

The Moisture Content Monitoring Device for PuO₂ Using Self Neutron Radiation

Valeriy I. BULANENKO, Victor SVIRIDOV, Vladimir V. FROLOV,
Boris G. RYAZANOV and Vladimir V. TALANOV

*Federal State Unitary Enterprise State Scientific Center of the Russian Federation –
Institute for Physics and Power Engineering named after A.I. Leypunsky,
Bondarenko sq.1, 249033, Obninsk, Kaluga region, Russia*

Solutions technology of plutonium dioxide powders production inevitably leads to free or chemically bound hydrogen to be present in these powders.

This work is devoted to the nondestructive method of PuO₂ powder moisture measurement based on application of the effect of neutron moderation caused by water.

Plutonium dioxide is fast neutron source, while ³He counters located in the nickel and polyethylene annular reflectors surrounding PuO₂ serve as detectors.

In the work wide range of issues are considered related to practical implementation of the moisture measurement method by detecting inherent neutron radiation of plutonium dioxide powder. The most practical design of the detector has been chosen, which include two ³He detectors having different reflectors mounted to the device. The absolute error of measurement does not exceed 0.2 wt % with confidence coefficient of 0.95. Duration of analysis ~ 5 minutes.

KEYWORDS: *criticality safety, plutonium oxide powder, moisture control, non-destructive assay*

1. Introduction

At present one of the issues of high priority consists in developing technologies of Pu useful application in national economy, first of all in nuclear power. The technologies of weapons grade metal plutonium conversion into PuO₂ and NPP reactor fuel fabrication on its basis are becoming the leading direction in solution of this problem. This fact results in the necessity to fulfill a set of practical tasks on ensuring safety and quality of technological processes. Water technologies for Pu reprocessing and refinement are the mostly well developed and promising. They ensure a high quality of PuO₂ powder as a material for nuclear fuel and its good purification from ²⁴¹Am accumulated during decades of storage and from other alloy additions.

In view of the use of water technologies the objectives to assure nuclear safety have become very topical. One of the parameters that needs control is PuO₂ residual moisture. Moisture measurements allow the control of both criticality safety conditions and quality of technology, e.g. calcinations procedures after which the powder must have the minimum moisture content ($\leq 1\%$ mass) and concentration of other impurities which could cause loss of ignition.^{1,2)}

There are some other arguments in favour of moisture NDA, for instance, the necessity to improve the accuracy of Pu inventory mass determination in MBAs.²⁻⁷⁾

Plutonium refers to the category of especially harmful toxic substances. For this reason and for the

reason of Pu leak prevention it is extremely undesirable to open containers with Pu and to take samples. And it is quite obvious that moisture NDA of PuO₂ powder in containers should be prompt and timely, automated and highly sensitive.

2. Analytical relations

Neutron moisture meters are known to be widely used in various industries of national economy, including uranium powder production technologies.⁸⁾ The method sensitivity gets higher if the container with UO₂ powder is surrounded with nickel or iron which are fast neutron inelastic scatterers.^{1,8)} The reflectors like these send part of fast neutrons back to the container but with lower energy, thus increasing the effect of their hydrogen moderation.

PuO₂ has its own intensive neutron radiation: $\sim 10^5$ n/s per 1 kg PuO₂ of low burn-up and $\sim 10^6$ n/s per 1 kg PuO₂ of high burn-up. So in order to control moisture with an external radio isotopic source the neutron yield should be within the range of $\sim (10^7 \div 10^8)$ n/s.¹⁾ This fact causes some problems related to personnel protection against fast neutrons and to measurements in nuclear material accounting system. Due to the own high background of fast neutrons PuO₂ moisture can be measured without any external fast neutron source.

Let us first consider a moisture meter with a fast neutron source of ²⁵²Cf type, whose design with metal reflector was proposed in.¹⁾

The analytical signal for the initial moisture range is expressed by the linear function

$$N = N_o \cdot (1 + K \cdot W) \quad (1)$$

where N – is the count rate being measured, pulse/s; W – is PuO_2 moisture, wt %; N_o – is the count rate at $W = 0$ wt %, which is a calibration parameter of the moisture meter; K – is the proportionality factor which represents the slope of calibration parameter or sensitivity of the method $(dN/N_o)/W$, it is independent of the source neutron yield.

