PANWAS: A Passive/Active Neutron Waste Assay System for the Radiological Characterization of Waste Packages at the Nucleco Facility at Casaccia

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ABSTRACT

CANBERRA has recently supplied Nucleco SpA with a new Passive/Active Neutron Waste Assay System (PANWAS) for use at their waste management facility at Casaccia in Italy. The system complements two existing CANBERRA high-resolution gamma spectrometry waste assay systems. The three waste assay systems have been integrated into a combined facility for the radiological characterization of the waste managed by Nucleco in order to provide the information required to:

- Determine the physical inventory of the nuclear material present for Safeguards purposes,
- Segregate the waste into different categories,
- Allow transportation to and storage in the final repository for the waste.

This paper describes the main characteristics of the PANWAS, how it is used (in conjunction with the two gamma monitoring systems) to determine the radionuclide inventory of the waste and how the system was calibrated and characterized for use in this application.

INTRODUCTION

Nucleco SpA is responsible for the management of radioactive waste produced in Italy by industrial research and medical processes. It operates a facility at Casaccia near Rome for the storage and treatment of low level and intermediate level waste in order to make it suitable for transport and final disposal.

Nucleco required a waste assay system for the measurement of a wide range of materials with different process histories to assess inventories of safety and Nuclear Safeguards relevant nuclides consisting mainly of uranium, thorium and plutonium.

The new monitor was designed to complement two existing gamma-based assay systems. It measures passive neutrons produced by spontaneous fission and neutrons from actively induced fission in the nuclear material present in the waste. The requirements for the integrated assay system are to identify and quantify specific radionuclides of interest for Safeguards purposes and for the safe transport and disposal of the waste.

THE REQUIREMENT

The items to be assayed consist of contact handleable waste in 200 liter and 400 liter drums as well as non-contact handleable waste in lead shielded *TSR* type transport and storage flasks. The PANWAS was designed to measure fissile content while taking the following factors of the waste form into account:

Highly random non homogeneous matrices,

- Wide range of matrix density variation (both compacted and uncompacted and also with and without cement added for the immobilization of the waste,
- Non uniform distribution of the activity,
- Containers of variable shape and dimensions,
- Great variety of radionuclides.

In order to meet Safeguards requirements, it is necessary to determine the mass inventory of the following:

- Natural uranium,
- Depleted uranium,
- U-235,
- Thorium,
- Plutonium.

The diverse waste assay capability provided by a combined passive/active neutron monitor was required to be combined with the assay results from the two existing high resolution gamma spectrometry based assay systems already supplied to Nucleco by CANBERRA. These two systems consist of a fully automated segmented gamma scanner (SGS) for the assay of contact handleable 200 liter drums and an in-situ object counting system (ISOCS), with associated modeling software, used for the assay of gamma-ray emitters in 400 liter drums and 200 liter drums that exceeded the weight and/or gamma-ray dose restrictions of the SGS.

A further requirement is for the combined waste assay system to be fully integrated using a single data repository and with the capability to combine the assay results from the different instruments into a single assay report (e.g. to use the measured isotopic ratios from the ISOCS system along with the Pu-240 effective mass from the PANWAS to calculate the total plutonium mass.

SYSTEM DESCRIPTION

The PANWAS uses passive neutron coincidence counting to determine the quantity of spontaneous fission radionuclides (Pu-238, Pu-240, Pu-242 etc.) in the waste. It uses the active neutron differential die-away (DDA) technique [1],[2] to quantify the fissile content (U-235, Pu-239 etc.). The system consists of a single graphite lined rectangular assay chamber (see Fig. 1) with a common set of cylindrical He-3 filled proportional counters which are used to detect neutrons in both passive and active mode.

There are a total of 117 He-3 detectors (each manufactured from stainless steel with 50mm diameter × 1000 mm active length, 4 atmosphere pressure of He-3) located in cadmium shielded polyethylene moderator assemblies in three of the four sides of the chamber (the left side, right side and the door at the front of the chamber). There are no detectors in the rear of the chamber, where the neutron generator tube (used for DDA measurements is housed, nor in the chamber roof and base.

