**USTUR 700: Measurement of Actinium in Tissue Samples -- Gamma Spectroscopy**

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| **Purpose** | Gamma spectrometry analysis of actinium in tissue | **Method Number** | USTUR 700 |
| **Original Date** | 10/10/95 | **Author** | LANL RT900 (11/92) |
| **Revision Number** | 0 | **Approved By** | Not approved for routine use |
| **Effective Date** | 10/10/95 | **Approval Date** | |

**NOTE: This method has been directly adopted from LANL RT900. It has not yet been fully implemented as of 08/24/04. Data listed is from LANL. Procedure will require modification according to WSU facilities.**

## 1. Principle of Method

1.1. Dissolved tissue samples containing isotopes of actinium are measured for gamma activity using gamma spectroscopy.

1.2. Detection limits for gamma activity are determined by using appropriate blank count data (independent of sample matrix type). These data are integrated within the specific energy areas associated with the gamma spectrum for the actinium nuclides of interest within each sample.

1.3. A tri-energetic source (NBS Mixed Radioactivity Point Source, SRM-4216, #33, using Cs-137 and Co-60 at 661.6, 1173.2 and 1132.5 keV) is used to set the energy calibration of the gamma counting system.

1.4. A set of NIST SRM-4339 #11 $^{228}$Ra standards (or similar NIST Standard Solution) in 8.0 N $HNO_3$ carrier is used to determine the peak centroid, range of integration, and counting efficiency of the detector. The set consists of five 500-mL Nalgene bottles with the same amount of standard and a varying volume of liquid in each bottle.

1.5. Quality-control records are maintained on the detector's background and on in-house quality-control samples. These records are used to monitor the detector performance and determine if the detector is out of control relative to previously established limits for background and QC samples (see Figs. 1 and 2).

1.6. A limit of detection ($L_d$) for $^{228}$Ac of 70 dis/min per sample (1.17 Bq) has been achieved at the 95% confidence level using a 500-mL sample and a 7200-s counting period (independent of sample matrix type).

## 2. Detection Limits

2.1. The limit of detection ($L_d$) for $^{228}Ac$ is a function of the instrument background counts within the integrated region of interest (ROI) of the $^{228}Ac$ peaks, the counting efficiency of the system, and the counting time for the samples.

2.2. The following equation is used to calculate the limit of detection ($L_d$):

$$L_d = \frac{2.71 + 4.65\sqrt{C_b}}{T \times E_v}$$

where $L_d$ = limit of detection,
2.71 = constant to allow for error of small-count statistics associated with Poisson distributions,
4.65 = constant to protect against Type I and Type II statistical errors at the 0.05 confidence level,
$C_b$ = background counts obtained from the analysis of chemical reagent blank quality-control samples,
T = count length (120 to 833.3 min), and
$E_v$ = volume-based efficiency.

2.3. The $L_d$ for a blank sample with a 7200-s count time is 70 dis/min per sample (1.17 Bq). This calculation assumes an average background of 21,200 counts, a 120-min count length, and a counting efficiency of 8.01% using a 500-mL sample volume. (The exact limit of detection will vary according to the background rate at the time of sample count). The $L_d$ will change as a function of the volume of the sample; it decreases as the sample volume decreases.

## 3. Accuracy and Precision

3.1. Quality-control samples (blind and non-blind) of "tissue equivalent" 8.0 N $HNO_3$ NIST SRM-4339 $^{228}Ra$ solutions are run with every set of samples.

3.2. The following table is a summary of the mean recoveries of the QC samples that were run during 1990 and 1991 (LANL data).

| Sample No. | Mean ± std dev (%) | (n) |
|---|---|---|
| #1 | 108.8 ± 13.0 | 2 |
| #2 | 101.0 ± 10.0 | 3 |
| #3 | 103.0 ± 11.0 | 3 |
| #4 | 100.0 ±8.0 | 5 |
| #5 | 97.0 ± 9.0 | 4 |
| #6 | 104.0 ± 7.0 | 3 |

## 4. Interferences

4.1. Any gamma emitter that decays with an energy between 800-1050 keV can interfere.

## 5. Apparatus

5.1. Bicron NaI(Tl) well counter with an efficiency of 48.6% at 661.6 keV, with a PHR of 7.5%. This is coupled to a Canberra Series 35 Plus 4096-channel pulse height analyzer through an external analog-to-digital converter and amplifier.