In this option the moisture meter has a number of drawbacks:

1. A high fast neutron yield is required from the ^{252}Cf point source.
2. A drastically non-uniform fast neutron field is formed in PuO_2 powder.
3. The effects of powder bulk density and mass become significant. They require an additional correction of the signal being measured.

There is another moisture meter option, which is more sensitive to moisture. In this option the PuO_2 own neutron radiation uniformly generated within the volume is used instead of ^{252}Cf . In this case the analytical moisture signal is generated additively by three effects:

- * The neutron spectrum gets softer and the detector efficiency grows.
- * (α, n) -reaction yield grows due to the increase in oxygen weight fraction.
- * The neutron multiplication grows due to additional elastic neutron scattering on hydrogen nuclei.

Numerical values of each effect at the registration of double neutron coincidences (doubles) for the HLNCC-II detector with a polyethylene moderator are given in.³⁾ The dominating contribution comes from the increase in the (α, n) -reaction yield.

The own fast neutron yield can vary and depends on PuO_2 mass in the container, Pu isotopic composition, ^{241}Am fraction and light element impurities in PuO_2 . In this case equation (1) does not meet the assay conditions because of N_o variations. One of the ways to eliminate this methodological error consists in introducing the second detection unit, independent of the first one, thus resulting in two-parametric moisture assay. So detector D_1 (nickel reflector) and detector D_2 (polyethylene reflector) are independent and the container with PuO_2 is loaded into them successively.

The operation of such a “combined” moisture meter can be described by two equations:

$$N_1 = N_{o1} \cdot (1 + K_1 \cdot W) \quad (2)$$

$$N_2 = N_{o2} \cdot (1 + K_2 \cdot W) \quad (3)$$

similar to (1).

Let us get the ratio

$$\frac{N_1}{N_2} = \frac{N_{o1}}{N_{o2}} \cdot \frac{1 + K_1 \cdot W}{1 + K_2 \cdot W}, \quad (4)$$

in which the ratio (N_{o1}/N_{o2}) does not depend on fast neutron yield variation in PuO_2 powder and is measured in the course of calibration tests.

Detectors D_1 and D_2 should be designed in such a way that $K_1 \gg K_2$. With this condition and due to a low value of moisture W equation (4) is modified in the following way

$$\frac{N_1}{N_2} = \frac{N_{o1}}{N_{o2}} \cdot [1 + (K_1 - K_2) \cdot W] \quad (5)$$

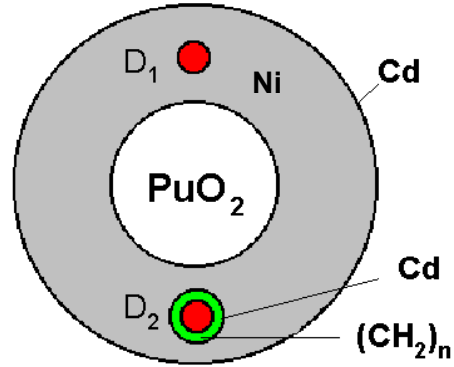


Fig.1 “Joint” detection unit design for moisture meter.

Detector D_2 could be designed so that K_2 would have a negative value. In this case the calibration curve slope (sensitivity) would increase $(K_1 + |K_2|)$. In this option the moisture meter is assumed to consist of two detectors with opposite responses to PuO_2 powder moisture. However detector D_2 cannot be considered as PuO_2 fast neutron yield monitor because this detector, similar to detector D_1 , also responds to the effect of fast neutron count rate growth due to their multiplication and the growth of (α, n) -reaction neutron contribution with the increase in moisture.

The two options considered above can be presented in the form of one “joint” configuration shown in fig.1. In this variant signals N_1 and N_2 are registered at the same time and with one operation of PuO_2 , container loading-unloading, thus reducing the time of one measurement and making its automation simpler. However, when combining detectors D_1 and D_2 the main effect (fast neutron moderation) can decrease.

3. Detection unit characteristics.

The main objectives of the study are to optimize the principal parameters of moisture meter detector and to determine the calibration curve parameters with estimation of influencing factors.

