PASSIVE NONDESTRUCTIVE ASSAY OF NUCLEAR MATERIALS

2007 ADDENDUM

DOUGLAS REILLY – Editor

The NDA 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 this 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 as a compact disk and files available on the internet website of the Safeguards Science and Technology Group, N-1, at LANL. As chapters are concluded, they will be added to the website, not necessarily in numerical order. The initial chapter to appear on the N-1 website was written by Phyllis Russo and Duc Vo entitled Gamma-Ray Detectors for Nondestructive Analysis, LA-UR-05-3813.

The following provisional Table of Contents shows the intended contents of the Addendum.

TABLE OF CONTENTS – PANDA 2007 ADDENDUM

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. Noncryogenic Portable Semi-Conductor Detectors 1-22
CPG CdZnTe 1-22
Electrically Cooled CdTe 1-23
HgI_2 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
- A. Purpose of This Chapter 2-1
- B. Isotopic Analysis Applications in Nondestructive Assay 2-1
  - 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
- A. Gamma-Ray Measurement of Isotopic Ratios 2-4
- B. Ratio Measurements for the Arbitrary Sample—Without Efficiency Corrections 2-4
- C. The Intrinsic Self-Calibration Technique 2-5
- D. The Relative Efficiency Concept 2-5
- E. Relative Efficiency Models 2-6
  - 1. Empirical Model 2-7
  - 2. Physical Model 2-7
  - 3. Isotopic Heterogeneity 2-8

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

IV. HOW FRAM WORKS 2-11
- A. Obtain Data 2-11
- B. 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 2-47
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
I. INTRODUCTION 4-1
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
D. Data Acquisition System 4-16
E. Data Analysis System 4-16
IV. Performance Evaluation in Blind Tests and Actual Facility Measurements 4-17
A. Performance Demonstration Program (PDP) 4-17
B. RFETS Pyrochemical Salts 4-19
C. Uranium Applications 4-21
1. RFETS Experience 4-21
2. Survey of Excess HEU Materials across the DOE Complex (Refs. 25-26) 4-22
3. Relative Comparison of TGS Measurements of HEU and Pu (Ref. 27) 4-22
V. Lump Correction Techniques 4-24
A. Plutonium Lump Detection and Correction Technique 4-24
B. Standard Pu Lump Correction Doesn't Work for Uranium 4-25
C. Proposed Technique for Uranium Lump Correction 4-25
VI. Commercially Available Systems 4-27
References: 4-28

5. NONDESTRUCTIVE ASSAY OF HOLDUP

T. Douglas Reilly

I. INTRODUCTION 5-1
II. GAMMA-RAY SIGNATURES AND EQUIPMENT 5-1
III. GENERALIZED GEOMETRY HOLDUP (GGH) ASSAY METHOD 5-3
A. Assumptions and Constraints 5-3
B. Calibration 5-4
C. Performing the GGH Measurement and Assay 5-5
D. CORRECTION FOR EQUIPMENT ATTENUATION 5-6
E. CORRECTION FOR FINITE SOURCE DIMENSIONS 5-6
1. Finite Sources in Holdup Measurements 5-6
2. Concept of a Finite Source 5-7
3. Correcting a Measured Holdup Deposit for Finite Source Dimensions 5-7
F. CORRECTION FOR SELF-ATTENUATION EFFECTS 5-8
1. Self-Attenuation Effects in Holdup Measurements 5-8
2. Determining the Self-Attenuation Correction from the Measured Areal Density 5-9
3. Correcting a Measured Holdup Deposit for Self-Attenuation 5-9
IV. HOLDUP MEASUREMENT SYSTEM EXAMPLES 5-9
V. NEUTRON HOLDUP MEASUREMENTS 5-11
VI. ACCURACY OF GAMMA-RAY AND NEUTRON HOLDUP MEASUREMENTS 5-14
VII.SUMMARY 5-15
REFERENCES 5-16

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
6.1.1 Purpose of the Chapter 6-1
6.1.2. Definition of Neutron Multiplicity Counting 6-1
6.1.3. Basic Principle of Neutron Multiplicity Counting 6-2
6.1.4. Historical Reasons for Multiplicity Counting 6-3
6.1.5. Areas of Application for Multiplicity Counting 6-4
6.1.6. Advantages and Disadvantages of Multiplicity Counting 6-4
6.2. MULTIPLICITY COUNTER DESIGN PRINCIPLES 6-4
6.2.1. Multiplicity Counter Design Goals 6-4
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 (α,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, α 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
7.1.4. Advantages and Disadvantages of Active Multiplicity Counting 7-2
7.2. ACTIVE MULTIPLICITY EQUATIONS 7-3
7.2.1. Overview 7-3
7.2.2. Singles, Doubles, and Triples Count Rates 7-3
7.2.3. Calculation of Sample Self-Multiplication 7-4
7.2.4. Definition of Sample Coupling 7-5
7.2.5. Determination of Uranium Mass 7-8
7.3. ACTIVE MULTIPLICITY MEASUREMENT PROCEDURES 7-8
7.3.1. Measurement Hardware 7-8
7.3.2. Measurement Software 7-9
7.3.3. Detector Characterization 7-9
7.3.4. Measurement Control and Calibration 7-10
7.3.5. Item Measurement Procedure 7-11
7.4. ACTIVE MULTIPLICITY COUNTING PRECISION 7-12
7.4.1. Calculation of Expected Counting Precision 7-12
7.4.2. Results for Thermal Neutron Counting 7-12

