PASSIVE NONDESTRUCTIVE ASSAY OF NUCLEAR MATERIALS

2007 ADDENDUM

DOUGLAS REILLY – Technical Editor

The nondestructive assay reference; Passive Nondestructive Assay of Nuclear Material; Reilly, Ensslin, Smith, and Kreiner, was published in 1991 although the major technical writing was completed by 1987. This book has become widely known by the acronym PANDA. Although much of the material contained therein is still valid, there has been considerable development in the field during the intervening twenty years. The two remaining editors/authors of the original book felt that it would be valuable to produce an Addendum to cover some of the more recent developments and some of the measurement technology omitted from PANDA.

In 2002, Norbert Ensslin proposed a project to the US Department of Energy that would develop an appropriate set of additional chapters to complement the original PANDA. The DOE agreed to fund this effort and work began on the Addendum early in 2003, when Doug Reilly returned to Los Alamos from the IAEA Safeguards Training Section.

As the writing project neared its conclusion, it was decided that the materials would be issued as Los Alamos National Laboratory reports on a compact disk and as .pdf files available on the internet website of the Safeguards Science and Technology Group, N-1, at LANL. The CD is available by contacting N-1.

Chapter 1, “Gamma-Ray Detectors for Nondestructive Analysis,” LA-UR-05-3813, was written by Phyllis Russo and Duc Vo. Whereas PANDA treated only high-purity germanium and NaI(Tl) detectors, this chapter treats other alkali-halide detectors, scintillator-photodiode detectors, lanthanum scintillators, non-cryogenic semiconductor detectors, high-pressure Xe detectors, Pb-loaded scintillators, and micro-calorimeters, in addition to advances in NaI and HpGe.

Chapter 2, “Plutonium Isotopic Analysis Using PC/FRAM,” LA-UR-03-4403, was written by Thomas Sampson. It covers the theory and operation of the PC/FRAM code and its performance in interlaboratory comparisons and various actual applications. It also covers the analysis of uranium spectra.

Chapter 3, “Measurement of Plutonium and Uranium Isotopics with MGA/MGAU,” is being written by staff at Lawrence Livermore National Laboratory and is not yet available. These codes are extensively used internationally and the chapter covers theory, operation, and applications.

Chapter 4, “Tomographic Gamma-Ray Scanning of Uranium and Plutonium,” LA-UR-07-5150, was written by Steven Hansen. This covers first the basic principles of tomography with simple examples of how tomographic images are formed and how voxel (volume element) mass and opacity are calculated. It then describes the design and operation of TGS systems and the performance of a system to characterize waste sent to the Waste Isolation Pilot Project (WIPP) in New Mexico. The chapter ends with a discussion of lump-correction techniques and an innovative technique for U lump correction.

Chapter 5, “Nondestructive Assay of Holdup,” LA-UR-07-5149, was written by Douglas Reilly. This is an extension of PANDA chapter 20 that covers new corrections developed by Phyllis Russo for the Generalized Geometry Holdup (GGH) method and new applications and performance results.
Chapter 6, “Passive Neutron Multiplicity Counting,” LA-UR-07-1402, was written by Norbert Ensslin, Merlyn Krick, Mark Pickrell, Doug Reilly, and Jim Stewart. PANDA covered neutron coincidence counting, but multiplicity counting was under development when it was published. The chapter covers the mathematical theory of triple coincidence counting, detector design, existing detectors and electronics, the multiplicity shift register, and multiplicity applications and performance.

Chapter 7, “Active Neutron Multiplicity Counting,” LA-UR-07-1403, was written by Norbert Ensslin, Bill Geist, Merlyn Krick, and Mark Pickrell. Active multiplicity counting was developed to supplement active coincidence counting for $^{235}$U materials. It does not have a “closed” solution as does passive multiplicity counting, but it may offer a promising technique for samples that are not amenable to coincidence counting. The chapter covers the theory and application of the technique.