## 6. Calibration and Standards for Actinium

6.1. Use a NIST (NBS) SRM-4216 #33 point source containing $^{137}Cs$ and $^{60}Co$ isotopes as the energy calibration source.

6.2. Calibrate the detector to respond to the peak energies (centroids at the following specified channels:

| Isotope | Energy, keV | Peak Channel |
|---|---|---|
| $^{137}Cs$ | 661.6 | 662 |
| $^{60}Co$ | 1173.2 | 1173 |
| $^{60}Co$ | 1332.5 | 1332 |

6.3. Use the 500-mL NIST SRM-4339 radium-228 solution standard as the source of signal to determine correct regions of integration. Collect approximately 5000 counts (to provide adequate counting statistics) within the peak centroid response (channel 910). Set the ROIs on the lower and upper limits of the peak, where the slope is approximately zero. These integration limits will encompass the 911.1 keV line (27.7% abundance), along with the second most abundant peak at 968.9 keV (16.6% abundance). Activate the integration function on the multichannel analyzer by depressing the “integrate” key. Read the integrated counts in the lower left-hand region of the display.

6.4. Use the NIST SRM-4339 $^{228}Ra$ standard in 7.2 N $HNO_3$ (5 samples with volumes varying from 100 to 500 mL) and count each standard for 7200 s. Record the integrated counts within the specified ROI.

6.5. Calculate the absolute efficiency for each volume from the known amount of standard added to the various volumes. Graph the efficiency as a function of sample volume (Fig. 3). Determine the equation of line that best fits this response (y=a+bx), i.e., the equation of line that produces the highest correlation coefficient (R=1.0 is a perfect fit). This will allow the interpolation of an efficiency for any volume between the calculated ranges.

6.6. NAI(Tl) systems characteristically have relatively poor resolution compared to HPGE systems, but the quality of the spectra can still be a measure of the quality of

the signal. If the full width at half maximum (FWHM) is greater than approximately 100 keV, because of the complexity of the combination of the singlet and doublet, the sample should be recounted as a matter of good spectroscopic technique. Another indicator of problems is spectrum shift. If the peak centroid is not evenly distributed within the ROIs, shift has occurred and will result in erroneous calculated data. If this occurs, recalibrate the system with the mixed $^{137}Cs/^{60}Co$ point source.

## 7. Calculation of Sample Activity for Actinium-228 Content

7.1. Routine samples are counted for 7200 s. Samples analyzed over the weekend are counted for 50,000 seconds.

7.2. A sample-counting log sheet accompanies the NAI(Tl) counting system. Record all pertinent sample information (sample identification, volume or weight of the sample, date counted, the integrated counts, and comments) on the log sheet. The data is used in the calculation of the final results.

7.3. Use the following equation to calculate $^{228}Ac$ activity:

$$A = \frac{(V_s - C_b)}{T \times E_v}$$

where $A = {}^{228}Ac$ activity (dis/min per sample),
$C_s$ = sample counts within the integrated region,
$C_b$ = background counts within the integrated region,
T = count length (120 or 833.3 min), and
$E_v$ = efficiency based on volume of sample

7.4. The following equation is used to calculate the 3-sigma standard deviation associated with the result:

$$SD = 3 \times \sqrt{A}$$

where SD = standard deviation of sample activity and
A = sample activity (dis/min per sample).

## 8. Proper Waste Disposal Practices

8.1. No waste is produced using this procedure.

## 9. Source Materials

9.1. L.A. Currie, "Limits for Qualitative Detection and Quantitative Determination," *Anal. Chem.* **40**, 586-593 (1968).

9.2. W.W. Bowman and K.W. MacMurdo, "Radioactive-Decay Gammas," *Atomic Data and Nuclear Data Tables* **13**, 89-292 (1974).

9.3. A.S. Goldin, P.J. Magno, and F. Geiger, "Radionuclides in Autopsy Samples from Thorotrast Patients," *Health Physics* **22**, 471-482 (1972).

9.4. R.A. Dudley, "Bone Irradiation in Thorotrast Cases: Results of Measurements at IAEA," *Health Physics* **35**, 103-112 (1987).

9.5. H.F. Lucas, Jr., N.S. MacDonald, and J. Sweeney, "Thorium Daughters in the Spleen of a Thorotrast Case," *Health Physics* **23**, 575-576 (1972).