A combined calorimetry, neutron coincidence counting and gamma spectrometry system (CANEGA) for enhanced plutonium mass and isotopic assay

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Abstract. A measurement approach for enhanced plutonium assay combining three passive non-destructive measurement techniques – calorimetry, neutron coincidence counting and gamma spectrometry (CANEGA) – has been designed and evaluated. The combination of the measured quantities obtained from the three techniques not only leads to an improvement of the plutonium assay through redundant and complementary measurement information, but also provides a more complete fingerprint for any plutonium-bearing sample under assay. Further, the combined measurement information allows one to derive a more reliable estimate for the ^{242}Pu isotope abundance not directly measurable by gamma spectrometry. A conceptual feasibility and design study for a transportable CANEGA prototype instrument has been carried out with the aim of defining the most promising and advantageous instrument configuration.

1. Introduction

The adopted primary non-destructive assay (NDA) approach in Safeguards for Pu mass measurements is to combine passive neutron coincidence counting (PNCC) with high-resolution gamma spectroscopy (HRGS). The Pu isotopic abundances are determined from gamma spectra taken by HRGS and analysed by codes like MGA and FRAM. However, there are some inherent limitations and drawbacks of this NDA approach for the Pu mass determination. The main limitation accrues from the missing information on the abundance of the isotope ^{242}Pu, which significantly contributes to the measured neutron coincidence rate in PNCC, but cannot be determined directly by HRGS. This problem is only partially solved through the application of isotope correlations estimating the relative abundance of ^{242}Pu from relations to ratios of other plutonium isotopes measurable by HRGS. Any error on the ^{242}Pu determination affects the performance both of the plutonium isotope abundance measurements and of the quantitative determination of the amount of plutonium. Another drawback of the combined PNCC+HRGS approach is related to the PNCC measurement itself, which is not really tamperproof (traces of ^{244}Cm, for example, can quickly invalidate the PNCC measurement), and which even for small samples requires careful neutron multiplication corrections in order to arrive at unbiased measurement results.

At the Institute for Transuranium Elements (ITU), Karlsruhe, we are also evaluating the alternative NDA approach for Pu mass determination, namely the combination of calorimetry and HRGS. Although calorimetry is so far not represented among the NDA instrumentation of the Safeguards authorities, it yet provides some attractive features compared to PNCC. Its main asset is certainly the ease of measurement interpretation through the insensitivity to all kinds of sample properties. From our recent investigations and application of calorimetry to small sample measurements of gram amounts of representative reactor-grade plutonium materials we have gained pertinent experiences with this technique with typical samples encountered in Safeguards verification measurements [1].

The experiences gained from the parallel use of calorimetry and PNCC in conjunction with HRGS for the non-destructive plutonium assay have led to the proposal of a combined calorimetry, neutron

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coincidence counting and gamma spectrometry (CANEGA) measurement approach for the non-destructive plutonium assay [2, 3]. The combination of the directly measured quantities from the three techniques not only allows a fairly good determination of the ^{242}Pu isotope abundance, but also contributes through the use of redundant and complementary information to an improved plutonium assay. The combined NDA approach also proved to be of value for the assay of special types of nuclear materials, for which a single NDA technique normally is not able to give an adequate measurement answer. Such kind of special nuclear materials arise, for example, from R&D work at ITU on future fuel cycles including minor actinide transmutation.

While in a Safeguards analytical laboratory like ITU the three NDA measurements can be performed, in principle, with resident separate instruments, it would be yet of practical advantage for a potential on-site application of CANEGA to integrate the three NDA techniques into one single, transportable instrument. A corresponding feasibility and design study has been carried out and will be discussed in this paper.

2. Concept and performance of CANEGA

The principle of the CANEGA assay approach, its performance as well as the improvements to be expected from the combined NDA measurements have been previously described [2, 3]. Only a brief summary with some additional performance data is therefore presented below.

2.1. Concept for the ^{242}Pu determination

The combined calorimetry, neutron coincidence counting and high-resolution gamma spectrometry measurements can directly determine a total of six quantities from a plutonium sample:

the thermal power P, the amount of m_{240}-*effective* and the plutonium isotope weight ratios m_{238}/m_{239}, m_{240}/m_{239}, m_{241}/m_{239} and the ratio m_{Am}/m_{239} from gamma spectrometry. In the following we denote the isotope weight ratios relative to ^{239}Pu as R_{238}, R_{240}, R_{241} and R_{Am}.