Evidently the most complete information about the dependence of information signal (assay parameter) on moisture can be only obtained by means of calibration procedure with the use of real PuO₂ powder samples. If considered in full scope this procedure is quite a labor-consuming task because it requires preparation of samples as reference materials with certified characteristics of moisture, Pu isotopic composition and Pu mass.

With all these factors in view, metal plutonium in the form of standard discs of the BFS-1 type was chosen for test modeling as an object of assay. These discs have the diameter of 46.7 mm and the height of 3.5 mm. Their external clad is made of stainless steel and has the thickness of 0.3 mm. Each disc contains ~ 52.5 g Pu. The total Pu mass in the container was equal to $m = 2534.4$ g Pu, and neutron yield was ~ 1.178×10^5 n/s, with the multiplication not taken into account.

Water was simulated with thin polyethylene films in the form of circles with diameters equal to disc diameter. The number of polyethylene circles was chosen from the condition of moisture range from 0 to 5 wt %. The equivalent moisture was calculated relative to Pu mass in view of different hydrogen concentration in water and polyethylene. In order to simulate different density of materials the aluminum discs were used with the diameter of 46.5 mm and the height of 5 mm.

The object under measurement consisted of 4 columns placed close to one another in a cylindrical cup with a cover. The following variants of stacking the container were studied:

- The high-density container with the average density ~ 5.3 g Pu/cm³. The polyethylene film was placed between each two Pu discs.
- The basic container with the average density ~ 3.1 g Pu/cm³. In each column between two BFS-1 discs one aluminum disc was placed.
- The low-density container with ~ 2.2 g Pu/cm³, with the double number of aluminum discs.

The set of these containers made it possible to simulate various characteristics of the object being measured (Pu density, moisture, arrangement geometry, etc).

In the experiments 3 options of detection units were studied:

- * A - "basic" detection unit D_1 , without any external neutron source.
- * B - "combined" detection unit, i.e. combination of two detectors, D_1 and D_2 .
- * C - "joint" detection unit shown in fig.1.

In all the moisture meter options SNM-18 neutron counters filled with ³He up to 4 atm, with the diameter of 32 mm and active part length of 270 m, or SHM-17 counters filled with ³He up to 7 atm, with the diameter of 18 mm and the length of its active part of 160 mm, were used as detector D_1 .

The "basic" detection unit with the nickel reflector (option A) had the outer diameter of 464 mm and the height of 480 mm. The measurement cavity had the diameter of 132 mm and the height of 280 mm. The group of 4 SNM-18 counters was uniformly distributed in the holes on the circumference with the radius of 90 mm, with the nickel layer thickness of 8 mm in front of the counter surface. And one SNM-17 counter was placed on the circumference with the radius of 86 mm. The second detector D_2 for the "combined" moisture meter (option B) had the same design, but its reflector was polyethylene and the outer diameter was reduced to 250 mm.

In the "joint" detection unit (option C, fig.1) the compound counters D_2 were located on the circumference with the radius of $R_1 = 150$ mm or $R_2 = 200$ mm. Each counter had the external layer of organic glass of various thickness d . Its outer surface can be used to locate a layer of sheet cadmium on it with the thickness of 0.4 mm (types 1 ÷ 4). The registration efficiency of these compound counters has different dependence on energy and responses in a different way on hydrogen content as part of the measured object.

The counters characteristics were determined with ²⁵²Cf neutron source and coaxial moderator made of organic glass, with both being located in the measurement cavity.

Table 1 Characteristics of counters for the "joint" option of moisture meter.