Chapter 8, “Fast and Epithermal Neutron Multiplicity Counter,” LA-UR-07-1602, was written by Mark Pickrell, Kevin Veal, and Norbert Ensslin. This chapter discusses the design and application of Epithermal Neutron (ENMC) and Fiber-Based Fast Neutron counters. Both of these counters address the problem of excessive accidentals from some samples by lowering the neutron die-away time of the counter. The ENMC is already in use and the Fiber-Based counter is still an experimental project.

Chapter 9, “Shufflers,” LA-UR-03-4404, was written by Phillip Rinard. The Shuffler uses the principle of Delayed-Neutron Activation Analysis that was investigated using pulsed neutron generators in the very early days of the Los Alamos safeguards program. Instead of a neutron generator, the shuffler uses an intense $^{252}$Cf neutron source that is moved close to the measured sample to induce fissions and quickly removed to a shield while delayed-neutrons from the fissions are counted. The chapter discusses basic theory, shuffler design, calibration, data analysis, and performance.

Chapter 10, “Principles and Applications of Calorimetric Assay,” LA-UR-07-XXXX, was written by David Bracken and Clifford Rudy. This is a more comprehensive exposition of calorimetry than is given in PANDA chapters 21 and 22. The chapter covers heat-flow calorimeter theory, operation, calibration, and performance. Calorimetry applications to plutonium and tritium are covered, in addition to recent work on calorimetry of HEU.

Chapter 11, “Useful Nuclear Data,” was compiled by Doug Reilly. This includes, among other things, a listing of nuclear data tables and figures in PANDA and Ray Gunnink’s tabulation of x rays and $\gamma$ rays from the principal isotopes of uranium and plutonium.

Index: A comprehensive topical index will be added at a later date. The following Table of Contents shows the contents of the Addendum.

Acknowledgements: I would like to acknowledge, especially, Norbert Ensslin for initiating this project and giving it his continued interest and support, even after his retirement from Los Alamos in 2005. Norbert obtained the necessary funding for the project and was a principal author of three of the chapters. This Addendum to PANDA would never exist were it not for Norbert’s efforts. Next, I wish to recognize the support and funding of the U. S. Department of Energy, SO-13/20, and the project manager, Lynne Preston. Of course none of the chapters included herein could exist without the efforts of the fifteen principal authors. Lastly, I must recognize the support and encouragement received from the N-1 group management. Thank you everybody.
# 1. GAMMA-RAY DETECTORS FOR NONDESTRUCTIVE ANALYSIS

**P. A. Russo** and **D. T. Vo**

I. Introduction and Overview 1-1

II. Sodium Iodide and other Alkali-Halide Scintillators 1-4

III. Scintillator-Photodiode Detectors 1-8

IV. Inorganic Lanthanum and Rare-Earth Scintillators 1-11

V. GE Detectors and Cryogenics 1-16

VI. Non-cryogenic Portable Semi-Conductor Detectors 1-22
   - CPG CdZnTe 1-22
   - Electrically Cooled CdTe 1-23
   - HgI₂ 1-25

VII. Gas-Filled Detectors: High-Pressure Xe Ionization Chambers 1-26

VIII. Organic Scintillators: Pb-Loaded Plastic for DNS 1-30

End Notes 1-32

References 1-35

# 2. PLUTONIUM ISOTOPIC ANALYSIS USING PC/FRAM

**Thomas E. Sampson**

I. INTRODUCTION 2-1
   - Purpose of This Chapter 2-1
   - Isotopic Analysis Applications in Nondestructive Assay
     1. Calorimetry. 2-1
     2. Neutron Coincidence Counting. 2-2
     3. Other Bulk Measurement Techniques. 2-3
     4. Process Control 2-3
     5. Treaty Verification. 2-4