Taking the ratio of the thermal power P over m_{239} yields:

$$P/m_{239} = P_{238} \cdot R_{238} + P_{239} + P_{240} \cdot R_{240} + P_{241} \cdot R_{241} + P_{242} \cdot R_{242} + P_{Am} \cdot R_{Am} , \qquad (1)$$

where the quantities P_i denote the specific thermal power of the respective isotope. In Eq. 1 the term $P_{242} \cdot R_{242}$ can be reasonably neglected without introducing a significant error (typicall less than 0.2%) because of the very low specific thermal power P_{242} of ^{242}Pu. Eq. 1 then allows to calculate the quantity m_{239} from the measurement observables P, R_{238}, R_{240}, R_{241}, R_{Am} and the known specific heat values P_i. With the knowledge of m_{239} the isotope ratio $R_{242} = {}^{242}$Pu/^{239}Pu is easily calculated from the following equation containing only known and/or measured quantities:

$$m_{240eff} /m_{239} = \gamma_{238} \cdot R_{238} + R_{240} + \gamma_{242} \cdot R_{242} \qquad (2)$$

In this equation we are using for the coefficients γ_i , which proportion the measured Reals rates from ^{238}Pu and ^{242}Pu relative to ^{240}Pu, our experimentally determined values reported in Ref. 4. With the value for R_{242} thus obtained, and with the measured gamma-spectrometric values for R_{238}, R_{240} and R_{241} the new isotopic composition is then calculated to yield improved values for P-eff and ^{240}Pu-eff as input data for the Pu mass determination from calorimetry and PNCC.

2.2. Performance of CANEGA

In addition to results previously reported we have applied the CANEGA concept to another set of comparative NDA measurement data recently produced at ITU. Sample data for the set of 19 PuO$_2$ powder, MOX powder and MOX pellet samples used for the test are listed in Table 1. In terms of

sample amount and isotopic composition the samples are representative for the type of verification samples encountered in the routine Safeguards analyses at ITU.

The non-destructive calorimetry, PNCC and HRGS measurements were performed before sample dissolution for the classical destructive analyses (IDMS and/or titrimetry for the U and Pu concentration, and TIMS for isotopic composition). The calorimetry measurements were carried out with a high-sensitivity small sample calorimeter (Model TAM III from Thermometric AB), while for the combined PNCC and HRGS measurements the samples were counted in the so-called OSL neutron/gamma counter [5]. Both the calorimeter and the neutron counter were calibrated with the same reference material (a sealed sample with 500 mg of isotopically pure $^{240}PuO_2$). The thermal power of the samples (in mW) as measured in the calorimeter, and the neutron-coincidence ('Reals') rates obtained from the samples with OSL counter (detection efficiency = 40%) are also listed in Table 1 for information. The right-hand column in the Table quotes for the individual samples the approximate counting times required to reach a 0.2% counting precision for the Reals rate.

Table 1. Sample and measurement data for the analysed samples.

Type of sample	Sample mass (g)	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	Am-241	P (mW)	Reals (cps)	Time for 0.2% NCC precision (h)
				wt. %						
PuO2	1.03	2.76	51.48	28.45	9.05	8.26	1.98	18.56	51.12	1.4
PuO2	1.24	2.84	51.58	26.97	10.25	8.37	0.74	21.93	61.47	1.1
PuO2	1.02	2.65	53.65	27.50	7.91	8.29	3.28	18.61	48.67	1.4
PuO2	1.03	2.35	55.26	27.05	7.66	7.68	1.91	16.27	47.32	1.5
PuO2	1.04	2.16	56.72	26.46	7.87	6.78	2.69	16.12	45.10	1.5
PuO2	1.06	1.68	59.00	25.73	7.50	6.09	2.39	13.82	42.35	1.6
MOX pellet	6.08	3.11	51.69	27.29	9.41	8.50	0.79	6.30	16.96	4.1
MOX pellet	7.91	2.93	52.27	27.00	9.51	8.30	0.80	9.06	25.13	2.8
MOX pellet	7.56	2.31	54.20	26.47	9.54	7.48	0.95	10.08	31.04	2.2
MOX pellet	7.25	2.43	54.79	26.06	9.23	7.48	0.89	8.54	25.25	2.7
MOX pellet	5.68	2.04	55.92	25.76	9.44	6.85	0.88	6.03	19.13	3.6
MOX pellet	6.62	2.11	57.21	26.40	7.39	6.89	2.40	5.39	15.52	4.5
MOX pellet	7.80	1.95	57.27	26.37	7.60	6.80	2.28	6.09	18.31	3.8
MOX pellet	7.23	2.04	57.30	26.14	7.78	6.74	2.42	9.65	27.88	2.5
MOX pellet	6.82	1.95	57.50	26.19	7.69	6.67	2.24	8.72	25.98	2.7
MOX powder	1.54	2.95	52.06	27.09	9.56	8.33	0.76	7.02	18.86	3.7
MOX powder	5.08	2.15	55.79	26.14	8.88	7.05	0.96	4.18	12.79	5.4
MOX powder	4.92	2.04	55.91	25.76	9.44	6.85	0.88	5.11	15.71	4.4
MOX powder	4.94	2.00	57.51	25.42	8.49	6.58	1.26	4.79	14.29	4.9

