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Rad Calc III: Radioanalysis Calculation Program for Plutonium and Americium Determination

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Introduction

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The radiochemistry team of the Analytical Chemistry Group has supported nuclear materials production and management programs at Los Alamos National Laboratory since the 1940s. Routinely, plutonium and americium contents in various matrices (such as metals, oxides, process solutions, and waste streams) are determined by direct alpha and gamma analyses¹. Over the years, analysts have written a number of computer programs^{2,3,4} to calculate analytical results. In 1999, the program was enhanced and upgraded to produce *Rad Calc III*⁵. The new program, written in Visual Basic 4.0, corrects limitations of previous versions, offers enhanced features, and incorporates user suggestions to customize the program and make it more user friendly.

Description

Rad Calc III calculates plutonium and americium contents in weapons-grade plutonium metals, oxides, and process solutions. The program also determines whether waste solutions meet the waste-discard limit for industrial, acidic, and caustic drain lines.

The user inputs the following information:

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- analysis type,
- customer and sample information,
- aliquot and dilution factors, and
- counting data.

When the user presses the "Calculate" button, results appear on the screen. The user may choose to print and/or save the file for later retrieval and data transfer. Once the results have been reviewed and approved, the operator can electronically report data on the Laboratory Information Management System (LIMS) and provide a hard copy of the results to the submitter of the sample.

Rad Calc III allows data collected from multiple detectors with different counting efficiencies to be entered into the calculation. The program also contains several features, including detector background correction, detector counting-efficiency correction, plutonium alpha-specific activity calculation based on isotopic information, and data outlier test (Q test⁶).

Detailed calculations and definitions for *Rad Calc III* are described in the user manual⁵. Salient features of the program, plutonium alpha specific-activity estimates and several correction factors that influence the final calculation of plutonium and americium concentrations, are presented here. These factors include americium and plutonium activities-overlap correction, alpha counter-coincidence correction, and salt-absorption correction.

Alpha-Specific Activity. Frequently, exact plutonium isotopic information is unknown, but an enrichment factor (wt %) for ^{240}Pu is usually associated with the samples to indicate different plutonium grades. To calculate plutonium and americium contents from gross alpha and gamma data, an estimate of plutonium alpha-specific activity is derived from more than 200 plutonium samples with known isotopic composition (Fig. 1).

Fig. 1

Alpha and Gamma Activities Overlap. Sodium iodide detectors are used to perform direct gamma measurements. However, this detector does not have sufficient resolution to discriminate between the gamma rays of plutonium and americium between

40 to 80 keV. Also, gross alpha counting does not differentiate plutonium and americium activities; therefore *Rad Calc III* uses simultaneous equations⁵ to determine final plutonium and americium concentrations.

Coincidence Correction for Direct Alpha Measurement. The alpha coincidence correction is based on the fact that when two alpha particles reach the gas proportional counter at the same or nearly the same time, the two events will be recorded as a single event.

Previous experimental data⁷ show that the following equation represents the determined coincidence-correction factor that can be applied to the obtained count rate.

$$CAVGA_{(cpm)} = AVGA_{(cpm)} + \left(\frac{AVGA_{(cpm)}}{100,000} \right)^2 \times 2000$$

where CAVGA is the coincidence corrected count rate and

AVGA is the average alpha count rate before coincidence correction.

Salt Absorption Correction. Direct alpha measurement by proportional counter involves counting alpha activity on a plate that contains a small deposited and dried aliquot of the sample solution. Experimental data showed the absorption of alpha particles caused by the salt residue left on the plate can be described by the following equation:

$$ABS = -0.0008 \times SALT^2 + 0.1774 \times SALT$$

where SALT is the amount of salt residue in μg and

ABS is the percent of alpha absorption.

The amount of salt residue on the sample plate is estimated by visual comparison to plates with known salt deposits.

Results

The *Rad Calc III* program provides a flexible, easy-to-use calculation tool for analysts to determine gross alpha activity in waste solutions and plutonium and americium contents in plutonium materials.

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