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VNIIEF-ORNL Joint Plutonium Measurements with NMIS and Results of Plutonium Attributes Preliminary Evaluations

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ABSTRACT

Within the frameworks of TO № 007 between ORNL and VNIIEF on Nuclear Materials Identification System (NMIS) mastering at VNIIEF in July 2000 there had been finalized joint measurements, in which NMIS-technique equipment was used that had been placed at VNIIEF's disposal by ORNL, as well as VNIIEF-produced unclassified samples of fissile materials. In the report there are presented results of experimental data preliminary processing to obtain absolute values of some attributes used in plutonium shells measurements: values of their mass and thickness. Possibility of fissile materials parameters absolute values obtaining from measurement data essentially widens NMIS applicability to the tasks relevant to these materials inspections.

1. Parameters of plutonium shells under study.

The plutonium shells mass-geometric parameters (δ-phase, ^{240}Pu percentage is 1,8) are presented in Table 1.

Table 1. Parameters of the plutonium assemblies

ASSEMBLY NUMBER	$R_{int.}$, cm	$R_{ext.}$, cm	Mass, g	Spherical components dimensions		Neutron multiplication, q
				$R_{int.}$, cm	$R_{ext.}$, cm	
1	1,00	4,02	4091,0	1,00	1,40	2,26
				1,40	3,15	
				3,15	4,02	
2	1,40	4,02	3768,0	1,40	3,15 *)	2,07
				3,15	4,02 **)	
3	3,15	4,66	4468,3	3,15	4,02	1,46
				4,02	4,66	
4	5,35	6,00	4004,4	-	-	1,16
5	1,40	3,15	1829,0	-	-	1,51
6	4,02	4,66	2315,1	-	-	1,12
7	4,66	5,35	3316,1	-	-	1,18
10	3,15	4,02	2153,2	-	-	1,22

*) without a stopper in the upper hemisphere
**) without stoppers in either of the hemispheres, stopper diameter ~ 2,20 cm.

For the above plutonium assemblies both active and passive measurements have been fulfilled for “bare” assemblies as well as for the same assemblies containerized into AT 400.

Radiation signatures of the studied system obtained in NMIS-measurements present an objective representation of its gamma-neutron characteristics and can be used to get diverse information relevant to the system. The way to deal with the measured signatures is different for classified and unclassified objects.

To obtain numerical values of such attributes of an object under study as, for example, values of fissile substance mass and dimension, absolute values of the measured signatures need to be used. Such analysis can be applied only to unclassified objects measurements.

Results of the performed measurements can be also presented in non-intrusive form – as a ratio of signatures of an inspected object and a “reference” one. Unclassified data obtained in such way may be useful for containerized classified objects safety (identity) non-intrusive inspecting. NMIS-measurements application to such tasks has been described earlier in ORNL reports, e.g. see [1] and[2].

2. Determining plutonium spherical shells mass and thickness.

2.1. Measurements with “bare” assemblies.

The signatures of plutonium spherical shells obtained in the measurements can be employed to find mass and thickness of “unknown” assemblies under certain subsidiary conditions. Videlicet, one need to be convinced that all the assemblies, both investigated and “unknown”, are spherical shells produced of δ-phase metallic plutonium, and all assemblies have the same isotopic composition. Such requirements being met, the signatures of NMIS-measurements depend only on two assemblies parameters – shells mass and thickness, for example.

An idea of shell mass and thickness values deriving from the measurements data consists in following. First, the results of measurements of several known plutonium shells are used to obtain dependencies that associate measured signatures peculiarities with shells parameters. The obtained dependencies are then used to determine other shells parameters from results of their signatures measuring.

As spherical shells are completely characterized by two parameters, two independent equations need to be derived, associating any pair of independent characteristics of the measured signatures with the shells mass and thickness (regression equations). First estimation of such possibility was carried out by one of the joint measurements participants – J. Mattingly (ORNL) in the course of measurements in summer 2000. To obtain the sought equations he used the results of measurements in seven assemblies, the values of mass and thickness for the eighth assembly (assembly № 10 from Table 1) were then derived from its signatures measurement results and the regression equations. Comparison of the calculated values with the factual ones characterizes an accuracy degree of such procedure of shell mass and thickness determining. In one variant of such analysis J. Mattingly simultaneously used the results of both active and passive measurements. The below characteristics of the measured signatures have been obtained in active measurements, where distance between a californium chamber and detectors face surface constituted 18,7 cm, and for the passive measurements at a distance of 13 cm between the “bare” assembly center and the detectors face surface. At in-container measurements, the distance was about 25 cm.