Three different sets of data for the isotope ratio R_{242} have been evaluated for comparison: one set derived from the CANEGA approach (applying Eqs 1 and 2), and two further sets derived from isotope correlations of the type $R_{242} = a \cdot (R_{238})^b \cdot (R_{240})^c$. In each case only gamma-spectrometric isotope ratios evaluated with the analysis code MGA (version 9.5) were used as further input data.

Appropriate coefficients a, b, and c in the above isotope correlation depend on the type of plutonium. According to our established criteria for the categorisation of the material type [7], the plutonium in all of the samples has been identified as PWR plutonium. With this classification we have applied two different sets of coefficients:

a = 1.313, b = 0.33, c = 1.7   as previously recommended [6], and
a = 1.441, b = 0.484, c = 1.149   as recently evaluated at ITU [7].

The R_{242} values obtained from the above correlations and from the combined calorimetry, PNCC and HRGS measurements (CANEGA) were then compared with the "true values" from mass spectrometry (TIMS). The average percentage differences and their standard deviations for the given set of 19 measurement samples are given in the 2nd column of Table 2.

Table 2. Average percentage difference and standard deviation between R_{242}, P-eff and ^{240}Pu-eff values calculated with HRGS /MGA and TIMS isotopic data.

Source of R_{242}	R_{242}	P-eff	^{240}Pu-eff
Isotope correlation (previous coefficients)	-1.23 ± 4.23	-0.08 ± 0.64	-0.64 ± 1.04
Isotope correlation (ITU coefficients)	-0.17 ± 3.54	-0.15 ± 0.61	-0.40 ± 0.85
CANEGA	1.11 ± 2.02	-0.14 ± 0.51	0.31 ± 0.64

We note that the R_{242} values obtained from the two applied correlations and from the CANEGA approach show comparable average differences to the TIMS reference values. However, the scatter of the CANEGA results relative to the TIMS values is reduced by about a factor of 2 compared to the correlation data.

One remark concerning the absolute accuracy of the R_{242} values derived from CANEGA may be appropriate. The R_{242} values calculated from Eqs. 1 and 2 critically depend on the accuracy of the measured thermal power P from calorimetry, and on the measured mass of m240-effective from PNCC, with the latter being the most critical quantity. Our previous sensitivity studies [2] have shown that a possible bias in the measured value of P would bias the derived R_{242} value about twice as large, and a given measurement bias in m240-eff would even magnify the resulting bias in R_{242} by a factor of 3-4. While the thermal power P can be normally measured with high accuracy, it is by no means a matter of course to achieve in real measurements a bias-free determination of m240-eff from PNCC, because even for small samples the measured Reals rates require corrections for neutron multiplication effects of up to 4%. This is illustrated in Fig. 1, which displays for the given set of samples the corresponding correction factors determined from MCNP calculations. A systematic bias of slightly more than a quarter of a percent in the PNCC measurement of the effective ^{240}Pu mass would therefore automatically lead to a bias of about 1% in the derived R_{242} values.

Fig. 1. Correction factors for neutron multiplication obtained from MCNP calculations for the set of samples listed in Table 1.

The R_{242} values from the isotope correlations and from CANEGA were combined with the gamma-spectrometric ratios for R_{238}, R_{240} and R_{241} to obtain the complete isotopic composition from gamma spectrometry. With the respective isotopic composition the isotope-specific help quantities P-eff and ^{240}Pu-eff required for the Pu mass evaluation from calorimetry and PNCC were finally calculated. The average percentage differences relative to the corresponding values obtained with TIMS isotopic data are listed in Table 2. It is obvious that the P-eff values are not as sensitive to R_{242} as the ^{240}Pu-eff values, which show some improvement with the improved R_{242} values from CANEGA.