An automated drum transfer mechanism is used to load a waste drum into the assay system onto a rotator mechanism built onto the inside of the chamber door. The rotator mechanism is used to turn the drum during the assay measurement to take into account (by partitioning the count for use in algorithms) any inhomogeneities in the fissile material or matrix distribution.

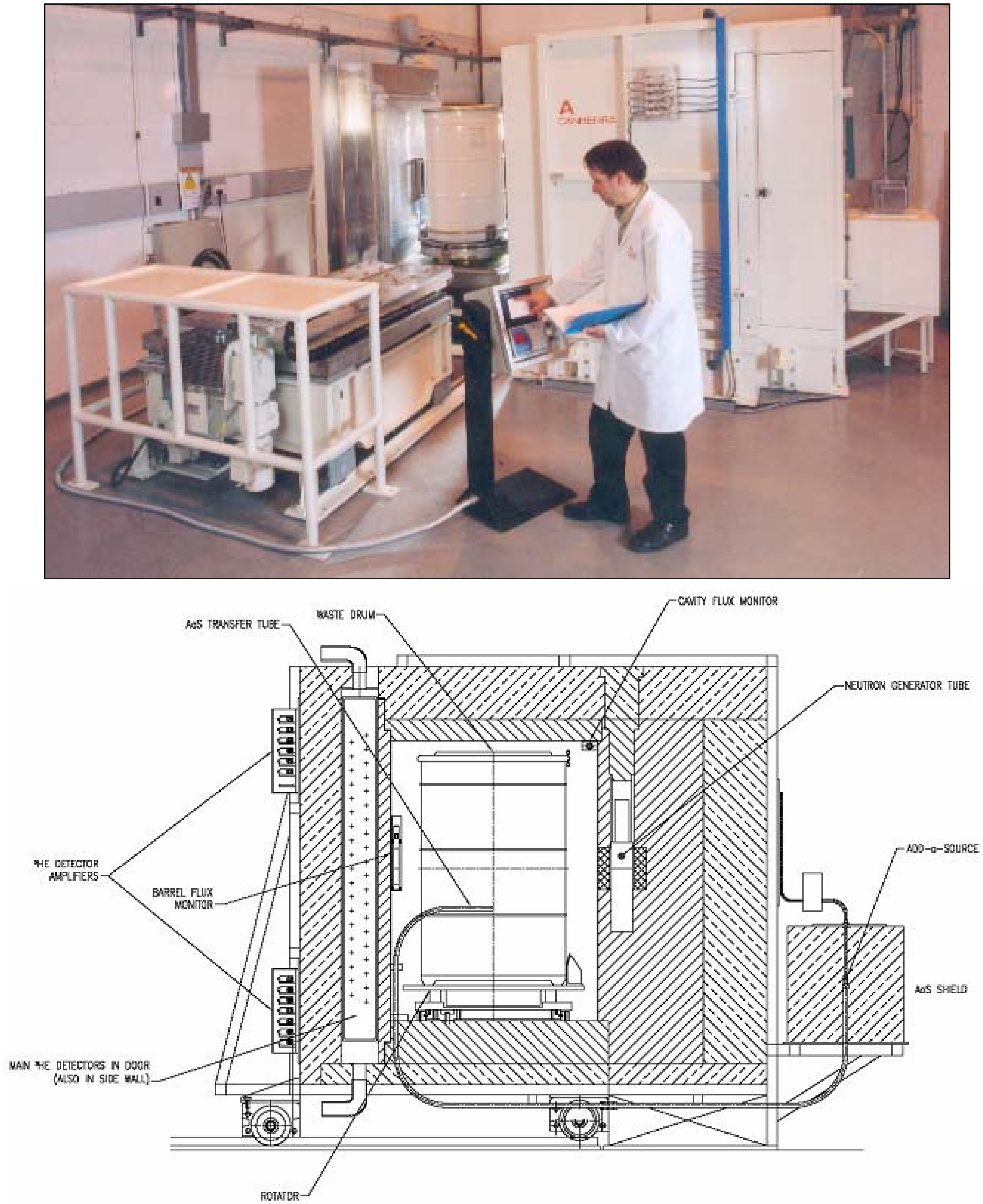

Fig. 1. Photograph and cut-away side view of the PANWAS System

The Add-a-Source (AAS) technique [3],[4],[5] is used to correct for the effect of the waste matrix on the passive neutron measurement. The AAS technique makes use of a small (10^5 n.sec^{-1}) Cf-252 neutron source that is driven into the chamber into a position near the waste drum during the assay measurement. The perturbation of the neutron count rate from the Cf-252 caused by the waste matrix (relative to that for an empty drum) is used to infer the effect of the matrix on the neutron signal from any fissile material in the drum. The AAS Cf-252 source is normally stored in a polyethylene shield at the rear of the chamber. At the appropriate point in the assay process, it is driven on the end of a Teleflex™ cable along a guide tube to a position alongside the waste drum on the rotator. The AAS method has been found to have significantly improved accuracy over alternative Passive Neutron Coincidence Counting (PNCC) matrix correction techniques that rely on the passive neutron signal from the waste, which can be affected by source location and the presence of (α, n) neutron emitters.

The source of pulsed neutrons for the DDA measurement is a Zetatron type neutron generator that makes use of the deuterium-tritium (D-T) reaction to produce 14 MeV neutrons at a rate of up to 10^8 neutrons per second in 10µs duration pulses. The neutron generator is installed in the back wall of the chamber. The tritium target of the neutron generator is surrounded by a lead annulus in order to amplify the neutron flux through (n,2n) reactions. The emergent spectrum is also softened.

Two un-moderated He-3 detectors are installed inside the chamber to monitor the neutron flux from the neutron generator during the active measurement – one of these is mounted horizontally near the neutron generator (to act as a cavity flux monitor) and the other (the barrel monitor) is fixed vertically on the chamber door on the opposite side of the waste drum from the neutron generator (to determine the effect of the waste matrix on neutrons from the neutron generator).

