Error reduction in gamma-spectrometric measurements of nuclear materials enrichment

D Zaplatkina[1], A Semenov[2], E Tarasova[2], V Zakusilov[2] and M Kuznetsov[2]

[1] St. Petersburg Nuclear Physics Institute n.a. B.P. Konstantinov, Gathina, Russia
[2] National Research Tomsk Polytechnic University, Tomsk, Russia

E-mail: kms@tpu.ru

Abstract. The paper provides the analysis of the uncertainty in determining the uranium samples enrichment using non-destructive methods to ensure the functioning of the nuclear materials accounting and control system. The measurements were performed by a scintillation detector based on a sodium iodide crystal and the semiconductor germanium detector. Samples containing uranium oxide of different masses were used for the measurements. Statistical analysis of the results showed that the maximum enrichment error in a scintillation detector measurement can reach 82%. The bias correction, calculated from the data obtained by the semiconductor detector, reduces the error in the determination of uranium enrichment by 47.2% in average. Thus, the use of bias correction, calculated by the statistical methods, allows the use of scintillation detectors to account and control nuclear materials.

1. Introduction

One of the systems providing the required control level over the activities in the nuclear industry is a system of accounting and control of nuclear materials. The system is designated to ensure continuous systematic accuracy of information on nuclear material and to detect unauthorized actions. Nuclear materials accounting is based on the measurements results of quantitative characteristics and attribute properties of nuclear materials.

The development and improvement of non-destructive methods of isotopic composition analysis are directly related to the solution of practical problems in the field of accounting and control of nuclear materials, as well as detection and prevention of illicit trafficking. One of the main methods of non-destructive analysis is the gamma-ray spectrometry which is widely applied for the nuclear materials control. This is a key method in determining the isotopic composition of the nuclear materials and uranium enrichment [1].

However any measurement process is inevitably linked with the need to process the data. The most important aspects of the test results processing are accurate and correct calculations which include errors at all stages of the work and their correct interpretation [2].

Thus the research goal is to optimize the process of the uranium isotopic composition analysis with spectrometric channel based on NaI (Tl) scintillation detector by means of measurement error reduction with statistical methods.

2. Results and Discussion

To determine the accuracy of measurements it is necessary to run a series of experiments to define the amount of nuclear material. We have used a spectrometric channel on the basis of the NaI (Tl)

scintillation detector (Canberra, model 802-2x2), spectra processing was performed in the Genie-2000 software, and uranium dioxide UO_2 with different mass of U^{235} enrichment by 2% was used as a sample. Nuclides were matched by the developed nuclide library. There were 3 measurements for each source spaced 0 cm from the detector at 1200 live time. The measurement results are shown in the Table 1 [3, 4].

Table 1. The results of the samples measurement with a scintillation detector

Sample mass, g.	4	6	8	10	12	14
	0.4557	0.3221	0.9272	0.4630	0.4886	0.4812
	0.3117	0.8896	0.9654	0.5847	0.9442	0.3015
Calculated enrichment, %	0.3037	0.2660	0.9389	0.6649	0.8236	0.5922
	0.8024	0.2670	0.5509	0.5418	0.8995	0.4091
	0.3157	0.8896	0.9674	0.6159	0.6552	0.4562
Moderate enrichment, %	0.3570	0.4926	0.9438	0.5709	0.7521	0.4583
Measured result dispersion	0.0574	0.4905	0.0004	0.0317	0.0985	0.1024
Absolute error of enrichment measurement	1.6430	1.5074	1.0562	1.4291	1.2479	1.5417
Relative error of enrichment measurement, %	82.1492	75.3710	52.8079	71.4569	62.3940	77.0843

As it can be seen from the table, the error of the results is significant and accounts for a prominent uncertainty in the measurements due to the plurality of the latter. To increase the accuracy of measurements the methods of mathematical statistics were applied.

The method for several samples was used to evaluate the systematic error and to introduce the bias correction. The high resolution semiconductor germanium detector Canberra in conjunction with the installed control environment Genie-2000 was used as a more accurate method [3].

To calculate the bias we performed measurements using the same set of uranium dioxide samples. There were 3 measurements for each source spaced 0 cm from the detector at 1200 live time. Average enrichment values measured by semiconductor detectors are considered as accepted ones.

The bias estimation was calculated by the formulas [5]:

$$\theta = \sum_{k=1}^{m} w_k \cdot (\overline{x}_k - \mu_k) \Big/ \sum_{k=1}^{m} w_k \tag{1}$$

$$w_k = (\sigma_k^2 + s^2 / n_k)^{-1} \tag{2}$$

$$s^2 = \sum_{k=1}^{m} (n_k - 1) \cdot s_k^2 / (n - m) \tag{3}$$

Bias estimation results are shown in the Table 2.