3. Design study for a CANEGA instrument

Having demonstrated the principle benefits of the CANEGA approach for an improved plutonium assay we decided to take a further step with a feasibility study for the design of a CANEGA instrument. For this purpose ITU has commissioned A.N. Technology, Wallingford, UK, which has experience with all three NDA techniques involved, to carry out a corresponding feasibility study into the design of a transportable combined calorimeter, neutron and gamma measuring device for on-site use in nuclear facilities. The study has been conducted in two phases, with phase 1 as a conceptual design phase reviewing design options, and phase 2 performing some detailed modelling for the finally selected configuration.

3.1. Design considerations

The design considerations were addressing all aspects of the system. This includes the overall performance, hardware and software requirements for the system as well as the specific requirements for each measurement type, i.e. calorimetry-, neutron coincidence- and gamma measurements. The basic specifications were:

- A CANEGA system for small sample measurements (gram-size PuO_2 powder or MOX powder and pellet samples);
- Measurement cavity with dimensions of 40 mm dia x 80 mm heigh;
- Neutron detection efficiency as high as possible, ideally close to 40% as obtained with the existing OSL neutron/gamma counter installed in the Euratom on-site laboratories;
- Calorimeter sensitivity as large as possible (larger than100 mV/mW) assuring a measurement repeatability of 0.1% at a thermal sample power of 10 mW;
- High-resolution HPGe detector for the low-to-medium energy range (up to 400 keV) subtending a solid angle relative to the sample of not significantly smaller than 10^{-2} sr.

It has been realized that the specifications were ambitious, and that at the end probably some compromises in terms of performance would have to be made in view of the sometimes conflicting requirements, especially for the calorimeter and neutron measurements.

3.2. Design options

Several designs have been considered to arrive at an optimum design for a combined measurement system. Advantages and disadvantages of each design have been assessed with respect to thermal block/moderator features, practicality of construction and ease of operation and maintainability.

After initial review of the design it was realised that a 40mm diameter by 80mm high sample chamber had implications for the performance and size of the system. This requirement was reduced to a 30mm diameter by 80mm high chamber.

For optimum performance the calorimeter should be preferably of the twin-cell design, with identical measurement and reference chambers. This leaves options for two fundamental configurations:

a) A side-by-side design, more closely resembling the classic design of ANTECH's small sample calorimeters, whereby both cups are mounted eccentrically in the thermal block as shown in the example given in Fig. 2.
b) An over/under design, in which the sample cup is placed axially directly above the reference cup and both are thermally linked. An example of this type of configuration is shown in Fig. 3.