As F_1 – first characteristic of the measured signatures – there was used a sum of values of cross-correlation functions between the detectors pairs within the limits of delay time values $\tau = \pm 50$ ns, obtained using the passive measurements data, divided by the total measurement time T. Summation over all the detector pairs is carried out to improve the results statistical accuracy.

$$F_1(M,\Delta) = \frac{1}{T}\sum_{i\neq j}\sum_{-50ns}^{50ns} CC_{ij}(\tau), \frac{1}{\sec} \qquad (1)$$

Here $CC_{ij}(\tau)$ is a value of the cross-correlation function between detector i and detector j at inter-signal delay time τ. T is the total measuring time = N×512×1 nsec, where N is the number of data blocks, employed to calculate $CC_{ij}(\tau)$ values. F_1 value is an average count rate of correlated events for all the detectors pairs, obtained at measurements with plutonium shell of mass M and thickness Δ. In linear regression representation $F_1(M,\Delta)$ function is approximated by a linear dependence on M and Δ:

$$F_1(M,\ \Delta) \approx A_{1M} \times M + B_{1\Delta} \times \Delta + C_1 \qquad (2)$$

Here M is the shell mass, Δ is the shell thickness, A_{1M}, $B_{1\Delta}$ and C_1 are the regression equation coefficients to be found from a condition of best fit for describing results of measuring seven assemblies selected as reference ones.

As F_2 – second characteristic of the measured signatures – there were used active measurements results: a sum of values of cross-correlation function between the californium chamber and the detector in the time interval corresponding to induced fission neutrons recording divided by the total measuring time T. This portion beginning corresponds to californium source neutrons recording cut-off due to the selected discrimination threshold. For the performed measurements conditions it corresponds to the point of time ~ 11 nsec:

$$F_2(M,\Delta) = \frac{1}{T}\sum_{-11ns}^{50ns}[CC_{12}(\tau) + CC_{13}(\tau)] \approx A_{2M} \times M + B_{2\Delta} \times \Delta + C_2 \qquad (3)$$

$$[F_2] = 1/\sec$$

where $CC_{12}(\tau)$ and $CC_{13}(\tau)$ are cross-correlation functions between the chamber and detectors № 2 and № 3 according to the active measurements data. Like the previous case, signatures summation improves statistical accuracy of the results.

Making use of linear regression simulating procedure available in Microsoft Excel applied to the results of measurements with seven samples, coefficients A, B and C have been obtained, which being used in the regression equations give the best description of the experimental data. If the mass M is expressed in grams, and the thickness Δ is expressed in millimeters, then the regression equations obtained in such a way look like:

$$F_1 \approx 0{,}19844 \times M + 20{,}158 \times \Delta - 399 \qquad (4)$$

$$F_2 \approx 0{,}670318 \times M - 105{,}345 \times \Delta + 5443{,}94 \qquad (5)$$

In Table 2 there are presented results of mass and thickness computation for the eighth (reference) sample, which was not involved into the regression equation elaboration.

Table 2

Mass, gram			Thickness, mm		
M_{fact}	$M_{computed}$	Error	Δ_{fact}	$\Delta_{computed}$	Error
2153,2	2461,8	14,3%	8,70	8,0	7,8%

There has been undertaken an attempt to construct regression equations using some other characteristics of the measured signatures, F_3 (F_4) in particular – a characteristic, representing the first gamma-peak square using active measurements data.
As the first gamma-peak square depends mainly on the shell thickness, at first, there has been tested one-parameter – exponential and power – dependence of the square upon the shell thickness.

$$F_3(\Delta) = \frac{1}{T}\sum_{\sim 0ns}^{\sim 3ns}[CC_{12}(\tau) + CC_{13}(\tau)] \approx A_{2\Delta} \times e^{-B_{3\Delta}\times\Delta} \qquad [F_3] = 1/\text{sec} \quad (6)$$

$$F_4(\Delta) = \frac{1}{T}\sum_{\sim 0нсек}^{\sim 3нсек}[CC_{12}(\tau) + CC_{13}(\tau)] \approx A_{4\Delta} \times \Delta^{-B_{4\Delta}\times\Delta} \qquad [F_4] = 1/\text{sec} \quad (7)$$

In Fig. 1 there are shown a graph and equation of the exponential regression, built (like the above) upon the results of seven assemblies measuring.

