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Comparing Metrological Characteristics of Methodologies for Determining Uranium Enrichment Using NaI and LEGe Detectors

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Abstract. The purpose of this work was to compare metrological characteristics of methodologies for determining uranium enrichment using different types of detectors and the IMCA software. This software includes several programs (applications), two of which were used in this work: IMCN was used for NaI detector, and IMCG was used for LEGe detector. The principle of enrichment measurement is that the uranium enrichment is directly proportional to the count rate at the 185.7 keV peak resulting from decay of ^{235}U , provided that the sample's thickness is sufficient ("infinite thickness" method). Based on the results of repeated measurements of samples with known enrichment, metrological characteristics of methodologies for different count times were calculated and compared. Analysis of the results led us to the following conclusions: reproducibility of results when performing measurements using LEGe detector is just as good as when using NaI detector; at that, uncertainty reported by the IMCG program is almost the same as the reproducibility estimate, while the IMCN program reports uncertainties that are several times smaller than the estimated reproducibility.

1. Introduction

At present, great importance is attached to safeguards and non-proliferation and security of nuclear weapons and nuclear materials. Activities related to upgrading nuclear materials physical protection, control and accounting system and developing state nuclear materials control and accounting system were launched by the Decree of the President of the Russian Federation dated September 15, 1994 "On Priority Measures to Improve Nuclear Materials Accounting and Security System."

The nuclear materials control and accounting system is an important safeguard to ensure security when performing activities related to use of nuclear materials. One of the primary goals of the system is to obtain data on quantitative and isotopic composition of plutonium, uranium or thorium, which are available, manufactured, obtained in the material balance area, or shipped from it[1].

All NM are subject to control and accounting, but control and accounting procedures themselves, as well as control and accounting methods, depend on physical, chemical and radiological properties of nuclear materials containing fissile substances. Most of control methods are based on measuring various types of radiation [2]. The most widely used methods are non-destructive gamma-spectrometry ones [3-5].



The purpose of this work was to compare metrological characteristics of methodologies for determining uranium enrichment using detectors of different types and the IMCA software, as well as to assess impact of the measurement time on the uncertainty of measurement results.

2. Description of Measurement Systems and Software

2.1. IMCA Software

IMCA (Inspector Multichannel Analyzer) Inspector is a portable gamma spectrometer designed to measure uranium enrichment. It includes a computer-controlled digital pulse analyzer (Inspector) made by Canberra, NaI or LEGe detector, and specialized software. The enrichment measurement program is based on specially designed data acquisition and processing procedures. The built-in IMCN (Inspector Multi-Channel Analyzer with Natrium Detector) program is used when working with NaI detector, and the IMCG (Inspector Multi-Channel Analyzer with Germanium Detector) program is used when working with LEGe detector.

Having the ability to quickly change detectors from NaI to LEGe and back, IMCA meets the requirements for both low and high-resolution gamma spectrometry systems. The device complies with the IAEA requirements for testing materials with enriched uranium [6, 7].

2.2. Principles Behind the Uranium Enrichment Measurement Method

The measurement method is based on the fact that uranium enrichment is directly proportional to the count rate at the 185.7 keV peak of ^{235}U , provided that the sample is "infinitely thick". The criterion for approximation of "infinite thickness" of the sample is the condition that the depth of the sample along the collimator axis is 5-7 times greater than free path of gamma quanta with the energy of 185.7 keV in the sample material. In this case, all samples with similar physical compositions are represented by the same volume, "visible" to the detector. For homogeneous samples of the same composition, the "visible" volume contains the same amount of uranium and the measured intensity at the 185.7 keV peak is proportional to the ^{235}U enrichment. The measurement system must be calibrated using reference materials [8].

Calibration is carried out by measuring count rate at the 185.7 keV peak from reference materials of known enrichment in containers made of known material and with known wall thickness. The calibration factors are determined based on the measurement results; and these factors are subsequently used to determine enrichment of unknown samples. When using IMCN, at least two reference materials are required, and when using IMCG, one is sufficient. If there is a need to enhance accuracy of determining calibration factors, a greater number of reference materials can be used.

The report is generated in the IAEA standard format. The IMCA software can operate both in the IAEA mode, which closely follows the existing IAEA procedures, and in a simpler general mode that is selected by the user at startup during setup.