The investigated options are essentially variations upon these two configurations, differing in the layout of the moderator/thermal block and in the way the gamma detector is incorporated into the system. There were in total eight different options that have been selected and reviewed.
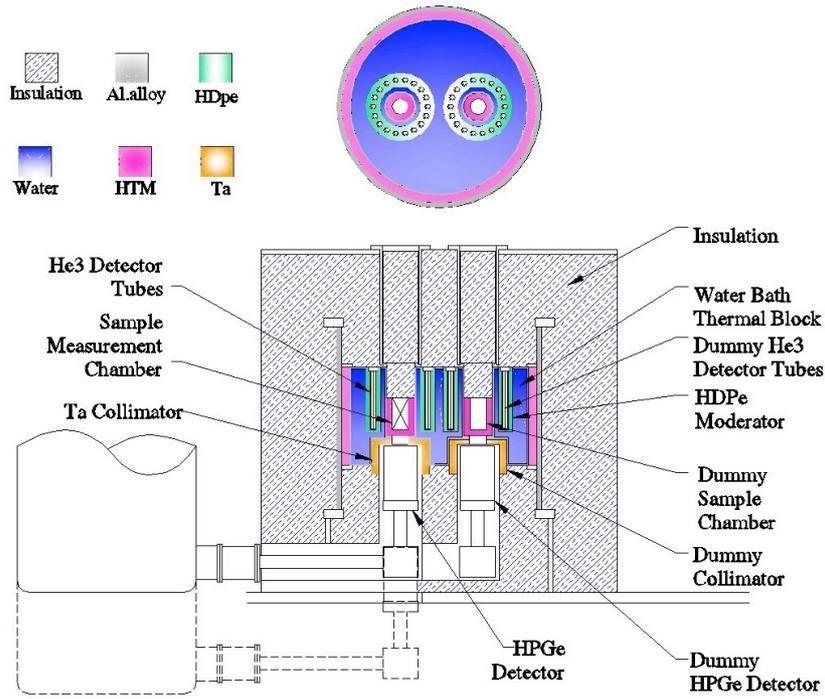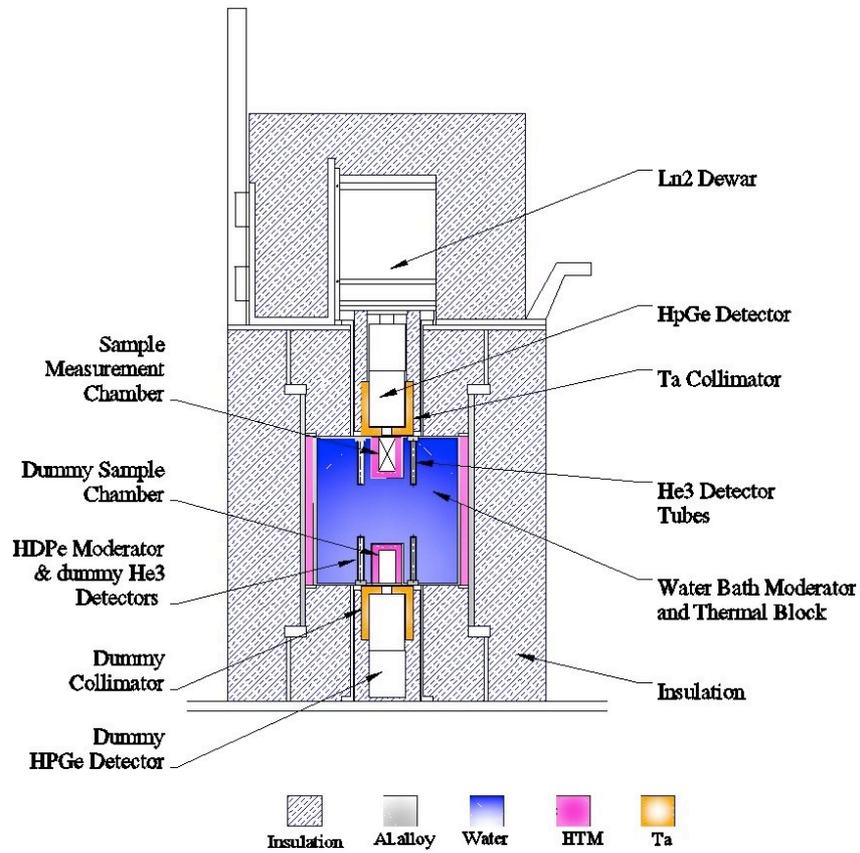
Fig. 2. Example for a side-by-side design.

Fig. 3. Example for an under/over design.

3.3 Proposed configuration

The option finally selected for further optimisation through modelling is a side-by-side configuration similar to that shown in Fig. 2, with ^{3}He tubes in close proximity to the sample chamber and a side-mounted gamma detector. The moderator/thermal block assembly consists of a mixed polyethylene/water configuration as shown in Fig. 4 in a simplified vertical section of the detector.
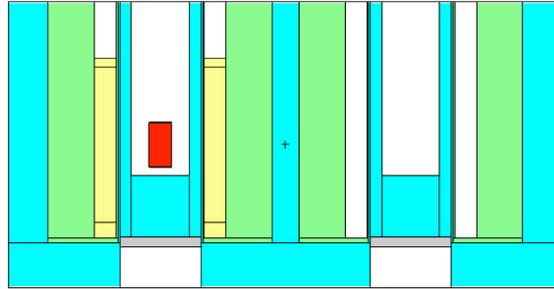

Fig. 4. Simplified cross-section of side-by-side model.

3.4 Monte Carlo modelling (neutron measurement)

The initial model consisted of a single row of 12 x 4atm ^{3}He tubes with an active length comparable to the height of the sample (80mm) mounted in a polyethylene moderator positioned in close proximity to the sample as per the sketch shown in Fig 4. The rationale behind this design was to look at the efficiency of a compact (low height) thermal element. This design yielded an efficiency of approximately 12%, far below that required.