II. BASIC PRINCIPLES OF GAMMA-RAY ISOTOPIC ANALYSIS FOR THE ARBITRARY SAMPLE 2-4
   - Gamma-Ray Measurement of Isotopic Ratios 2-4
   - Ratio Measurements for the Arbitrary Sample—Without Efficiency Corrections 2-4
   - The Intrinsic Self-Calibration Technique 2-5
   - The Relative Efficiency Concept 2-5
   - Relative Efficiency Models
     1. Empirical Model 2-7
     2. Physical Model 2-7
     3. Isotopic Heterogeneity 2-8

III. PC/FRAM 2-9
   - Development 2-9
   - Single Detector System 2-9
   - Choice of Detector Type 2-10
   - Shielded Samples 2-10
   - Uranium Isotopic Analysis 2-10
   - Version 4 2-11

IV. HOW FRAM WORKS 2-11
   - Obtain Data 2-11
   - Perform Analysis 2-12
1. Internal Calibrations 2-12
   a. Energy Calibration 2-12
   b. Initial Background 2-12
   c. FWHM Calibration 2-12
   d. Peak Shape/Tailing Calibration 2-13
2. Analysis of Spectral Data 2-13
   a. Calculate Peak Areas Using Response Functions 2-13
   b. Calculate Relative Efficiencies 2-14
   c. Calculate Relative Activities 2-14
   d. Calculate Isotopic Fractions 2-14
   e. Calculate Isotopic Correlation for $^{242}$Pu and $^{236}$U 2-14
V. Parameter files, the key to FRAM’s versatility 2-15
VI. FRAM USER INTERFACE 2-15
   A. File 2-15
   B. Edit 2-16
   C. Measure 2-16
   D. Options 2-16
VII. FRAM PERFORMANCE 2-18
   A. Measurement Precision or Repeatability 2-18
      1. Definitions 2-18
      2. Influencing Factors 2-19
         a. Count Rate and Throughput 2-19
         b. Electronic Settings 2-20
         c. Count Time 2-20
         d. Energy Range 2-21
         e. Detector Type 2-22
         f. Sample Characteristics 2-23
      3. Prediction of Precision in the FRAM Code 2-23
      4. Examples of FRAM’s Statistical Precision 2-24
   B. Measurement Bias 2-27
      1. Introduction 2-27
      2. Plutonium Measurement Bias 2-27
      3. Uranium Measurement Bias 2-29
      4. MOX Measurement Bias 2-29
   C. Intercomparison Exercises 2-30
      1. The PIDIE Exercise 2-30
      2. Uranium Enrichment Measurement Exercise, IRMM 1996 2-31
      3. The Pu-2000 Exercise 2-31
   D. Factors Influencing Measurement Bias 2-32
      1. Sample Composition Characteristics 2-32
      2. Branching Ratios 2-32
      3. Coincidence Summing 2-32
         a. Uranium 2-33
         b. Plutonium 2-33
      4. Peak Area Determination 2-33
         a. Background Shape 2-33
         b. Interferences 2-34
   E. Bias Correction 2-35
      1. Adjustment of Branching Ratios 2-35
      2. Observation of Peak Area Biases 2-36
3. Least-Squares Adjustment of Branching Ratios 2-36
4. Use of Standards 2-36

VIII. MAKING MEASUREMENTS FOR FRAM ANALYSIS 2-36
A. Choice of Detector 2-37
B. Choice of Energy Range 2-38
C. Collection of Pulse Height Spectra 2-38
   1. Electronics 2-38
   2. Count Rate Considerations 2-38
   3. Pulse Pileup 2-40
      a. Coincidence Summing 2-40
      b. Random Summing 2-40
   4. Filtering 2-40
   5. Shielding 2-42