Determination of peak area used in IMCA generally depends on smoothness and extent of the Compton continuum. There are several factors that can distort this continuum, which can lead to biased results when working with a NaI detector:

- the main contribution to the Compton continuum comes from scattered gamma quanta with energies of 765 and 1001 keV from $^{234\text{m}}\text{Pa}$, a daughter product of ^{238}U . Daughter decay products are usually in equilibrium with ^{238}U . Processing, cascade enrichment of UF_6 and conversion of UF_6 to UO_2 results in removal of daughter products, changing the background region. The equilibrium is established after 3 months.
- newly recycled materials may have increased ^{237}U and ^{232}U content, emitting 208 and 238 keV gamma rays, respectively, which can interfere with the 186 keV peak.

Use of LEGe detectors helps in overcoming these problems, however, their efficiency is not as high as that of NaI [9, 10].

3. Experimental Results

In the course of this work, measurements of three uranium samples with different enrichments (Table 1) were carried out using scintillation NaI and LEGe detectors. For each sample and type of detector, three series of three repeated measurements with different count times (60, 120, 180, 300, and 600 s) were performed, 270 measurements in total.

Table 1. Reference Material Data.

RM #	Atomic Enrichment, %	Mass, g	Matrix Type	Container Material	Container Thickness, mm
1	1.3	150	UO ₂	Polyethylene	2
2	1.8	150	UO ₂	Polyethylene	2
3	3.0	150	UO ₂	Polyethylene	2

Based on the data obtained, repeatability and reproducibility of the repeated measurements results were calculated [11]. Results of assessing metrological characteristics of the methodologies for measuring uranium enrichment, as well as uncertainty of a single measurement generated by the program in the report, are presented in Table 2. Comparison of the results demonstrated that root-mean-square (RMS) deviations obtained under repeatability conditions for the two detectors do not differ significantly. Reproducibility indices of the methodologies are also comparable, and when measuring sample #1 (Figure 1), the LEGe detector yields the best results.

Table 2. Summary Results of Estimating Repeatability σ_{ml} , Reproducibility σ_{Rm} and Absolute Uncertainty ΔE_{ml}^{prog} Reported by the Program for Sample #1.

Sample #	1				2			3		
	t, s	σ_{ml}	σ_{Rm}	ΔE_{ml}^{prog}	σ_{ml}	σ_{Rm}	ΔE_{ml}^{prog}	σ_{ml}	σ_{Rm}	ΔE_{ml}^{prog}
IMCN	180	0,0218	0.029	0.013	0.0142	0.010	0.013	0.0173	0.041	0.013
	300	0,0066	0.030	0.010	0.0080	0.008	0.010	0.0064	0.038	0.010
	600	0,0024	0.022	0.007	0.0027	0.005	0.007	0.0032	0.047	0.007
IMCG	180	0,0148	0.021	0.020	0.0097	0.018	0.031	0.0239	0.034	0.035
	300	0,0057	0.012	0.017	0.0077	0.011	0.028	0.0118	0.030	0.033
	600	0,0083	0.010	0.015	0.0097	0.020	0.027	0.0051	0.041	0.031