Table 2. Bias estimation of the sample mass determination

Sample number, k	σ2	s2	w	θ
1	0.00008	0.05738	23.00663	-1.35436
2	0.00366	0.49054	21.25564	
3	0.00181	0.00043	22.12301	
4	0.00022	0.03171	22.92836	
5	0.00003	0.09854	23.02916	
6	0.00005	0.10243	23.02232	

The resulting bias estimate is introduced as a correction for every measured value with the opposite sign, thus compensating the calculated bias. Corrected results of uranium enrichment determination by scintillation detector are presented in Table 3.

Table 3. Corrected uranium enrichment determination by NaI(Tl) detector

Sample mass, g	4	6	8	10	12	14
Enrichment without correction, %	0.438	0.527	0.870	0.574	0.762	0.448
Relative enrichment error without correction	78.109	73.656	56.501	71.297	61.890	77.598
Enrichment with correction, %	1.792	1.881	2.224	1.928	2.117	1.802
Relative enrichment error with correction	10.390	5.938	11.217	3.579	5.828	9.880

According to the measurement results analysis, it was found that the measurement bias correction determined using a more accurate method of standard samples analysis as well as mathematical and statistical techniques can reduce the relative systematic error component in the measurements performed by the detector approximately by 47.2% in average. The main condition used for the bias estimation is that the samples' physical, chemical and material composition should match that of the samples investigated by the detector as close as possible. The studied experiment complies with this condition, because one set of uranium samples was used.

Since the systematic error is a main contributor to the measurement error, we need to consider the dependence of the unaccounted material amount (UMA) on the systematic error.

The dependence of the UMA value change, its variance, standard deviation and confidence intervals are expressed by the formulas [5]:

$$\Delta UMA = 0.472 \cdot \sqrt{S} \tag{4}$$

$$\Delta(\sigma^2_{UMA}) = S \cdot (\sigma^2_{\delta 2} - \sigma^2_{\delta 1}) = -0.3244 \cdot S \tag{5}$$

$$\Delta(\sigma_{UMA}) = \sqrt{S} \cdot 0.0131 \tag{6}$$

$$\Delta Di = 2\sqrt{S} \cdot 0.472 \cdot (z_{1-\alpha/2} + 1) \tag{7}$$

According to the calculations by the formulas stated above, we plotted the graph showing the dependence of UMA value change on the stratum size and average mass of each stratum element.

With a decrease in the confidence interval the UMA value range decreases as well. This fact can be conditioned by the measurement errors, thus increasing the detection probability of loss or change of materials. The increase in the detection probability of material loss or change with the use of the studied method equals 14.6%.

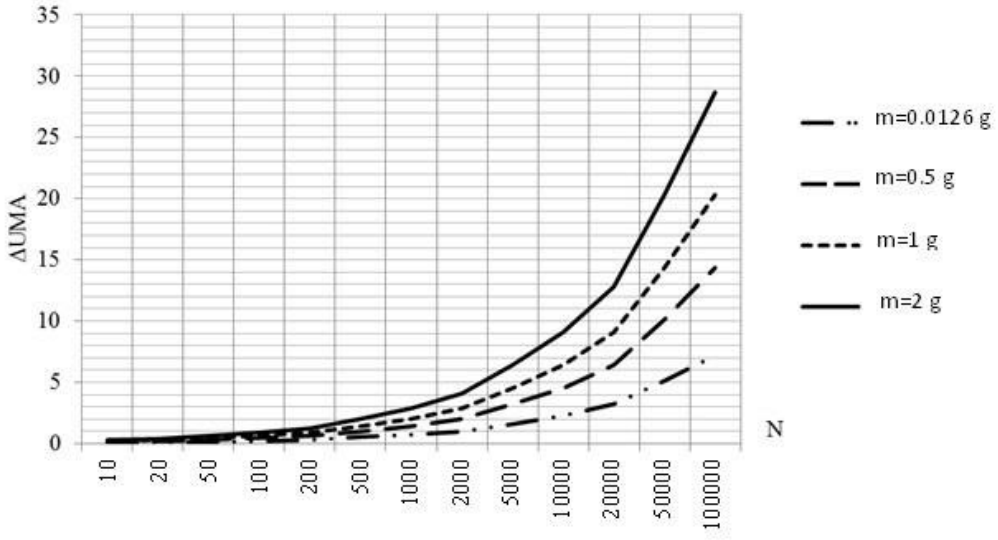

Figure 1. UMA value change depending on the stratum size and elements' mass.

3. Conclusion

The results analysis was used to estimate the bias correction estimation which equals 1.35%. The use of bias correction reduced the relative systematic error of measurements made by scintillation detector by 47.2%.

We have stated the dependence of the change in the unaccounted material amount, variance, standard deviation, confidence intervals of UMA, and the detection probability of nuclear materials change on the change in the measurement error. The described method allows for the increase in the detection probability of nuclear materials change by 14.6%.

Thus, the use of statistical methods to reduce the errors in uranium enrichment measurements made by the scintillation detector may be used to solve the accounting and control issues problems with acceptable accuracy.

References

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