D_1		$No_1^{*})$	$\Delta N/No_1^{*})$	$No_1^{*})$	$\Delta N/No_1^{*})$
		291.8	3.10	285.6	3.15
D_2	d (mm)	$No_1^{*})$	$\Delta N/No_1^{*})$	$No_1^{*})$	$\Delta N/No_1^{*})$
		$R_1=150$ mm		$R_2=200$ mm	
1	—	90.9	1.077	52.4	0.819
2	10	1726.4	0.050	1023.1	0.015
3	15	2963.9	- 0.015	1753.2	- 0.021
4	25	6583.7	- 0.041	4017.0	- 0.094
*) The relative error < 2 %					

Table 1 presents 2 sets of measurements for different types of detectors D_2 , located at the distances R_1 and R_2 . The obvious increase in the registered count rate of the ³He counter is caused by the reduced average energy of moderated neutrons. When the distance between the detectors D_2 and measurement cavity goes up, the registered neutron flux goes down. At the same time compound detector present in the reflector introduces local perturbation

into the neutron field, thus resulting in the decrease in the average neutron energy and corresponding increase in D_1 readings by (20÷50) %, if D_1 detectors are at the distance less than 80 mm from the D_2 .

The D_2 detector quality criterion is the ratio of effect/background, i.e. the ratio of $[S - N_o] / N_o$, where S – is the signal in the presence of moderator, and N_o – is the signal only from the neutron source. Pay attention to the negative value of the effect/background ratio for the compound detectors of type 3 and 4.

So Table 1 shows that for the “joint” option of moisture meter the use of compound counters (type 3 and 4) is more preferable. In this case the D_2 detector can be placed as it is shown in fig.1 or outside the nickel reflector as an additional mending plate.

4. Study of moisture meter response functions.

Table 2 and fig.2-3 illustrate some results of model measurements: count rate for different detectors and detectors’ reading ratio: D_1 (SNM-18 or SNM-17) and D_2 (SNM-17; type 2, R=150 mm).

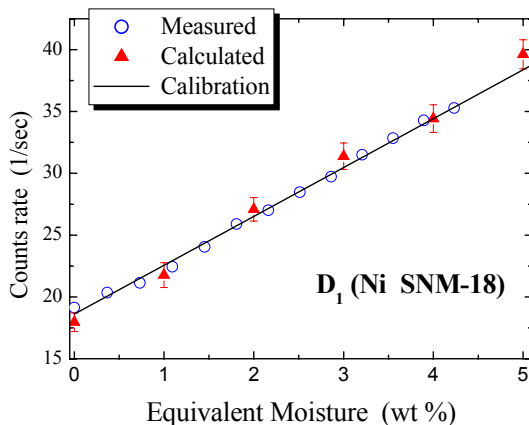


Fig.2 Count rate in nickel reflector as a function of equivalent moisture of “basic” container

The indicated data confirm the initial points of moisture assay technique stated earlier. In particular, within the studied range of equivalent moisture values (0 ÷ 5) wt %, the functional dependence of analytical signal is linear and follows equation (1).

It should be stated that the experimental and calculated results with MMKFK-2⁹⁾ show a good agreement that is illustrated in fig.2. The calculated values indicated here are normalized on the value of equivalent moisture of 4 wt %.

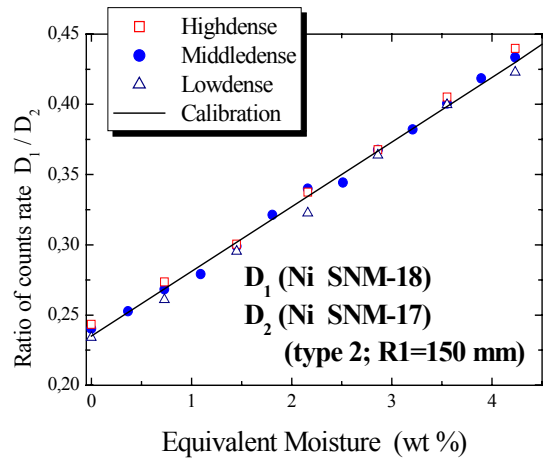


Fig.3 Count rate ratio as a function of equivalent moisture for “joint” moisture meter at different density of the object under measurement.

The effect of density of the measured objects is actually reduced only to the shift of measured count rates because the slope factors practically coincide. The ratio of both detectors readings N_1 / N_2 does not depend on Pu density in the objects under measurement (fig.2).

Table 2 presents the calibration curve parameters for two options of moisture meters (A and B), when count rate is the information parameters, which is measured.

Table 2 Control parameters for moisture meter options A and B.