IX. DIFFICULT MEASUREMENT SITUATIONS 2-43
A. Using FRAM With Rate-Loss Correction Sources 2-43
B. Simultaneous FRAM/AWCC Measurements 2-43
C. Measurements Through Thick Shielding 2-43
   1. Steel Shielding 2-43
      a. Plutonium 2-43
      b. Uranium 2-44
   2. Lead Shielding 2-46
   3. 9975 Shipping Container 2-46
D. Measurements of Am-Be Neutron Sources 2-46
E. Measuring Samples with $^{237}$Np 2-47
F. Measuring Samples with Very High $^{241}$Am 2-48
G. Measuring Heat-Source Grade $^{238}$Pu 2-49

X. FRAM APPLICATION WITH CADMIUM TELLURIDE (CdTe) DETECTORS 2-50

XI. FRAM APPLICATIONS IN AUTOMATED SYSTEMS 2-51
A. ROBOCAL 2-51
   1. Intelligent Isotopic Unit Autoanalysis 2-51
   2. Intelligent Isotopic Unit Hardware 2-52
B. ARIES NDA System 2-53

REFERENCES 2-54

3. MEASUREMENT OF PLUTONIUM AND URANIUM ISOTOPICS (MGA/MGAU)

4. TOMOGRAPHIC GAMMA-RAY SCANNING OF URANIUM AND PLUTONIUM

J. Steven Hansen 4-1

I. INTRODUCTION 4-2
II. MEASUREMENTS PRINCIPLES 4-2
   A. General Concepts 4-2
   B. Understanding Tomographic Image Reconstruction 4-6
   C. Defining the Geometry 4-10
   D. Attenuation and Emission Maps 4-12
   E. Detector Response Function 4-13
III. TGS Design Considerations 4-14
   A. Transmission Source assembly 4-15
   B. Detector Assembly 4-15
   C. Sample Positioning System 4-16
5. NONDESTRUCTIVE ASSAY OF HOLDUP

T. Douglas Reilly

I. INTRODUCTION
II. GAMMA-RAY SIGNATURES AND EQUIPMENT
III. GENERALIZED GEOMETRY HOLDUP (GGH) ASSAY METHOD
   A. Assumptions and Constraints
   B. Calibration
   C. Performing the GGH Measurement and Assay
   D. CORRECTION FOR EQUIPMENT ATTENUATION
      1. Finite Sources in Holdup Measurements
      2. Concept of a Finite Source
      3. Correcting a Measured Holdup Deposit for Finite Source Dimensions
   E. CORRECTION FOR SELF-ATTENUATION EFFECTS
      1. Self-Attenuation Effects in Holdup Measurements
      2. Determining the Self-Attenuation Correction from the Measured Areal Density
      3. Correcting a Measured Holdup Deposit for Self-Attenuation
IV. HOLDUP MEASUREMENT SYSTEM EXAMPLES
V. NEUTRON HOLDUP MEASUREMENTS
VI. ACCURACY OF GAMMA-RAY AND NEUTRON HOLDUP MEASUREMENTS
VII. SUMMARY
     REFERENCES

6. PASSIVE NEUTRON MULTIPLICITY COUNTING

N. Ensslin, M. S. Krick, D. G. Langner, M. M. Pickrell, T. D. Reilly, and J. E. Stewart

6.1. INTRODUCTION
   6.1.1 Purpose of the Chapter
   6.1.2. Definition of Neutron Multiplicity Counting
   6.1.3. Basic Principle of Neutron Multiplicity Counting
   6.1.4. Historical Reasons for Multiplicity Counting
   6.1.5. Areas of Application for Multiplicity Counting
   6.1.6. Advantages and Disadvantages of Multiplicity Counting
6.2. MULTIPLICITY COUNTER DESIGN PRINCIPLES
   6.2.1. Multiplicity Counter Design Goals
6.2.2. Monte Carlo Design Calculations 6-5
6.2.3. Figure of Merit Calculations 6-6

