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Reference Material Manufacture and Certification for the AVNG

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ABSTRACT

Testing and demonstration of any radiation measurement system requires the use of appropriate radioactive sources. The AVNG implementation that we describe is an attribute measurement system built by RFNC - VNIIEF in Sarov, Russia. The AVNG detects neutron and gamma radiation signatures and displays the three unclassified attributes of "plutonium presence," "plutonium mass > 2 kg," and "plutonium isotopic ratio (^{240}Pu to ^{239}Pu) < 0.1."

The AVNG was tested using a number of reference material (RM) sources with masses and isotopic ratios above and below these thresholds. The AVNG was demonstrated in June 2009 using several of these sources in addition to detector calibration sources. Since the AVNG was designed to measure multi-kg plutonium sources, the RM was manufactured specifically for use with this system. In addition, the RM was used to test the thresholds in the AVNG, so the size and composition of each RM was certified prior to use. In this presentation, we will describe the various steps in the manufacture and certification of these RM sources.

INTRODUCTION

AVNG (Attribute Verification Systems with Information Barriers for Plutonium with Classified Characteristics utilizing Neutron Multiplicity Counting and High-Resolution Gamma-ray Spectrometry – or in Russian, ПАНГ) is an attribute measurement system that was built as part of a cooperative effort between the Russian and American federal scientific agencies. The AVNG was built by The All-Russian Research Institute for Experimental Physics at the Russian Federal Nuclear Center (RFNC-VNIIEF) in Sarov, Russia, and it is the first example of an attribute measurement system with an information barrier designed and built in Russia.

The RFNC-VNIIEF (Российский Федеральныйй Ядерный Центр - Всероссийский Научно-Исследовательский Институт Экспериментальной Физики), like Los Alamos National Laboratory, was founded in 1946 for the production of nuclear weapons. Today its primary mission is to ensure the safety and reliability of nuclear weapons in Russia.

The AVNG is designed to measure the composition of sensitive nuclear materials behind an information barrier and to display only the unclassified and agreed upon attribute properties.

The AVNG measurements are based on neutron multiplicity counting and high-resolution gamma spectroscopy. The pre-defined attributes that are reported by the AVNG are:

1. Whether plutonium is present in the material,
2. Whether the plutonium mass is in excess of 2 kg, and
3. Whether the $^{240}Pu/^{239}Pu$ ratio in excess of 10%.

The AVNG measurement system is discussed in detail elsewhere in this and previous volumes (e.g. Pucket et al., 2001, Budnikov et al., 2005, and Smith et al., 2010).

Since the AVNG was designed to measure high-enrichment, multi-kilogram plutonium sources, it was necessary to manufacture standard reference materials (SRMs) specifically for the testing, calibration and demonstration of the system. In total, four such SRMs were created and certified by the Russian Federal Agency on Technical Regulation and Metrology (FATRM, Федеральное Агентство по Техническому Регулированию и Метрологии). The SRMs contain plutonium dioxide (PuO_2) with plutonium masses and isotopic ratios which cover a range above and below the pre-defined attribute thresholds. The AVNG was successfully tested and demonstrated in June 2009 using the specifically developed SRMs. This paper describes the various steps in their manufacture and certification.

The development and manufacture of the SRMs can be broken down into the following steps:

1. Determination of basic material requirements
2. Development of procedures and containers for the safe transportation and handling of sensitive material
3. Manufacture of appropriate PuO_2 mixtures for use in the SRMs
4. Analysis of the reference materials to determine certified measurement values
5. Non-destructive acceptance testing of the SRMs to verify the material content
6. Final assembly of the SRMs
7. Certification by FATRM

MATERIAL REQUIREMENTS

A total of four PuO_2 SRMs were manufactured covering a range of isotopic ratio and plutonium mass. The major constituent requirements including total plutonium content, the isotopic mass fractions of ^{239}Pu and ^{240}Pu, and the allowable $^{240}Pu/^{239}Pu$ isotopic ratio, are listed in Table 1. The allowable range for the mass fraction of plutonium in each of the PuO_2 mixtures was 86-89%. The total concentration of impurities in the PuO_2 powder was limited to 0.2% and the fluorine and chlorine concentrations were each capped at 0.005%. It was anticipated that the relative content of ^{241}Am must be determined and reported for a specific assay date. Each plutonium mixture must be of homogeneous isotopic composition. It was also determined that the loss of plutonium dioxide powder during calcination of the plutonium oxalate (see the section on Manufacture) must not exceed 2%.

Table 1. Basic requirements for the set of Standard Reference Materials produced for testing and demonstrating the AVNG.