#	Detector	N_o (1/s)	K
1	D_1 (Ni, SNM-18)	18.60 ± 0.14	0.213 ± 0.005
2	D_1 (Ni, SNM-17)	8.57 ± 0.13	0.241 ± 0.010
3	D_2 (Ni, SNM-17; type 2, $R_1=150$ mm)	79.86 ± 0.32	0.0067 ± 0.0017
4	D_2 (Ni, SNM-17; type 4, $R_2=200$ mm)	213.7 ± 1.0	-0.0088 ± 0.0021
5	D_2 (Polyethylene, SNM-17)	478.4 ± 3.5	-0.0168 ± 0.0030

As the measurement results similar to those indicated in table 2 were obtained in the course of independent measurements all the data were processed in order to be able to compare the relative values of measured parameters and summarized in table 3. These data show that the two-parametric method of moisture assay by means of D_1 and D_2 reading ratio provides a rather high sensitivity of moisture assay.

Tables 2 and 3 illustrate that when the SNM-17 counter is used the calibration curve slope (coefficient K) turns out to be higher by 10% as compared to the SNM-18 counter. These systematically overestimated

parameters of neutron moderation effect being observed with the SNM-17 counter encouraged additional studies of the given difference. For this purpose the radial count rate distribution, N_o and effect/background ratios $[S - N_o] / N_o$ were measured depending on the nickel thickness between the measurement cavity and counter surface.

Table 3 Control parameters for “combined” and “joint” moisture meter.

Analytical signal: count rate ratio.

#	D ₁	D ₂	N _{o1} /N _{o2}	K
“Combined” meter (option B; D ₁ ⇒Ni; D ₂ ⇒CH ₂)				
1	SNM-18	Polyethylene CH ₂	0.0387 ± 0.005	0.243 ± 0.008
2	SNM-17	Polyethylene CH ₂	0.0178 ± 0.0003	0.276 ± 0.013
“Joint” meter (option C; only Ni)				
1	SNM-18	Ni; type 2, R ₁	0.2337 ± 0.0020	0.200 ± 0.005
2	SNM-18	Ni; type 4, R ₂	0.0871 ± 0.0080	0.226 ± 0.006
3	SNM-17	Ni; type 2, R ₁	0.1078 ± 0.0017	0.228 ± 0.010
4	SNM-17	Ni; type 4, R ₂	0.0400 ± 0.0007	0.256 ± 0.011

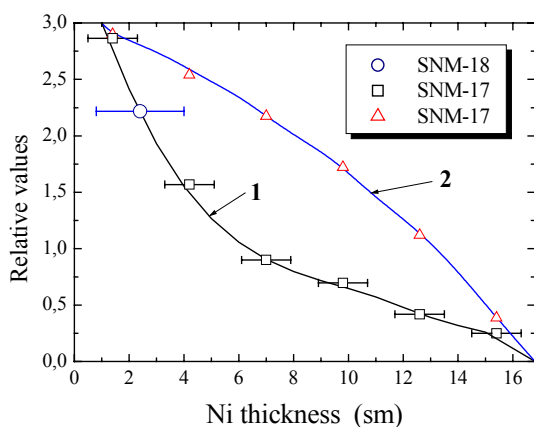


Fig.4 Ratio dependence: effect/background (1) and background (2) as a function of neutron detector location in the nickel reflector

Fig.4 shows the radial distribution of the dependence of both signals on the nickel thickness between the measurement cavity and outer SNM-17 surface. The counter moved with the step of 28 mm. A cylindrical rod made of nickel was installed in the blind opening instead of the counter. The horizontal lines indicate the counters’ diameters within which the additive counter signal is generated. The background signal N_o is seen to be a monotonous function, which smoothly dies out to the reflector periphery. At the

same time, the dependence for the effect / background ratio has a different pattern, because the moderator effect manifests itself first of all near the measurement cavity. So with the increase in the counter diameter the relative contribution of useful signal will be less as it is observed for the SNM-18 counter.

5. Discussion of the results.

The specific yield of PuO₂ own neutron radiation exceeds 10⁵ n/s per 1 kg PuO₂. This intensity of own neutrons allows the moisture assay by the methods of neutron radiometry.