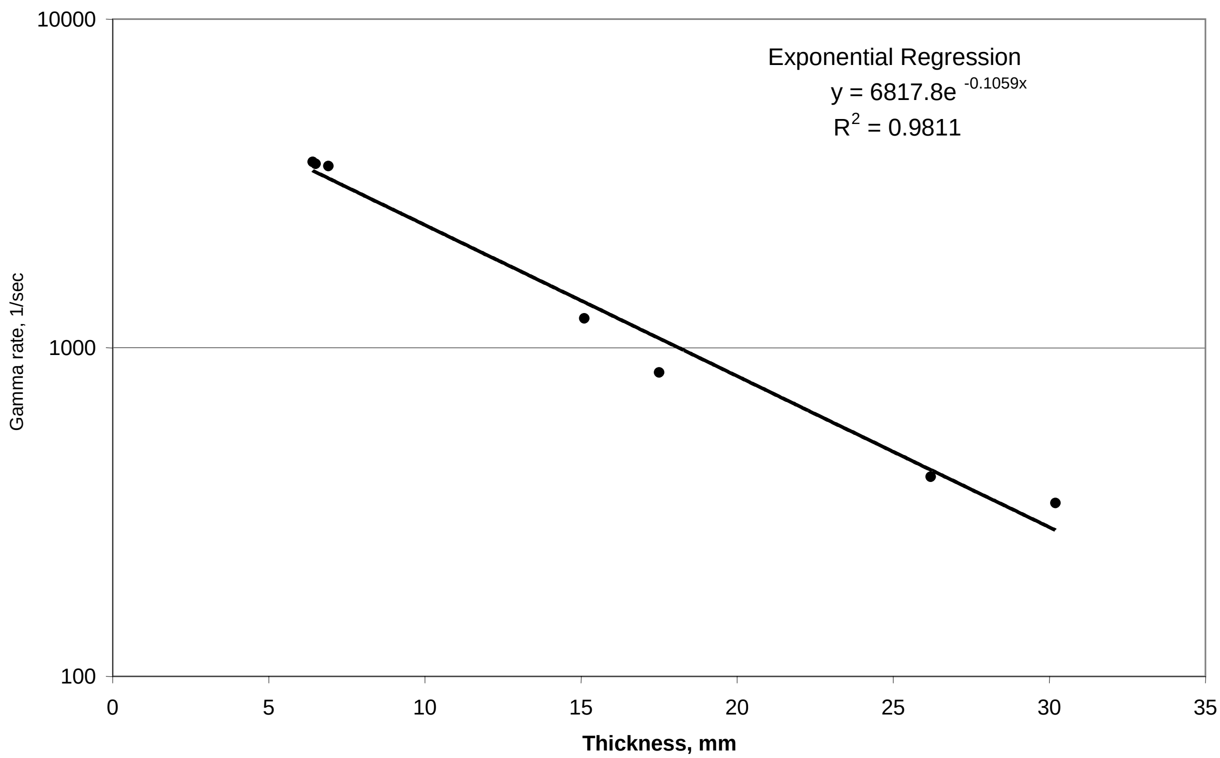

Fig. 1. Exponential regression for average count rate of the source gamma-rays, built on results of measurements of seven “reference” samples (assemblies).

In terms of the above designations the exponential regression equation for the shell thickness is given by:

$$F_3(\Delta) = 6817{,}8 \exp(-0{,}1055\Delta)\ ,\ [\Delta] = \text{mm}, \qquad (8)$$

As it is seen from the experimental data marked in Fig.1 with separate points, gamma-ray count rate dependence on the sample thickness differs from the simple exponent, particularly at large values of the thickness. It seems to be due to the fact that exponential dependence is more suitable under “good” geometry, when multiple scattering quanta contribution to the signal value is absent, but that was not fulfilled in our measurements.