	Pu №0.07-11	Pu №0.07-12	Pu №0.25-21	Pu №0.1-31
$^{240}Pu/^{239}Pu$	0.07 ± 0.01		0.25 ± 0.02	0.10 ± 0.01
^{239}Pu (%)	91.5 ± 1.5		70 ± 3.0	86.5 ± 1.5
^{240}Pu (%)	6.6 ± 0.6		17.2 ± 1.3	8.65 ± 0.45
Total Pu (g)	1000 ± 5	3500 ± 5	1000 ± 5	2000 ± 5

The SRM set required the production of three different plutonium mixtures. One SRM is produced from each of the materials relatively low in fissile nuclides ($^{240}Pu/^{239}Pu$ ratios of 0.25 and 0.1), and the high-enrichment mixture ($^{240}Pu/^{239}Pu$ ratio of 0.07) is used in the production of two SRMs with different total mass contents.

The shelf life of the SRM set is 10 years, taking into account the half-life of the fissile materials. The plutonium and ^{241}Am concentrations must be adjusted for decay over the 10-year lifetime. Any certified error values must also be adjusted over that period to account for errors in the values of the isotope half-lives.

HANDLING REQUIREMENTS

The SRMs contain a highly controlled substance (special nuclear material) with fairly high concentrations of fissile material. Therefore, the production and handling of the SRMs involved elevated concerns for material security as well as the basic requirements for the security, safety, transportation, storage and labeling of radioactive materials. It was necessary to design and construct appropriate containers for the storage and transportation of the SRMs themselves as well as the analysis and archive samples.

The final SRM containers (Figure 1) were constructed of stainless steel with constrained limits on the allowable wall thickness (3mm side walls and 4mm bottom). Containers were designed with an internal vessel containing PuO_2 powder with a screw-top and metal gasket. The outer tube, or housing, is sealed with argon arc welding. To prevent the outer tube from losing its seal and radioactive substances potentially entering the environment, opening or deforming the shell of the tube is prohibited, as is exposing it to corrosive media not specified in the technical documentation. The SRM containers were leak tested after being loaded with the PuO_2 powder. Each measurement standard has its own set of design documents. Each standard has an outer diameter of about 11cm. However, the heights varied depending on the mass of plutonium oxide and range between 17 and 32cm. Care was taken to eliminate any contamination on the outside of the containers.

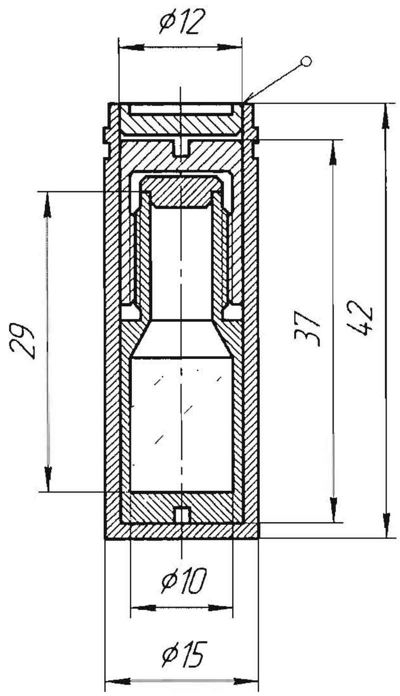

Figure 1. Diagram of the SRM container (left) and actual photograph of a closed container (right). Measurements are in centimeters. Container walls are constructed from stainless steel.

Smaller containers were also created for storing and transporting archive and measurement samples.

It was necessary to obtain a permit to transport the SRMs to a separate facility for acceptance testing. The permit describes the packaging for shipping and other requirements. Each shipping container contains room for three transfer casks on the top and bottom levels. The shipping container cover is a welded structure formed by two steel cylindrical shells separated by heat insulating foam. The corresponding end pieces are also separated by foam.

MANUFACTURE

The plutonium dioxide mixtures for the SRMs were produced at the RT-1 plant of the Mayak Production Association (Производственное объединение Маяк) in Russia. Three different plutonium dioxide materials were produced with different isotopic ratios. Each mixture was a plutonium dioxide powder containing about 86% plutonium by weight.

The plutonium mixtures were manufactured from 6 VVER-440 and 2 BN-600 spent nuclear fuel assemblies. The VVER (Водо-водяной энергетический реактор, or Water-Water Energetic Reactor) is a 440 kW thermal power reactor. The BN-600 (Быстрых Нейтронах, or Fast Neutron) is a fast, sodium-cooled power reactor. Both reactors are fueled with uranium dioxide.

The fuel element processing was carried out according to standard procedures at the Mayak production plant. The processing included the dissolution of the spent fuel, isolation of plutonium, and deposition as an oxalate. The steps in the material manufacture can be summarized as below:

1. Fuel elements were cut, dissolved, and filtrated
2. Uranium, plutonium and neptunium were extracted from the fuel solution
3. Uranium was separated from plutonium (leaving a mixture of plutonium and neptunium)
4. Plutonium was separated from neptunium
5. Plutonium was purified to remove fission fragments
6. Plutonium oxalate was deposited out of the solution and calcinated to form PuO_2
7. PuO_2 was annealed and packed into three containers (one for each mixture).

