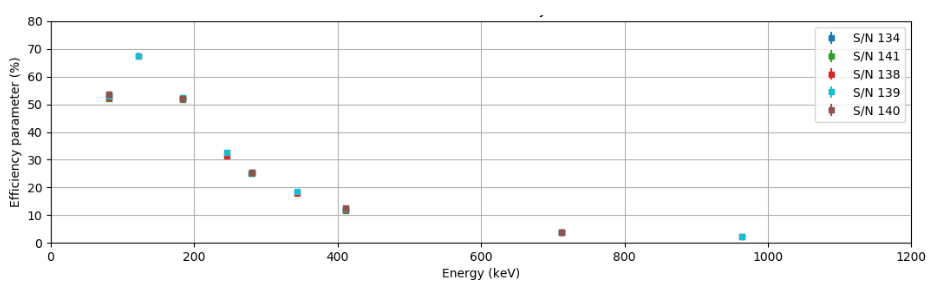

Figure 5. Efficiency parameter as a function of energy, based on point sources at 25 cm between the source and detector surfaces.

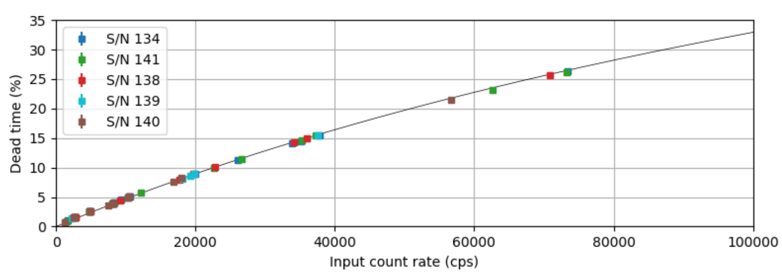

Figure 6. Dead time (DT) as a function of count rate (CR) and fit with the function DT=100/(1+a/CR), parameter a=203675.67.

Table 1. Relative standard deviation on various parameters from a set of 1-hour spectra with modules kept on and measuring for about 65 hours. The average random component of the RSDs (due to counting statistics) over all modules, for total count rate and ROI net count rates, is about 0.024% and 0.18%, respectively.

Parameter	Relative standard deviation on the measurement set (%)				
	134	138	139	140	141
Total count rate	0.041	0.045	0.037	0.036	0.043
ROI net count rate	0.190	0.179	0.197	0.221	0.222
Centroid	0.0068	0.0059	0.0241	0.0145	0.0164
FWHM	0.67	1.53	0.76	0.66	1.01
FWTM/ FWHM	0.74	0.66	1.07	0.73	0.68

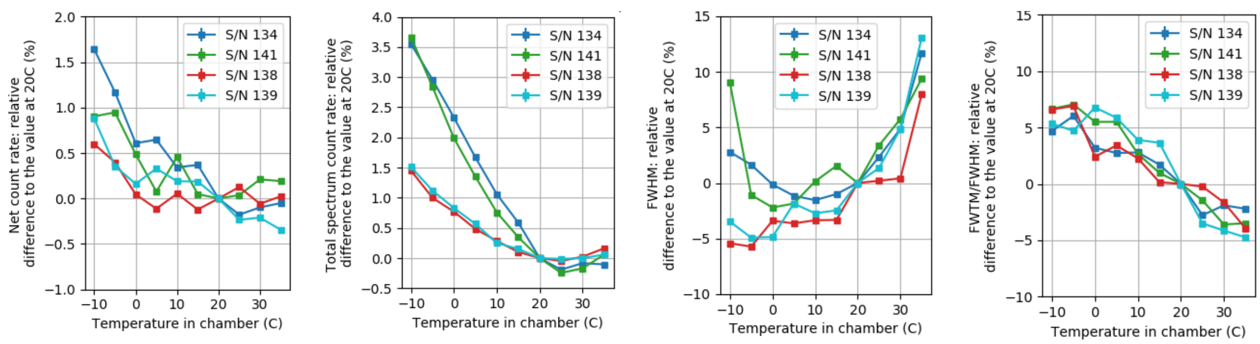

Figure 7. Temperature test results in terms of the relative difference of a parameter to its value at 20 ℃, as a function of temperature, for the following parameters (left to right): net ROI count rate, total spectrum count rate, FWHM, FWTM/FWHM ratio. The source used was Ho-166m and the region of interest addressed the 184-keV peak.