The output signals from the main He-3 detectors are amplified, with a group of three detector outputs feeding a single amplifier/discriminator. The detectors are fitted horizontally and arranged into nine counting banks, three per side (top, middle and bottom). The summed digital pulse train combining the outputs of all of the amplifier/discriminators for the detectors in each of the nine banks is collected separately whether the PANWAS is operated in passive or active mode. The positional information is used to facilitate the identification of waste that contains fissile material present as a single point source. This Neutron Imaging Technique (NIT) allows a geometrical correction to be applied to the measurement data and therefore improves the measurement accuracy in such cases where the system response would otherwise differ significantly from the default volume weighted average (or uniform distribution) calibration.

The individual detector bank pulse trains are counted separately in nine CANBERRA Multiport-II multichannel scaler (MCS) units. Three further Multiport-II MCS units are used to count the summed pulse train from all nine banks as well as the counts from the cavity flux and barrel flux monitors. A derandomized summed pulse train from all nine banks is also fed to a CANBERRA JSR-14 multiplicity coincidence analyzer that is used to collect the combined correlated passive neutron information.

The nucleonics is located in an electronics cubicle in a separate control room next to the room housing the assay chamber. The nucleonics cubicle also houses the neutron generator drive chassis (ATC type N-250).

The entire measurement process is fully automated by means of a GE Fanuc type 90-30 Programmable Logic Controller (PLC) which is in direct control of the various mechanisms for drum handling as well as the AAS drive mechanism and the neutron generator. The PLC acts under the direction of the application software running on a PC in the control room, which forms the main user interface. Following the loading of a waste drum onto the trolley of the drum transfer system (using a fork lift truck and a dedicated crane), the assay measurement process is initiated following vacation of the measurement room by the operator personnel. The chamber door opens and the drum is driven inside and deposited onto the rotator assembly. The measurement begins after the chamber door is closed with the passive measurement, followed by the active measurement. At the end of the measurement, the drum is unloaded from the chamber and returned to the drum transfer trolley for collection by the operator.

