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Combined calorimetry/neutron coincidence counting/gamma spectrometry (CANEGA) measurements for plutonium mass and isotopic assay

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Abstract

A combined Calorimetry/Neutron Coincidence Counting/Gamma Spectrometry (CANEGA) assay approach offering the potential for improved nondestructive plutonium assay is described. The combination of the six directly measurable quantities as obtained from the three measurement techniques, namely the thermal power P from calorimetry, the effective mass $m_{240\text{eff}}$ of ^{240}Pu from neutron coincidence counting (NCC), and the plutonium isotope ratios 238/239, 240/239 and 241/239 and ^{241}Am/239 from high-resolution gamma spectrometry (HRGS) allow to determine the ^{242}Pu isotope abundance, which otherwise cannot be directly obtained from a HRGS measurement. In this manner the CANEGA measurement approach provides a more complete fingerprint for any plutonium-bearing sample under assay, which in turn helps to improve the overall accuracy of the plutonium mass determination. Pertinent features and performances of the individual and combined measurement techniques are presented and discussed.

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1. Introduction

The amount of plutonium in arbitrary plutonium-bearing nuclear materials can be determined nondestructively either by means of neutron coincidence counting (NCC) through the detection of time-correlated fission neutrons from the spontaneous fission of the plutonium isotopes ^{238}Pu, ^{240}Pu and ^{242}Pu, or by means of calorimetry (CAL) through a measurement of the thermal power generated from the radioactive decay of plutonium. For an accurate quantitative

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plutonium assay both techniques, NCC and CAL, require the knowledge of the isotopic composition of plutonium. In a completely nondestructive scheme of analysis the isotopic information is obtained, within certain limits, from a complementary high-resolution gamma spectrometry (HRGS) measurement.

In the international safeguarding of nuclear materials the prime nondestructive assay (NDA) technique currently employed for plutonium measurements is NCC, combined with gamma spectrometry. One limitation of this kind of plutonium assay arises from the fact that the abundance of the isotope ^{242}Pu cannot be determined by gamma spectrometry because of the absence of a detectable gamma-ray signature from this isotope. This deficiency starts to limit the accuracy of the plutonium assay particularly for higher burn-up materials, where ^{242}Pu contributes 25% or more to the measured neutron coincidence rate. In this situation recourse is therefore often made to isotope correlations in order to obtain a rough estimate for the ^{242}Pu isotope abundance. The recently proposed isotope correlations [1,2] work reasonably well only for defined categories of plutonium materials of known burnup history. However, this information is in practice not readily available for the materials to be assayed, leaving therefore often relatively large uncertainties for the estimated ^{242}Pu abundances. This uncertainty normally affects the performance both of the plutonium isotope abundance measurements and of the quantitative determination of the amount of plutonium.

The use of a combined Calorimetry/Neutron Coincidence Counting/Gamma Spectrometry (CANEGA) assay can improve this unsatisfactory situation, because the added information gained from the three separate NDA measurements allows to derive a fairly accurate value for the missing ^{242}Pu isotope abundance. Moreover, the availability of analysis results from two independent NDA techniques (CAL and NCC) for plutonium element assay, together with improved ^{242}Pu abundance values derived from the combined CAL/NCC/HRGS measurements, offers increased measurement performance and assurance for the plutonium mass determination as well.

In this paper we shall outline the principle of the CANEGA assay approach, with the emphasis put on the ^{242}Pu isotope abundance determination. Expected measurement performances as estimated from parameter studies are compared with results from actual measurements. In addition, measurement examples illustrating the increased confidence and measurement assurance gained for the Pu element assay from the combined nondestructive measurements are also presented and discussed.

2. Concept for the ^{242}Pu determination

The combined CAL, NCC and HRGS measurements can directly determine a total of six quantities from a plutonium sample:

- the thermal power $P = P_{238}m_{238} + P_{239}m_{239} + P_{240}m_{240} + P_{241}m_{241} + P_{242}m_{242} + P_{Am}m_{Am}$ from CAL, where P_{xxx} and m_{xxx} denote the specific thermal power and the amount of the respective isotope. The quantities P_{xxx} are well known physical constants (Table 1);
- the amount of m_{240}-effective $= \gamma_{238}m_{238} + m_{240} + \gamma_{242}m_{242}$ from NCC, where the coefficients γ_{238} and γ_{242} proportion per unit amount of isotope the contribution of ^{238}Pu and ^{242}Pu to the measured neutron coincidence rate relative to ^{240}Pu. Empirical γ-coefficients for the neutron counter used for the present studies have been recently determined to $\gamma_{238} = 2.714 \pm 0.011$ and $\gamma_{242} = 1.667 \pm 0.005$ [3];
- the plutonium isotope weight ratios m_{238}/m_{239}, m_{240}/m_{239}, m_{241}/m_{239} and the ratio $m(^{241}\text{Am})/$

Table 1
Specific thermal power values P [4]

Isotope	P (mW/g isotope)	Std. dev. (%)
^{238}Pu	567.57	0.05
^{239}Pu	1.9288	0.02
^{240}Pu	7.0824	0.03
^{241}Pu	3.412	0.06
^{242}Pu	0.1159	0.22
^{241}Am	114.2	0.37