In Fig.2 it is presented a power regression of gamma-ray counting dependence on the sample thickness constructed using the same seven experimental points. It is seen to approximate the experimental data rather better than an exponent does.

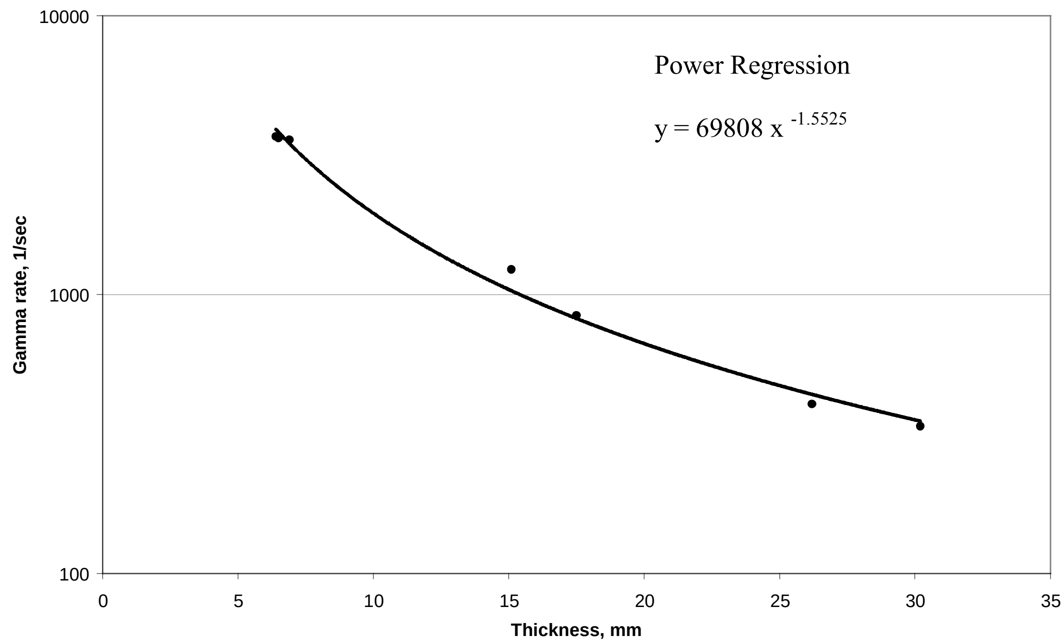

Fig. 2. Power regression for average count rate of the source gamma-rays, constructed to fit seven experimental points

In terms of the above designations power regression for the shell thickness is given by:

$$F_4(\Delta) = 69808\ \Delta^{-1{,}5525}\ ,\ [\Delta] = \text{mm}. \qquad (9)$$

In Table 3 there are presented values of mass and thickness of the eighth (reference) sample, which was not involved into the regression construction, computed using measured value of the correlated gamma-peak square from the equations of exponential and power regression:

Table 3

Factual thickness, mm	Exponential regression		Power regression	
	Calculation	Error	Calculation	Error
8,70	9,64	10,8%	8,64	0,7%

It is seen that power type regression being used gives much better approximation of the computed value to the factual thickness value.

In addition to this there has been considered two-parameter linear dependence of logarithm of the first gamma-peak square as a function of the shell mass and thickness.

$$F_5(M,\Delta) = \frac{1}{T}\sum_{\sim 0ns}^{\sim 3ns}[CC_{12}(\tau) + CC_{13}(\tau)]$$
$$Ln[F_5(M,\Delta)] \approx A_{5M} \times M + B_{5\Delta} \times \Delta + C_5 \qquad [F_5] = 1/\text{sec} \qquad (10)$$

Regression constructed using data for the same seven assemblies looks like:

$$LnF_5 \approx 6{,}1625 \times 10^{-5} \times M - 0{,}107586 \times \Delta + 8{,}644 \qquad (11)$$

In Table 4 there are presented values of mass and thickness of the eighth (reference) assembly, which was not involved into the regression construction, computed using diverse combinations of the measured signatures characteristics (the factual value of this assembly thickness constitutes 8,7 mm, value of mass is 2153 g).