A safety interlock system, which incorporates sensors for the detection of the presence of personnel and the state of the doors to the measurement room, prevents operation of the drum transfer system and the neutron generator when personnel are present in the room.

The PANWAS application software is the CANBERRA NDA2000 waste assay software. This is a fully integrated software package providing overall control of the entire PANWAS assay measurement process; it provides the main user interface, directs the assay measurement sequence via the PLC, acquires data from the nucleonics, analyses the results and produces the assay report. The NDA2000 software also provides maintenance functions for system calibration, parameter editing, diagnostics as well as measurement control (through regular quality check measurements making use of the AAS Cf-252 source to verify correct operation of the nucleonics) and a comprehensive data review and reanalysis function.

Because NDA2000 is a standard software package that is also in use on the other two pre-existing (SGS and ISOCS) gamma spectrometry based assay systems, it has been possible to integrate the three systems so that they share (via a local area network) a common setup and assay database. In this way, assay data for a given waste drum measured on the PANWAS is combined with the corresponding measurement data from one or both of the gamma systems into a single multi-modal assay result, independently of the order in which the individual measurements were carried out.

In addition to the NDA2000 software, CANBERRA supplied a separate *Interpreter* data analysis software package in order to provide Nucleco with a means of facilitating the necessary expert data review of the waste assay results. The Interpreter software is integrated with the NDA2000 results database and automatically carries out a comparison between the data supplied by the original waste consignor and the measured results from the PANWAS, SGS, and ISOCS systems on a drum-by-drum basis. Using a set of pre-defined rules, any significant anomalies are 'flagged' so that the drum can be subject to further more detailed expert review.

DATA ANALYSIS

The PANWAS data analysis software takes the passive mode count data from the multiplicity shift register and calculates the Pu-240 effective mass from the background and dead time corrected Reals (Doubles) count rate. Corrections are also applied for the effect of the waste matrix (using the AAS perturbation data) and (through the NIT analysis) for any point source effects as well as corrections (using the neutron multiplicity data) for the effects of cosmic rays on the background neutron count rate.

In active mode, the background corrected count rate in the *early gate* of the MCS data (normalized to the neutron generator output) is used to calculate the fissile content of the waste (expressed as a Pu-239 equivalent mass). As with the passive assay, corrections are applied for the effects of the waste matrix (using the barrel monitor counts and the passive mode AAS results) and for point sources using the NIT results. Corrections are also applied for the dependence of the active background count rate on the waste matrix type and for the effects of self-shielding [6] in any lumps of fissile material present assuming a realistic maximum lump size.

CALIBRATION AND CHARACTERIZATION

Calibration and characterization of the PANWAS was required in order to define the fundamental relationships between the quantities measured by the equipment (neutron count rates) and the amount of nuclear material present in the monitor. The calibration and characterization results are embodied in a set of parameters together with the algorithms that use them.

For this purpose, a set of standard drums of each size (200 liter and 400 liter) and inactive simulated waste matrix materials (including simulated supercompacted waste pucks for the 400 liter drums) representative of the actual waste form were used. Details of the drums and contents are as follows:

- 8 different 200-liter drums containing the following matrices: air, polyurethane, DAW (diverse active waste composed of low density material), neoprene, low and medium density wood (wood chip, glue and melinex), metallic and cemented.
- 2 different 400 liter drums, one of them empty and the other one with a cemented grout liner, to accept the various possible combinations of simulated puck simulants made up of material ranging from low density (neoprene), medium density fibreboard (MDF) to high density (metallic).

Table I shows the main characteristics of the different calibration drums and the matrix materials used.