6.3. SOME EXISTING MULTIPLICITY COUNTERS 6-7
6.3.1. Basic Differences between Multiplicity and Conventional Coincidence Counters 6-7
6.3.2. In-Plant (Pyrochemical) Multiplicity Counter 6-7
6.3.3. Plutonium Scrap Multiplicity Counter 6-9
6.3.4. FB-Line Multiplicity Counter 6-9
6.3.5. 30-Gallon Multiplicity Counters 6-9
6.3.6. High-Efficiency Neutron Counter (HENC) 6-10
6.3.7. Epithermal Neutron Multiplicity Counters 6-11
6.3.8. SuperHENC Multiplicity Counter 6-11

6.4. MULTIPLICITY ELECTRONICS 6-11
6.4.1. Thermal Neutron Detection Electronics 6-11
6.4.2. Derandomizer Circuit 6-12
6.4.3. The Neutron Pulse Stream and Coincidence Gates 6-13
6.4.4. Predelay Circuit 6-13
6.4.5. Multiplicity Shift Register Basics 6-14
6.4.6 Deployed Multiplicity Shift Registers 6-16

6.5. MULTIPLICITY MATHEMATICS 6-16
6.5.1. Assumptions in the Equations 6-16
6.5.2. The Spontaneous Fission Process 6-17
6.5.3. Description of \( (\alpha,n) \) Reactions 6-17
6.5.4. Definition of Sample Multiplication 6-18
6.5.5. Measured Singles, Doubles, and Triples Count Rates 6-18
6.5.6. Analytical Definition of Singles, Doubles, and Triples Count Rates 6-19
6.5.7. Final Solution for Sample Mass, Multiplication, \( \alpha \) 6-20
6.5.8 Weighted Point Model Equations 6-21

6.6. MULTIPLICITY CALIBRATION AND MEASUREMENT CONTROL 6-21
6.6.1. Multiplicity Data Analysis Software 6-21
6.6.2. Detector Characterization Measurements 6-22
6.6.3. Multiplicity Calibration Procedure 6-22
6.6.4. Multiplication Bias Correction 6-23
6.6.5. Ring Ratio Diagnostic 6-24
6.6.6. Measurement Control Procedures 6-24

6.7. PASSIVE MULTIPLICITY APPLICATIONS AND PERFORMANCE 6-25
6.7.1. Expected Assay Precision 6-25
6.7.2. Typical Assay Bias 6-25
6.7.3. Plutonium Metal 6-26
6.7.4. Plutonium Oxide 6-26
6.7.5. Plutonium Scrap 6-28
6.7.6. Plutonium Residues 6-28
6.7.7. Plutonium Waste 6-29
6.7.8. Mixed Uranium/Plutonium Oxide 6-29
6.7.9. Plutonium Inventory Verification 6-29