The main element of a neutron moisture meter is a well-type detector with a nickel reflector, which provides the required sensitivity of PuO₂ assay.

The presented model studies show the principal possibilities to implement the following moisture meter options:

- A. Moisture meter with one detection unit;
- B. “Combined” option of two detection units;
- C. “Joint” detector option.

For the A option of moisture meter the analytical moisture signal can be expressed as a linear function in the form of (1). The N_o value for each measured container is not measured but only corrected according to the Pu passport data. All the three above – mentioned effects caused by the PuO₂ moisture make an additive contribution to the method sensitivity. So in order to implement the first option the product passport data must be known, i.e. PuO₂ mass Pu, isotopic composition and ²⁴¹Am fraction.

For the “combined” option the analytical moisture signal is expressed as a linear function of type (5). It is a ratio of two detectors’ readings, D_1 (nickel reflector) and D_2 (hydrogen-containing reflector). For this option the analytical signal is primarily related to fast neutron hydrogen thermalization effect. In this case neutron multiplication and (α ,n)-reaction neutron contribution do not actually increase the assay sensitivity and the moisture assay results do not depend on these effects.

The advantage of the “combined” moisture meter consists in assay independence on Pu isotopic composition and its mass, i.e. there is no need to introduce the correction of the parameter N_{o1} / N_{o2} for PuO₂ powder in each measured container. In other words, the N_{o1} / N_{o2} readings ratio is a meter calibration constant. Along with that this type of moisture meter has a drawback, which consists in the fact that it requires twice as much time to perform two consecutive measurements, thus making the measurement procedure automation more complicated.

The “joint” option has all the advantages of the “combined” option. In this case N_1 and N_2 signals are registered simultaneously during one operation of

loading – unloading the cup with PuO₂ powder. So the “joint” option does not have any drawbacks indicated above and typical of other moisture meter options. In view of that the “joint” option is more preferable to control Pu moisture.

The experimental and calculation model studies performed with metal plutonium have shown that all the types of moisture meters provide practically the same assay sensitivity. The particular value of proportionality coefficient K is determined by the type of counters being used and is equal to 0.20÷0.27 at the moisture of 1% mass.

Let us estimate the expected moisture assay error. For the N parameter (count rate or count rate ratio) the moisture is equal to

$$W = \frac{N/N_o - 1}{K} \quad (6)$$

Fig.5 shows the example of calculated dependence of the assay relative error (curve 1) on PuO₂ moisture (with the mass of 3.0 kg). The option of two-parametric assay with the “joint” design of moisture meter has been considered. During the estimation consideration was given to the increase in (α,n)-neutron yield of the wet PuO₂ powder.

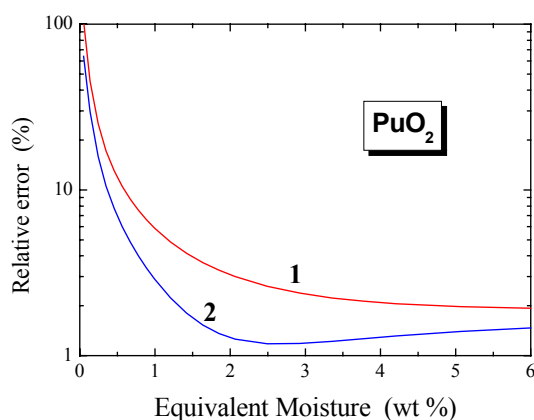


Fig. 5 Relative error of PuO₂ moisture assay:

- 1 – the error calculated with equation (5);
- 2 – the total error, which only considers the uncertainty of curve parameters.

The uncertainty of calibration curve parameters for the well-certified reference materials gives the dependence (curve 2) indicated in fig.5. Actually this function shows the limit of the assay accuracy achievable by the method. The lower assay limit is less than 0.1 wt % and the moisture higher than 1.0 wt % can be measured with the relative error better than 6%. The absolute value of moisture measurement is ~ 0,1 wt % and is practically constant up to the moisture value of wt 2%. It is determined by the volume of

statistical information being accumulated. At higher moisture value the uncertainty of calibration curve parameters starts to play the determining role. Due to that the absolute value increases twice at the moisture of 5 wt %.

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