Table I. Details of the Drums and Simulated Matrices used for the PANWAS Calibration/Characterization

Drum Size	Matrix type	Gross Weight [kg]	Net Weight [kg]	Container Volume [l]	Matrix Density [kg.l^{-1}]
200 liter	Empty	17	0	217.5	0.00129
200 liter	Polyurethane	21	4	217.5	0.019
200 liter	Diverse active waste (DAW) - Mixed waste containing mainly paper and some plastic.	46.2	29.2	217.5	0.150
200 liter	Neoprene	113.8	96.8	217.5	0.454
200 liter	Wood (low density)	100.5	83.5	217.5	0.387
200 liter	Wood (high density)	170.5	153.5	217.5	0.714
200 liter	Metallic	296.5	279.5	217.5	1.561
200 liter	Cemented	453.5	436.5	217.5	2.109
400 liter	Empty drum	58		377	0.00129
400 liter	Cemented	800	742	377	2.044
400 liter*	Ungrouted 400 liter drum either empty or with various matrices made up of pucks of metal, high density wood (MDF) and neoprene	160 - 610	100-550	377	0.5 - 2.0
400 liter*	Grout-lined 400 liter drum either empty or with various matrices made up of pucks of metal, high density wood (MDF) and neoprene	355 - 810	100-550	377	0.5 - 2.0

(*Package density and weight depend on combination of pucks used)

A set of 10 sealed plutonium oxide (PuO_2) calibration reference standards with masses in the range 0.1g to 25g was used for the calibration measurements. All of the standards are in doubly encapsulated stainless steel capsules. Each capsule has an outer diameter of 30mm and height of 50mm, with a total wall thickness on the base of 2.4mm. The plutonium oxide is in the form of a powder and is retained within the inner cell using a plunger that is held in place using a spring. The plutonium standards were supplied with a certificate detailing certified values for the amount and isotopic composition of the plutonium within the standards, together with information on the impurity contents determined during the analysis, meaning that their total neutron output is accurately known (to ±1.8% relative standard deviation, limited by basic nuclear data and impurity uncertainties). A range of analytical techniques was used to determine the actinide and impurity contents of the materials used in the preparation of the standards. Full details of preparation and characterization of the PuO_2 sources are given elsewhere [7]. The Pu-240 effective masses [8] of the items are consequently well known and self-multiplication effects have been studied in detail using Monte Carlo methods. Similarly, the Pu-239 equivalent mass [8] of the items is well known and the self-shielding factors for each have been calculated.

The following measurements were carried out to characterize the ^{3}He detectors and assay chamber:
- High voltage plateau measurements,
- Fission neutron die-away time measurement in passive mode.

The following sets of measurements were carried out to characterize and calibrate the PNCC sub-system:
- Dead-time measurement using different combinations of different Cf-252 sources,
- Measurement of PNCC chamber efficiency using both plutonium and Cf-252 sources,
- Passive neutron background count rates,
- Passive neutron background reduction calibration, to take into account the effect of the matrix on the passive background,
- Passive neutron mass calibration using plutonium standards,

- Passive neutron Add-a Source calibration,
- Passive neutron matrix correction calibration,
- Passive neutron imaging technique (NIT) calibration – used to identify and locate the position of a point source in the waste to improve the accuracy of the matrix correction,
- Passive neutron ‘high Z’ background correction calibration to take into account the increase in background neutrons caused by spallation of cosmic rays in certain matrix materials.

The following sets of measurements were carried out to characterize and calibrate the Differential Die-away (DDA) sub-system:

- Active neutron interrogation background calibration,
- Active neutron mass calibration / Differential Die-away calibration,
- Active neutron matrix correction calibration,
- Active NIT calibration.

Space prevents us from describing the experimental calibration work in depth here but in future published reports we hope to expand on this important aspect. It should be note that supporting simulation work was also performed.

SYSTEM PERFORMANCE

A number of key performance parameters for the PANWAS in passive mode are summarized in Table II.