The plutonium in the spent fuel from the fast reactor has relatively high $^{240}Pu/^{239}Pu$ ratio. To produce the SRM mixtures with the necessary range of isotopic ratios, fuel from the VVER-440 was processed first. Material was continuously sampled during the transition to the high-enrichment BN-600 assemblies. The mixtures with the desired enrichments were used for the SRMs. As a result of the purification processes, the PuO_2 mixtures were purified with respect to ^{241}Am at the time of production.

ANALYSIS

It was necessary to accurately characterize the SRM materials for certification. Each material was randomly sampled for both archiving and analysis. The subsamples were packed into small stainless steel containers. Two samples each with 1g of PuO_2 were reserved for archiving at the Mayak production facility and two 2-gram samples were reserved for analysis at Mayak. Ten 3-gram samples were reserved from each mixture for isotopic analysis. Isotopic analysis was performed at the Russian Federal Institute of Physics and Power Engineering (IPPE or Физико-Энергетическии Институт), a facility dedicated to the development of nuclear power technology and home of the world's first nuclear power plant.

The relative mass of plutonium in each PuO_2 powder was determined from separate measurements of the total powder mass and total plutonium mass. The powder mass was directly measured with certified balances and the plutonium mass was determined with potentiastatic coulometry. These measurements were carried out on analysis subsamples at Mayak.

The mass fractions of ^{238}Pu, ^{239}Pu, ^{240}Pu and ^{241}Pu and were determined by gamma spectroscopy. The elemental impurities, as well as the ^{242}Pu and ^{241}Am fractions, were determined with mass spectrometry. The Gamma Spectroscopy and Mass Spectroscopy measurements were carried out at IPPE. The certified values an arithmetic average of 10 independent measurements. The certified plutonium isotopic concentrations are given in Table 2 and the impurities are listed for each mixture in Table 3.

Table 2. Certified concentrations of the major constituents in the four Standard Reference Materials (SRM) created for testing and calibrating the AVNG.

	Pu №0.07-11	Pu №0.07-12	Pu №0.25-21	Pu №0.1-31
$^{240}Pu/^{239}Pu$[1]	0.072 ± 0.0002		0.254 ± 0.0005	0.107 ± 0.0002
^{238}Pu (%)[2]	0.954 ± 0.05		1.876 ± 0.09	3.679 ± 0.18
^{239}Pu (%)[2]	90.952 ± .09		70.639 ± 0.07	84.312 ± 0.08
^{240}Pu (%)[2]	6.522 ± 0.01		17.97 ± 0.03	9.022 ± 0.02
^{241}Pu (%)[2]	1.139 ± 0.02		5.698 ± 0.17	2.012 ± 0.07
^{242}Pu (%)[2]	0.434 ± 0.02		3.817 ± 0.15	0.975 ± 0.04
^{241}Am (%)[4]	0.11		0.79	0.46
Plutonium (%)[3]	286.02 ± 0.34		86.03 ± 0.34	86.04
Plutonium (kg)[3]	0.9937 ± .004	3.4770 ± .01	0.9990 ± 0.004	2.0063

[1]Absolute errors in the isotopic ratio are based on the propagated uncertainty of the ^{240}Pu and ^{239}Pu measurements

[2]The isotopic contents are given as the mass fraction of the plutonium. Absolute errors are based on the reproducibility of ten mass spectrometry measurements. The values are assayed on Jan 11, 2008

[3]The plutonium content is given as a mass fraction of the total PuO2 mixture as well as an absolute amount in each SRM. Absolute errors are reported at the 95% confidence interval. Measurements performed with certified balances and coulometry. The plutonium mass values are assayed on Oct 28, 2007.

[4]The isotopic contents are given as the mass fraction of the plutonium. The ^{241}Am content was assayed on Aug 20, 2007.

The independent measurements insured that the homogeneity of the mixtures was accurately characterized. The error in the certified values of isotopic fraction (δ) is calculated as

$$\delta = \sqrt{\delta_{rand}^2 + \delta_{meas}^2}$$ where $$\delta_{rand} = \frac{tS}{\bar{x}}$$, and

σ_{rand} = random measurement error at 95% confidence level

σ_{meas} = systematic error inherent in the measurement technique.

S = root mean square of the 10 independent measurements

t = Student's factor for 10 independent measurements with a confidence interval of 95%.

$\bar{x}$ = arithmetic average of 10 independent measurements.

