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A Combined Calorimetry, Neutron Coincidence Counting and Gamma Spectrometry System (CANEGA) for Enhanced Plutonium Mass and Isotopic Assay

S. Abousahl[a1] , H. Ottmar[a], P. van Belle[a], Th. van Vuure[a], K. Burke[b], J. Mason[b], A. Tolchard[b], A. Towner[b]

[a]European Commission, Joint Research Centre,
Institute for Transuranium Elements,
Karlsruhe, Germany

[b]ANTECH, A.N. Technology,
Wallingford OXON, OX10 9TA,
United Kingdom

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. ….

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}.

[1] Present address: European Commission, DG Joint Research Centre, Nuclear Activities Unit, SDME 10/60, B-1049 Brussels

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. ….

2.2. Performance of CANEGA

….. 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. ….

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. ….

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 than 100 µV/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. …

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. …

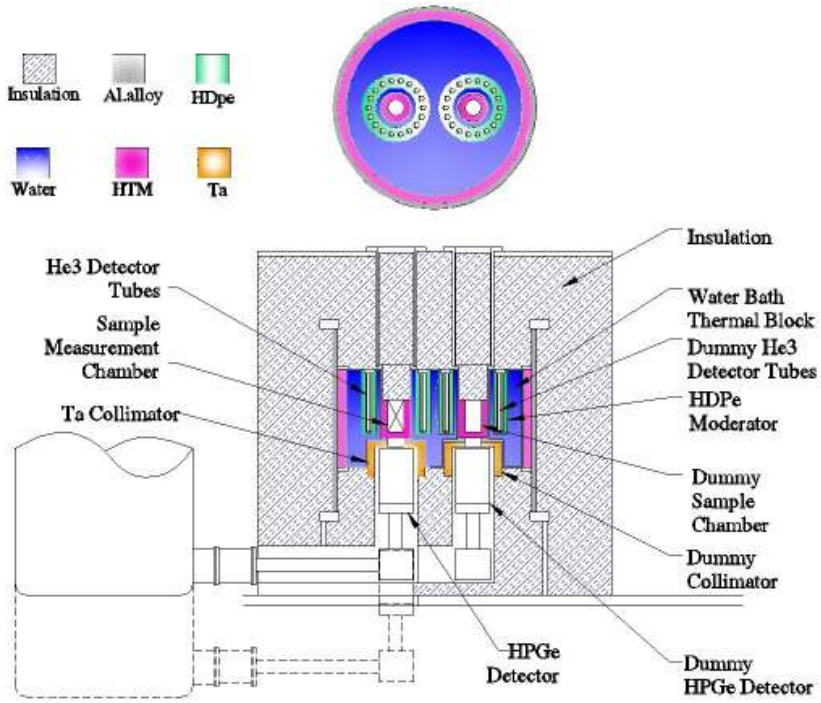

Fig. 2. Example for a side-by-side design.

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. ….

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