4.2 Application-specific performance of the MCCM

Some initial evaluations related to the MCCM's overall suitability for designated safeguards applications have already been completed, and further system performance assessments are ongoing.

The initial application-specific evaluation mostly addressed the quasi infinite thickness application. A batch of five M400 modules equipped with a specifically designed collimator (see Figure 1) was tested at the IAEA laboratories in Seibersdorf. Measurement setups were similar to those used for the experimental tests of the GEM software [7]: reference samples consisting of U_3O_8 discs (20 mm thick x 163 mm diameter) at enrichment levels ranging from 0.217% to 19.82% U-235 wt.% encapsulated in aluminium cans with a wall thickness of 2 mm. Sets of measurements were performed either without an attenuator or with 4 mm to 16 mm thick steel attenuators, with measurement times from 300 s to 2500 s. For each MCCM system, the enrichment calibration was determined from the 3.105% – no attenuator configuration, and the wall thickness calibration from the 19.82% – 16-mm steel attenuator configuration. Data analysis was performed using the GEM analysis algorithm of the MCAT software. Statistical analysis was performed in a similar way as described in [7], with the random (U_R) and systematic (U_S) uncertainty components, as well as the combined (U_C) uncertainty derived from the relative differences $\{\Delta_i\}_{i=1..N}$ between the measured (M_i) and reference (R_i) enrichment for the set of N measurements performed (excluding calibration measurements), as per the following formulas:

$$\Delta_i = \frac{M_i - R_i}{R_i} . 100\%,\ \ U_S = \frac{1}{N} . \sum_{i=1}^{N} \Delta_i,\ \ U_R = \sqrt{\frac{1}{N-1} \sum_{i=1}^{N} (\Delta_i - U_S)^2},\ \ U_C = \sqrt{{U_S}^2 + {U_R}^2}$$

Performance of the system is compared to international target values (ITVs [8]) and performance of other detector types [7]. The preliminary values obtained in this initial evaluation (Table 2, Figure 8) are generally in line with those reference values.

Table 2. Uncertainty component estimates from the tests performed at Seibersdorf.

	M400 serial number					All M400
	138	139	134	140	141	
U_S	-1.12	0.04	-1.00	-0.84	-1.13	-0.81
U_R	1.54	2.14	1.93	2.18	1.64	1.91
U_C	1.91	2.14	2.18	2.33	2.00	2.08

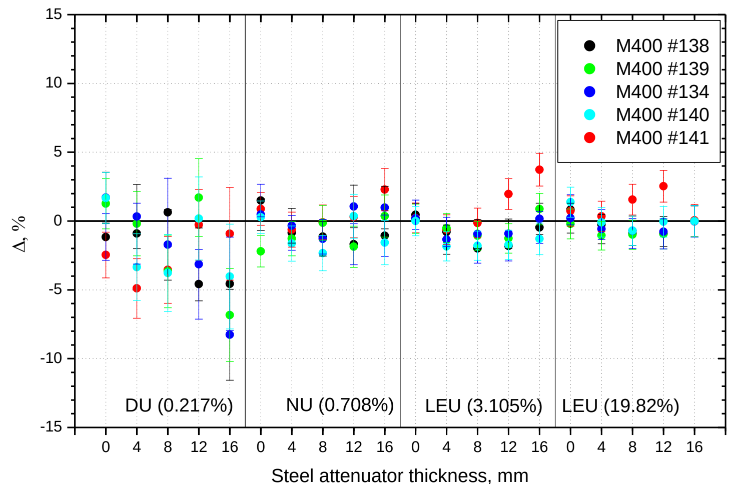

Figure 8. Relative difference between measured and reference enrichment for all detectors tested.