The reasons for the low efficiency were considered to be due to a combination of factors such as tubes having a short active length, low ^{3}He gas fill pressure, not enough detectors and an insufficient volume of moderator.

Following this the design was refined to include a double row of He^3 tubes with a much longer active length above and below the chamber. The thickness of polyethylene moderator was increased, the tube fill gas pressure varied and the respective radial distance of each row of tubes from the centre of the sample axis also varied.

The results of the modelling conclude that 40 % efficiency can be met by using a double ring of ^{3}He tubes, one ring positioned at a distance of 6.65cm and the other 11.15 cm from the vertical axis of the sample. It can be achieved with a fill gas pressure of 6 atm and active length of 24 cm or a fill gas pressure of 8 atm and active length of 22 cm.
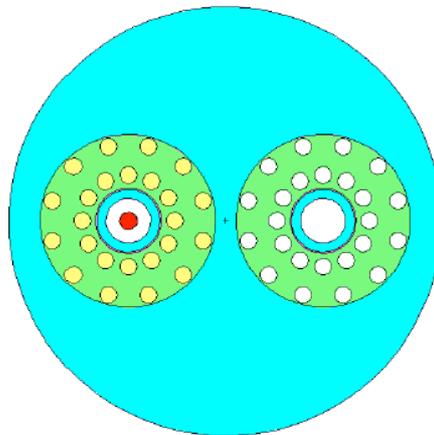

Fig 4. Optimised neutron detector array

Achieving maximum efficiency will need to be balanced with other aspects of the design relating the gamma and calorimeter measurements. The polyethylene moderator is in close proximity to the outer wall of the sample chamber. This directly influences how closely the gamma detector can be coupled to the bottom of the sample and the amount of water that can be circulated between the moderator and sample chamber wall - hence the cooling efficiency. These are design issues that will need to be taken into account before any such system is manufactured.

3.5 Modelling gamma measurement

The gamma detector is mounted under the sample, looking up through the base of the sample chamber. Surrounding the detector head is a tantalum shield with an aperture through which the sample is viewed. The performance of the gamma detector has been modelled for a number of cases of varying detector to sample distances and absorbing media.

For the initial model PNNL (Pacific North-west National Laboratories) SYNTH was used to generate synthetic gamma ray spectra after defining various parameters for the sample, source, absorbers, detector and electronics. The basic arrangement proposed for the gamma component of the CANEGA system was configured and parameters varied to produce a range of spectra.

A coaxial Ge detector was selected as it was considered the most appropriate to enable the correct parameters for the detector to be modelled. For this option the efficiency, diameter and length can be changed to match most shapes of detector. The resolution can be varied but SYNTH doesn't enable a resolution of 0.6keV to be achieved in the 100keV region of the spectrum. This, therefore, means that the SYNTH spectra cannot be used directly with MGA to give a realistic value for the accuracy of the MGA result for real spectra.

There are a number of peaks that are isolated and not part of a multiplet, ^{239}Pu at 129 keV, ^{241}Pu at 148 keV, ^{238}Pu at 152 keV and ^{240}Pu at 160 keV. These peaks were used for a cross comparison with real spectra. The real spectra were analysed with MGA and spectra with an error on the specific power value of less than 1% were chosen. The gross and net counts in the above peaks were then counted using a set of ROI's. These ROI's were then used to calculate the gross and net counts in the peak for the SYNTH spectra. Since these peaks are isolated the counts in these peaks will be comparable between the real and SYNTH spectra. It is assumed that if the SYNTH spectra have a greater number of net counts in the peak, then that SYNTH model will, for a real measurement with a detector of resolution 0.6 keV, give an error on the specific power value of less than 1%. A summary of the cases modelled is shown below.

Table 2: Summary of results for a range of gamma modelling parameters.

Case [Note 1]	Pu Isotopic composition	Distance between source and detector (cm)	Detector cross sectional area (cm2)	Detector depth (cm)	Absorbing material (cm) [Note 2]	Count time (h)	Net counts OK [Note 3]
1	Recycled MOX	18	3.6	1.5	1 Al	1	All OK
2	Recycled MOX	18	1.6	1.5	1 Al	1	129keV Pu239 failed
3	AGR	18	1.6	1.5	1 Al	1	129keV Pu239 failed
4	AGR	18	3.6	1.5	1 Al	1	All OK
5	AGR	18	3.6	1.5	1.5 Al 5 H2O	1	All OK
6	AGR	25	1.6	1.5	1.5 Al 5 H2O	1	129keV Pu239 failed
7	AGR	25	3.6	1.5	1 Al	1	All OK
8	AGR	25	3.6	1.5	1.5 Al 5 H2O	1	All OK