Table II. PANWAS Passive Mode Characteristics

No. ^{3}He Detectors	117
^{3}He partial pressure	4 atmospheres
Efficiency (^{240}Pu Spont. Fission Neutrons)	26.4%
Die-away Time	41.5 µs
Totals Dead Time	0.581 µs
Reals Dead Time	2.40 µs
Characteristic Multiplicity Dead Time	55 ns
Doubles Gate Utilization Fraction	0.65
Pre-delay	5.0 µs
Gate Width	52 µs
Sensitivity (Reals Rate)	36.1 sec^{-1}/g $^{240}Pu_{eff}$
Sensitivity (Ones Rate)	29.1 sec^{-1}/g $^{240}Pu_{eff}$
Typical background count rate (Totals)	15-17 sec^{-1}
Typical background count rate (Reals)	2.7-3.1 sec^{-1}
Passive assay count time	1440 sec

From the results of measurements carried out during the initial on-site commissioning of the PANWAS on the various test drums, the passive detection limits for the system were determined, in the form of the Minimum Detectable Activity (MDA) for Pu-240 effective, for a range of different waste matrices inside the different drum types. These are given in Table III assuming that the activity is uniformly distributed within the waste and that the background rates and the count time used are as given in Table II.

Table III. PANWAS Passive Mode Ddetection Limits

Drum Type	**Matrix Type**	**Passive MDA (mg Pu-240 effective)**
200 l	Empty	4.5
200 l	Heterogeneous	5
200 l	Cement	25
200 l	Neoprene	20
400 l ungrouted	Empty	5
400 l ungrouted	High Density Wood	30
400 l ungrouted	Neoprene/HD wood	25
400 l grout liner	Cement	65
400 l grout liner	Empty	10
400 l grout liner	High Density Wood	70
400 l grout liner	Neoprene/HD wood	45

Table IV gives the corresponding characteristics of the PANWAS in active mode and Table V gives the detection limits in the form of the MDA for dilute Pu-239 equivalent, based on measurements carried out during the initial on-site commissioning of the system.

Table IV. PANWAS Active Mode Characteristics

Early Gate Start	1,200 μs
Early Gate Width	5,800 μs
Late Gate Start	7,002 μs
Late Gate Width	2,498 μs
Sensitivity (200 l drums)	1.33 sec^{-1}/g Pu-239 equivalent per flux monitor rate
Sensitivity (400 l drums)	1.22 sec^{-1}/g $^{239}Pu_{eq}$ per flux monitor rate
Interrogation Background (empty 200 l)	0.048 sec^{-1} per flux monitor rate
Interrogation Background (empty 400 l)	0.043 sec^{-1} per flux monitor rate
Typical Zetratron neutrons per pulse	10^6
Zetratron Repetition Rate	100 Hz
Active assay count time	300 seconds
Typical Zetatron neutrons per second	10^8
Zetatron pulses per Assay	30,000

Table V. PANWAS Active Mode Detection Limits

Drum Type	**Matrix Type**	**Active MDA (mg Pu-239 equiv.)**
200 l	Empty	3.5
200 l	Heterogeneous	3.6
200 l	Cement	25
200 l	Neoprene	40
400 l ungrouted	Empty	4
400 l ungrouted	High Density Wood	10
400 l ungrouted	Neoprene/HD wood	12
400 l grout liner	Cement	25
400 l grout liner	Empty	4
400 l grout liner	High Density Wood	25
400 l grout liner	Neoprene/HD wood	30

CURRENT STATUS AND FUTURE WORK

The PANWAS system has been in full operation for a year at Nucleco’s site during which it has completed its first major measurement campaign for the purposes of nuclear material physical inventory taking.

As a result of the continuing review of the system performance, a number of possible improvements to the system design were identified that would result in a reduction in the detection limits for the PANWAS operating in both passive and active mode. Many of these design modifications have now been implemented and the system calibration has been updated. A new performance review will be undertaken before the PANWAS is put back into service for its next measurement campaign later in 2006.

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