5 Integration into the IAEA safeguards instrumentation environment

Before a new instrument can be used for safeguards verifications, it needs to pass a two-stage process: validation and authorization. A prerequisite to authorization, validation is internal to the Division of Technical and Scientific Services and confirms the design compliance of any type of safeguards equipment; it provides assurance, through verification, that safeguards equipment meets all required design characteristics, features and functions. Validation applies to new instruments, as well as to upgrades, configuration changes, software and individual components, whether designed and built in-house or commercially sourced. Safeguards equipment authorization is a departmental process which provides assurance that safeguards equipment is fit for purpose, i.e., that the results provided by the instrument can effectively be used in support of verification processes. Furthermore, the authorization process evaluates potential joint use scenarios, application-dependent vulnerability assessment, as well as application-specific user manuals and trainings.

For the MCCM, the validation process consisted of two component validations (the M400 CZT module and revalidation of the MCAT software for interfacing with the CZT module) and one safeguards equipment (instrument) validation of the MCCM. All three validations were approved in February 2023.

Once the MCCM instrument was validated, the demonstration of its suitability for the envisaged safeguards verification methods could be started. As detailed in section 4.2, the authorization activities include the demonstration of the measurement performance by the analysis of experimental measurement data from the laboratory and/or the field to provide an estimate of provisional RSD compared to the ITVs [8] to support the conclusion that the measurement performance of the MCCM meets the safeguards applications requirements. Completion of the authorization is expected later this year.

Additionally, the MCCM instrument needed to be added to the Safeguards asset management system for proper inventory control, and checklists and performance and failure tracking forms and procedures created as quality control and quality assurance measures.

6 Looking ahead

Future activities are mainly related to:

- Further assessing application-specific performance for full deployment as a primary verification tool;
- Monitoring long-term performance and remedying the observed limitations, in particular via an extended inventory of instruments;
- Further improving performance and expanding the range of applications covered by the instrument, in particular by using inherent features of the technology, e.g., list-mode data including 3D location of each time-stamped interaction in the crystal;
- Integrating the M400 CZT module into a handheld form factor (to replace and expand uses of the standard, NaI-based, handheld HM-5 inspector instrument [1]);
- Assessing the applicability of the M400 CZT modules for the verification of plutonium-bearing material (already initiated).

7 Conclusion

The new MCCM system, which is based on a large-volume CZT detector and standard data acquisition and analysis software, is being introduced into the pool of instruments used for IAEA safeguards verifications of unirradiated uranium compounds. The system evaluations conducted so far show that its general performance is in line with the expected target performance per the relevant ITVs. The IAEA internal processes necessary to achieve full readiness for deployment for inspection use of this new instrument are in progress, with finalization (authorization) expected later this year. Efforts are ongoing to further assess and improve performance, address observed limitations, improve performance and expand the range of applications.

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[8] International Target Values for Measurement Uncertainties in Safeguarding Nuclear Materials, STR – 368 (Revision 1.1), September 2022.

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[1] A 3D-printer was used for rapid creation and testing of different holder and stand prototypes, e.g., the pellets holder or the magnetic holder for UF_6 cylinders depicted in Figure 1.

[2] Full width at half maximum

[3] Full width at tenth-maximum

[4] However, some temperature dependency remains at low ambient temperatures (typically below 0 °C). While in-depth studies are ongoing in order to reach a detailed understanding of the physical processes governing this behaviour, the empirical characterizations were performed and led to the implementation of a temperature-dependent correction of the efficiency based on the device's internal temperature sensor and factory calibration of each device across the temperature range.