Table 3. Certified concentrations of the minor constituents in the four Standard Reference Materials (SRM) created for testing and calibrating the AVNG.
All values are reported as a percentage of the total PuO2 mass.

	Pu №0.07-11,12	Pu №0.25-21	Pu №0.1-31
Al	9×10^{-4}	3×10^{-4}	10×10^{-4}
Ba	1×10^{-3}	1×10^{-3}	1×10^{-3}
B	3×10^{-5}	3×10^{-5}	3×10^{-5}
Be	1×10^{-5}	1×10^{-5}	1×10^{-5}
V	3×10^{-4}	3×10^{-4}	3×10^{-4}
Fe	6×10^{-3}	5×10^{-3}	12×10^{-3}
Ca	4×10^{-3}	1×10^{-3}	7×10^{-3}
K	1×10^{-3}	1×10^{-3}	1×10^{-3}
Co	1×10^{-3}	1×10^{-3}	1×10^{-3}
Si	3×10^{-3}	12×10^{-3}	3×10^{-3}
Mg	4×10^{-3}	14×10^{-3}	11×10^{-3}
Mn	3×10^{-4}	60×10^{-4}	3×10^{-4}
Na	1×10^{-3}	2×10^{-3}	1×10^{-3}
Ni	10×10^{-3}	14×10^{-3}	12×10^{-3}
Pb	3×10^{-4}	3×10^{-4}	3×10^{-4}
Cr	13×10^{-3}	8×10^{-3}	5×10^{-3}
Cl	$< 1 \times 10^{-3}$	$< 1 \times 10^{-3}$	$< 1 \times 10^{-3}$
F	0.83×10^{-2}	2.4×10^{-2}	12.7×10^{-2}
Sm/Cd/Gd	16×10^{-4}	16×10^{-4}	16×10^{-4}
Total impurities	0.05574	0.09254	0.18554

ACCEPTANCE TESTS

The purpose of the Acceptance Tests was to verify that the material characteristics of the SRMs match the certified values. The total mass of PuO_2 powder in the SRMs was determined during assembly from the difference in weight between the filled and empty SRM containers at Mayak.

Gamma spectroscopy was used during the acceptance tests to verify the plutonium isotopic ratios in each mixture. The spectroscopy was carried out with two high-resolution coaxial HpGe detectors and two planar HpGe detectors. This information was also used in combination with calorimetry measurements and neutron multiplicity counting to verify the relative mass of plutonium in each SRM material.

State certified plutonium standards from a previous contract with the Commission of the European Communities were used to test and calibrate the measurement systems. Additionally, three ^{252}Cf sources were purchased and certified by VNIIF for the same purpose.

Calorimetric measurements were only performed on samples No 0.07-11 and No 0.25-21 because the other two SRMs produced too much heat to be measured in the calorimeter system. Neutron multiplicity counting was complicated by the production of neutrons from (α,n) reactions with light-element impurities and the large dimensions of the SRMs. A combination of four different multiplicity counters was used such that each SRM was analyzed at least once.

ASSEMBLY

In actuality, the entire assembly process includes several of the production steps outlined above.

1. Cleaning and drying of the component parts of the SRM containers
2. Weighing of the SRM containers
3. Sealing PuO_2 in a temporary container with an argon environment
4. Homogenization of the PuO_2 powder in a tumbler
5. Sampling of the PuO_2 powder for archive and analysis
6. Packaging of the PuO_2 powder into the inner, screw-top bottle of the SRM container
7. Loading of the inner bottle into the external housing
8. Sealing of the housing with argon arc welding
9. Decontamination of the outer surface of the housing
10. Final weighing of the filled SRM container
11. Course leak testing of the housing
12. Seal testing of the housing with a helium leak detector
13. SRM containers were loaded into modified AT-400R storage containers and sealed

CERTIFICATION

The analysis results from the plutonium isotope fraction measurements and PuO_2 mass measurements were processed by the Ural Scientific Research Institute of Metrology (UNIIM, Уральский Научно-Исследовательский Институт Метрологии). UNIIM was also responsible for determining the certified measurement errors. Several documents were required as part of the certification process. These include:

1. Technical report on the measured SRM characteristics
2. SRM Certificate
3. SRM Labels
4. SRM Type Description
5. SRM Application guide

CONCLUSIONS

A specifically developed set of plutonium dioxide standards were successfully manufactured and certified for the testing, calibration and demonstration of the AVNG attribute measurement system. In total, four such SRMs were created and certified by the Russian Federal Agency on Technical Regulation and Metrology (FATRM). The AVNG was successfully tested and demonstrated in June 2009 using the SRMs.

The most labor intensive portion of the manufacturing process was in performing precise and accurate analyses of the standard mass and composition. However, the final certified values for the standards are well within the predefined requirements. In addition, the certified measurement errors are below the predefined limits for uncertainty.

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