REFERENCES 6-31

7. ACTIVE NEUTRON MULTIPLICITY COUNTING
N. Ensslin, W. H. Geist, M. S. Krick, and M. M. Pickrell
7.1. INTRODUCTION 7-1
7.1.1. Definition of Active Neutron Multiplicity Counting 7-1
7.1.2. Historical Reasons for Active Multiplicity Counting 7-1
7.1.3. Areas of Application for Active Multiplicity Counting 7-2
a. Savannah River Uranium Scrap Shuffler 9-3
b. Savannah River Billet Shuffler 9-3
c. The “Standard” 200-liter-Drum Shuffler and Its Predecessors 9-3
d. The “Pass-Through” 200-liter-Drum and Boxed-Waste Shufflers 9-5
e. Spent Naval Fuel Shuffler 9-5
f. Dounreay Reprocessing Solid-Waste Shuffler 9-6
4. Applications of Shufflers in Europe 9-7
C. Shuffler Basics 9-7
1. Delayed Neutrons 9-7
2. The Shuffler Principle 9-8
3. Pertinent Properties of $^{252}$Cf 9-9
4. Factors That Complicate Assays 9-12
II. SHUFFLER PERFORMANCE 9-12
A. Precision 9-13
B. Accuracy 9-13
C. Sensitivity or Minimum Detectable Mass 9-16
D. Choosing the Assay Time 9-17
III. CREATING A SHUFFLER 9-18
A. Basic components 9-18
1. Hardware 9-18
2. Software 9-19
3. Safety features 9-19
B. Physics Design 9-20
1. User Specifications 9-20
2. Minimum Delayed Neutron Count Rate 9-20
3. The Minimum $^{252}$Cf Mass, Detection Efficiency, and Assay Chamber Shape 9-20
4. The Initial $^{252}$Cf Mass 9-21
5. $^{252}$Cf Shielding 9-21
   a. Storage Block 9-22
   b. Assay Chamber Walls 9-22
6. Assay Chamber 9-23
7. Detector Tube Arrays 9-23
8. $^{252}$Cf Motion Requirements 9-23
IV. CALIBRATION PROCEDURES 9-24
A. Matrix Issues 9-24
B. Calculated Calibrations 9-25
C. Enrichment Issues 9-26
V. DATA ANALYSIS 9-26
A. Raw Count Rates 9-26
1. Background Counts 9-27
2. Flux Monitor Counts 9-27
3. Post-irradiation Counts 9-27
B. Background Subtraction 9-27
C. $^{252}$Cf Decay Correction 9-27
D. Flux Monitor Matrix Correction 9-27
E. Calibration Curve 9-27
F. Measurement Control 9-28
REFERENCES 9-28
10. PRINCIPLES AND APPLICATIONS OF CALORIMETRIC ASSAY

D. S. Bracken, and C. R. Rudy

I. Introduction

10-1

Uniqueness of Calorimetry NDA
Advantages
Limitations
Thermal Power Production from Radionuclides

II. Heat-Flow Calorimeter Operation, Calibration, and Calculations

Heat-Flow Calorimeters
Calorimetric Assay Overview
Mass Calculation
Specific Power
Effective Specific Power
Passive Mode
Servo Mode
Assay Error Determination
Mixed Radionuclides Example: Plutonium and $^{241}$Am Mass
Single Radionuclide Example Tritium Mass

III. Measurement Performance

Calorimetric Assay Precision and Bias Data
Calorimetry Exchange
Heat Standards
Tritium
Automated Plutonium Assay System (APAS)
Possible Sources of Bias Due to Calorimeter Design
Weight Effects
Heat Distribution Error (HDE)
Heater-Lead Error
Possible Assay Interferences
Parameters Affecting Assay Time

IV. Types of Heat-Flow Calorimeters

Water-Bath Calorimeter
Solid-State Calorimeter
Large Volume Calorimeter (LVC)
Isothermal “Air-Bath” Calorimeter
Rod Calorimeter

V. Calorimetric Assay Applications

Plutonium
$^{238}$Pu Heat Standards Calibration and Traceability
Tritium
Highly Enriched Uranium (HEU)
$^{235}$U

VI. Calorimetry Applied in Unconventional Ways

Combined Calorimetry/Neutron/Gamma-Ray Assay
Uranium Enrichment by Combined Calorimetry/Neutron Counting
Calorimetric Specific Activity Determination

VII. References
11. Useful Nuclear Data
    1. Principal NDA Gamma-Ray Signatures
    2. Major Gamma-Rays from Common Calibration Sources
    3. Major K α x rays of uranium and plutonium
    4. Gamma Rays of $^{238}\text{Pu}$
    5. Gamma Rays of $^{239}\text{Pu}$
    6. Gamma Rays of $^{240}\text{Pu}$
    7. Gamma Rays of $^{241}\text{Pu}$
    8. Gamma Rays of $^{237}\text{U}$
    9. Gamma Rays of $^{241}\text{Am}$
    10. Kα x-ray energies, intensities and intrinsic line widths
    11. Mass attenuation coefficients for various elements
    12. Spontaneous Fission Neutron Yields
    13. (α,n) Neutron Yields
    14. Thick-Target Yields from (α,n) Reactions
    15. Nuclear Data Table and Figures in PANDA

INDEX