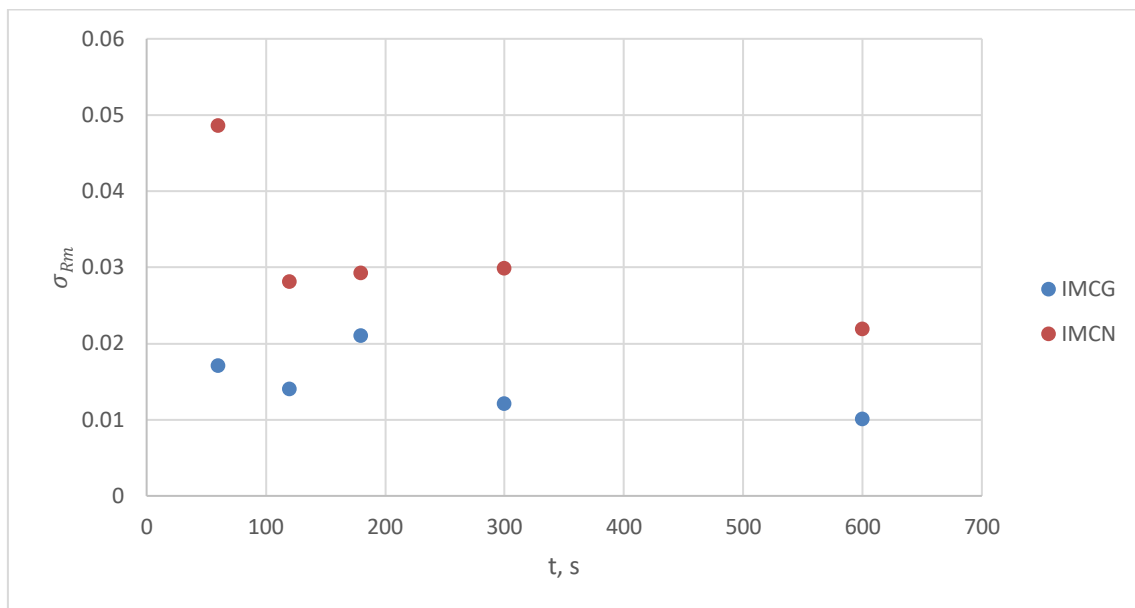


Figure 1. Graphs of Reproducibility of Analysis Methods as a Function of Measurement Time for Sample #1.

Comparison of absolute uncertainties resulting from using the programs as a function of the measurement time (Figure 2) demonstrated that use of IMCN results in absolute uncertainty of 2-3 times smaller than that of IMCG, but this contradicts the experimental results of reproducibility estimates obtained during measurements with NaI detector. The absolute uncertainty values reported by the IMCG software are marginally higher than experimental reproducibility estimates.

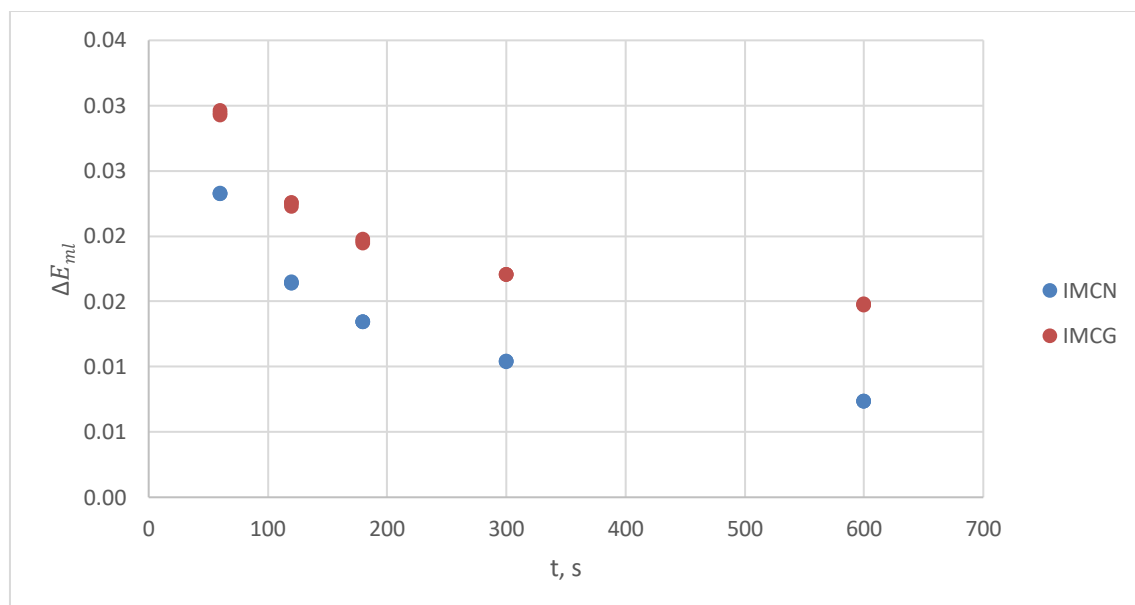


Figure 2. Graphs of Absolute Uncertainty as a Function of the Measurement Time for Sample #1 Reported by the IMCN and IMCG Programs.

4. Conclusion

To assess metrological characteristics of the methodologies, numerous repeated measurements of uranium samples with different enrichment were carried out under repeatability and reproducibility conditions using two measurement systems. Based on the measurement results, the metrological characteristics of the methodologies for different count times were calculated and compared, which demonstrated that reproducibility of measurements with LEGe detector is just as good as with NaI detector, and that the uncertainty reported by the IMCG program is almost the same as the reproducibility estimate, while IMCN reports the enrichment measurement uncertainty that is several times lower than the reproducibility estimate. To clarify the results of this study, it is advisable to continue performing repeated measurements in order to identify the cause of this discrepancy.

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