Table 4

Signature characteristic to construct regression	Shell thickness		Shell mass	
	Calculation	Error	Calculation	Error
F_2, F_1	8,0	7,8%	2461,8	14,3%
F_3, F_1	9,64	10,8%	2297,6	6,7%
F_3, F_2	9,64	10,8%	2715,7	26,1%
F_4, F_1	8,64	0,7%	2399,6	11,4%
F_4, F_2	8,64	0,7%	2558,0	18,8%
F_5, F_1	9,1	5,0%	2349,0	9,1%

In the described investigations of a possibility of sample mass and thickness predicting either signatures of active measurements only were used, or signatures of both active and passive types together. When only passive signatures are used, the equation (1) can be taken as one of the equations and the second one can be obtained by constructing a linear regression for the detector signal autocorrelation function

$$F_6(M,\Delta) = \frac{1}{T}\sum_{i=1}^{i=4}\sum_{\tau=-50}^{\tau=50} AC_i(\tau) \approx A_{6M} \cdot M + B_{6\Delta} \cdot \Delta + C_6 \qquad (12)$$

Here $AC_i(\tau)$ is an autocorrelation function of i'th detector at the passive measurements.
The equations set for reference sample mass and thickness predicting has been obtained by results of measurements with six assemblies. Assembly № 2 has been excluded from consideration, as it has slightly broken spherical symmetry. In this case coefficients of the regression equation F_1, obtained earlier, when results for seven assemblies were used, slightly change (see (4)):

$$F_1' \approx 0{,}19999 \times M + 22{,}011 \times \Delta - 418 \qquad (4')$$

$$F_6 \approx 57{,}432 \times M - 6770 \times \Delta + 163945 \qquad (13)$$

In Table 5 there are presented results of mass and thickness computation of the seventh reference sample, which was not involved into the regression equation elaboration:

Table 5

Signature characteristic to construct regression	Shell thickness		Shell mass	
	Calculation	Error	Calculation	Error
F_6, F'_1	9,95	14,4%	2253,1	4,6%

Comparison between data from the Tables 3…5 shows that accuracy of the shell mass prognosis based on the passive measurements data only is ~ 2 times better, than that obtained using mixed data of active and passive measurements, and accuracy of the shell thickness prognosis on the contrary is much worse. The best accuracy of the shell thickness prognosis is achieved when data on the prompt source gamma-quanta passage in active measurements are used.

1.1. Containerized assemblies measuring.

In Table 6 there are presented results of mass and thickness evaluations based on the available data of passive measurements with assemblies placed inside AT400 container. To construct the regression equations the measured values of the detectors autocorrelation functions and cross-correlation functions between the detectors pairs have been used.

Table 6

Shell parameter	Factual values	Computed values	Error
Mass, g	2153,2	2996,3	39,2%
Thickness, mm	8,7	6,54	-24,8%

An accuracy of sample mass and thickness predicting for containerized assemblies is seen to be essentially worse than that for “bare” assemblies. Partially it may be due to reduction of number of reference points used to select coefficients for the regression equations (in the passive measurements assemblies № 6 and 7 from the list presented in Table 1 were not included into a container), partially to design of the AT400 container, having in its composition materials with considerable content of light elements (wood, polyethylene doped with boron), which results in noticeable neutron weakening and worsening experimental data statistical accuracy. Calculations carried out in ORNL [3] have shown that to improve measurements accuracy for strongly protected containers and to reduce exposition time large plastic scintillation detectors 50x50x10cm placed on the container opposite sides need to be employed in the passive measurements.

Above there are presented results of the simplest approach to parameters evaluation of plutonium assemblies to be studied (inspected), built upon the regression equations coefficients selection. For the same purposes more complicated mathematical approaches may be used, e.g. based on the neuron-net method. At VNIIEF the first attempt has been made to apply this method to evaluate parameters of plutonium assemblies the same as presented in the report and on the base of results of the same measurements. This work was encouraged by the results obtained in the report [4].

It may be mentioned that, when experimental data obtained for the same “bare” assemblies both in active and passive measurements are used to “train” a neuron net, accuracy of “unknown” assembly parameters predicting may constitute 1-2 %, what is essentially better, than that provided by the regression approach.

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