7.4.3. Results for Epithermal Neutron Counting 7-13
7.4.4. Results for a Future Fast Neutron Counter 7-14
7.5. ACTIVE MULTIPLICITY MEASUREMENT PERFORMANCE 7-14
7.5.1. Factors that Affect Measurement Performance 7-14
7.5.2. Uranium Metal Field Measurements 7-15
7.5.3. Uranium Oxide Field Measurements 7-16
7.5.4. Laboratory Measurements of Uranium Metal Standards 7-16
7.5.5. Uranium Metal and Oxide Modeling Results 7-18
7.5.6. Modeling of Inhomogeneous Uranium Items 7-19
7.5.7. Expectations for Other HEU Materials 7-20
REFERENCES 7-20

8. FAST AND EPITHERMAL NEUTRON MULTIPLICITY COUNTERS
Mark M. Pickrell, Kevin Veal, and Norbert Ensslin
I. Introduction and Problem Statement 8-1
II. Measurement Problems Addressed by Epithermal Neutron Counting 8-1
A. Impure Plutonium 8-2
B. HEU Materials 8-2
C. Mixed HEU/Pu 8-2
D. Additional Applications for Fast Neutron Counters 8-3
III. The Physics of High Accidentals Multiplicity Counting 8-3
IV. Fast and Epithermal Neutron Multiplicity Counters 8-9
A. Epithermal Neutron Multiplicity Counter 8-10
B. Fast Neutron Counters 8-15
V. References 8-16

9. SHUFFLERS
Phillip M. Rinard
I. INTRODUCTION 9-1
A. Purpose of this Chapter 9-1
B. The Motivation for Shufflers 9-1
1. History of Shufflers 9-1
2. The Shuffler's Role Among Other NDA Instruments 9-1
3. Applications of Los Alamos Shufflers 9-2
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 10-1
Advantages 10-1
Limitations 10-2
Thermal Power Production from Radionuclides 10-2
II. Heat-Flow Calorimeter Operation, Calibration, and Calculations Heat-Flow Calorimeters 10-3
Heat-Flow Calorimeters 10-4
Calorimetric Assay Overview 10-5
Mass Calculation 10-6
Specific Power 10-6
Effective Specific Power 10-6
Passive Mode 10-7
Servo Mode 10-9

Assay Error Determination 10-10
Mixed Radionuclides Example: Plutonium and ^{241}Am Mass 10-10
Single Radionuclide Example Tritium Mass 10-11
III. Measurement Performance 10-12
Calorimetric Assay Precision and Bias Data 10-12
Calorimetry Exchange 10-12
Heat Standards 10-14
Tritium 10-14
Automated Plutonium Assay System (APAS) 10-15
Possible Sources of Bias Due to Calorimeter Design 10-15
Weight Effects 10-15
Heat Distribution Error (HDE) 10-16
Heater-Lead Error 10-16
Possible Assay Interferences 10-17
Parameters Affecting Assay Time 10-18
IV. Types of Heat-Flow Calorimeters 10-19
Water-Bath Calorimeter 10-19
Solid-State Calorimeter 10-22
Large Volume Calorimeter (LVC) 10-23
Isothermal "Air-Bath" Calorimeter 10-24
Rod Calorimeter 10-24
V. Calorimetric Assay Applications 10-25
Plutonium 10-25
^{238}Pu Heat Standards Calibration and Traceability 10-26
Tritium 10-27
Highly Enriched Uranium (HEU) 10-27
^{233}U 10-27
VI. Calorimetry Applied in Unconventional Ways 10-30
Combined Calorimetry/Neutron/Gamma-Ray Assay 10-30
Uranium Enrichment by Combined Calorimetry/Neutron Counting 10-32
Calorimetric Specific Activity Determination 10-33
VII. References 10-33

11. Useful Nuclear Date

1. Principal NDA Gamma-Ray Signatures
2. Major Gamma-Rays from Common Calibration Sources
3. Major K x rays of uranium and plutonium
4. Gamma Rays of ^{238}Pu
5. Gamma Rays of ^{239}Pu
6. Gamma Rays of ^{240}Pu
7. Gamma Rays of ^{241}Pu
8. Gamma Rays of ^{237}U
9. Gamma Rays of ^{241}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