Notes:
1. The basic arrangement was as follows:
   a. 1g sample of Pu metal, 0.1749 cm^2 cross sectional area, 0.3 cm thick.
   b. Coaxial Ge detector with dimensions varied as per columns 4 and 5
   c. Detector end cap thickness 1mm Aluminium
   d. 8192 channels defined with a gain of 0.075 keV/ch
   e. Count time of 3600 s

2. The remaining absorbing material that makes up the total distance between source and detector is air.
3. In the cases where the 129 keV peak failed the counts in the peak were between 4958 and 8060 compared to the real spectra values of between 9472 and 9864. This indicates that with double the count time, i.e. 2 hrs, the required number of net counts in the peak could be achieved.

Detailed gamma modelling suggests that if a 3.6 cm^2 cross sectional area by 1.5 cm deep coaxial detector is chosen it could be up to 25 cm away from the sample and view it through up to 5cm of water, 1.5 cm of aluminium and 18.5 cm of air. The detailed design may allow us to decrease this distance and the amount of absorber material but this shows that the minimum requirements of 1 % precision at 100 keV can be met with the chosen concept design.

3.6 Modelling calorimeter measurement

The same design used to model the neutron and gamma measurement criteria was used to model the calorimeter properties.

The heat output from a sample is measured with a series of thermopile junctions installed in an annular air gap between the inner cylinder forming the sample chamber and an outer cylinder forming a leak-tight barrier between the sample chamber and water bath. The thermopile array measures the heat flowing from the sample to the water bath and provides a voltage output proportional to the heat flow.

The sample measurement chamber construction has been modelled using two types of heat flow sensors:

a) An array of close coupled pad thermopiles mounted on flat surfaces machined on the outer surface of the inner cylinder,
b) A single continuous strip of thermopile junctions installed to run in vertical strips within the air gap between the inner and outer sample chamber cylinders.

Both types of sensors exceed the requirement for a measurement sensitivity greater than 100 ↘V per mW. The design utilising pads offers the best solution as it provides the greatest signal output per unit area and could potentially give up to 4 mV per mW. Vertical strips of thermopile junctions would be expected to give up between 0.1 to 0.5 mW per mW.

A measurement repeatability of 0.1% at 10 mW is routinely achieved with ANTECH's Model 601 Small Sample Calorimeter. This employs an aluminium thermal block to balance the need for fast measurements with thermal stability and repeatability. The proposed design for a CANEGA system will use a large volume water bath which has a higher thermal inertia, hence is more stable, in conjunction with more sensitive measurement and control electronics.

Further modelling demonstrated that to minimise the heat distribution error over the volume of the measurement cavity it will be essential to have an insulating layer of air between the bottom of the sample chamber and the outer chamber to minimise the heat flow through the bottom of the sample. In addition it will be important to have a highly insulating plug unit to prevent heat leaks from the top of the sample.

4. Conclusion

We consider the combined calorimetry, neutron and gamma measurements a viable approach for an improved non-destructive plutonium assay in smaller verification samples. By modelling the individual measurements in a combined instrument for a 30 mm diameter by 80 mm high sample chamber, and refining the model as we proceed, we have shown it is feasible to achieve a neutron measurement efficiency of 40 % and, although this constrains the gamma measurement by having to place the detector further away from the sample than desirable and view the base of the sample through a medium of water, air and aluminium it is possible to meet the gamma measurement requirements. Given the time it takes for the calorimeter and neutron measurement to complete,

conducting the gamma measurement for up to 2 hours, or more, in parallel with the calorimeter measurement would improve the measurement accuracy.

As anticipated, incorporating neutron and gamma measurement systems into the calorimeter thermal element affects the calorimeter measurement. By utilising a high density of thermopile junctions to measure the sample heat output the required sensitivity can be achieved. Furthermore, using a water bath construction in conjunction with more sensitive measurement and control electronics improves the stability, hence, repeatability of the system.

Minimising the heat distribution error across the sample presents challenges. It is likely that forced circulation of the water, in particular in the region between the sample chamber and the polyethylene moderator rings, will be required to obtain good heat transfer. Being able to incorporate a small volume of water, typically up to 5 cm depth, between the gamma detector window and the sample base is beneficial.

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