MASTER

GENERATION OF A MEASUREMENT CATALOG FOR SAFEGUARDS

S. C. Suda and W. Marcuse
TSO, Brookhaven National Laboratory

L. A. Kull
JRB Associates, Inc.

INTRODUCTION

During the past several years increased emphasis has been placed on the development and application of chemical analysis and nondestructive assay techniques for Safeguards measurements of special nuclear materials. The result of this work is a large collection of alternative measurement techniques for many production materials. These techniques have been applied to a bewildering array of container sizes, to materials of widely different chemical and physical forms, and in a variety of sampling patterns.

This situation places a very real burden for staying up-to-date with a rapidly changing and expanding technology on those working to assure effective Safeguards control of special nuclear material in the nuclear industry. For example, an operator of a fuel fabrication plant must attempt to keep abreast of new developments in nondestructive assay methods and techniques. As part of his operation, the plant operator would like to be able to access the applicability, efficacy and accuracy of new assay techniques. In choosing to purchase a gamma spectrometer for use in his fuel fabrication production line, the plant operator must be able to critically evaluate the assay accuracies claimed by the manufacturer. He must be aware of possible shortcomings of the method due to self-absorption, sample alignment, count rate corrections, etc. Therefore, he must either be familiar with the gamma spectrometer technique or have access to current information on the capabilities of the technique. Similarly, in deciding to upgrade his chemical analysis capability, the operator must have a rational basis for a choice among a large number of possibilities. He must be aware of possible limitations

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due to pretreatment techniques, interfering elements and calibration procedures. In either case, knowledge of the technical capabilities, measurement uncertainties, and problems associated with implementing the techniques will provide him with a basis for evaluating each alternative solution.

The preliminary work reported here results from a recognition of the need for meaningfully cataloging both available and forthcoming data on measurement techniques and existing applications, in a convenient handbook specifically designed for use in the nuclear industry. This paper is directed toward selecting the kind of data useful for inclusion in this catalog and also the format that might be used for presentation of this data.

This paper is presented with three objectives in mind. The first is to obtain feedback on the proposed selection of and presentation of the data. Assistance that is provided by those engaged in making measurements will make the format more useful and meaningful. The second objective is to alert experimenters, demonstrators and operators that data should be collected in sufficient detail so that entries such as the one used in the example are as complete as possible. The third is to request that experimenters submit their measurement results to OSMM for inclusion in the measurements catalog. The ultimate value of the catalog will be in direct proportion to the participation of those involved in making and recording the measurements.

It is envisioned that the catalog of techniques and data will consist of a collection of entries or exhibits; each one of these exhibits describing an application of a particular measurement setup to a given type of process or product material. The content of each exhibit in the catalog will include sufficient information on the measurement method and the type of material to provide the user with an understanding of the equipment, the effort required, and the quality of data which can be expected under the specified conditions. A proposed format for the presentation of data in the measurement catalog is given in Table I. A specific example of a catalog entry following this format is given in Exhibit I. (The example is not complete, but is representative of the level of detail which is useful.)

The authors have searched a number of reports attempting to construct examples such as Exhibit 1. An incomplete, but seemingly representative sampling of existing reports revealed that regardless of its origin (Safeguards R&D effort or commercial Safeguards activity), one cannot derive

enough information to complete an entry similar to that shown in the exhibit. Since none of these reports have been specifically written for cataloging, this is not a surprising finding. For example, many of the R&D reports are directed toward detailed descriptions of a new and specific aspect of their measurements, while results from production facilities concentrate heavily on the end results, omitting many of the details of the equipment and setup. It is hoped that investigators will consider the data requirements and the format of Exhibit 1 and will arrange to collect and record such data when future measurements are made.

It is hoped that investigators will present their results following a format similar to that shown in the example presented here. If these are transmitted to OSMM for inclusion in a measurement catalog, a giant step will have been taken toward the efficient dissemination of this information. The authors recognize that the proposed format is very general and that not all the items will apply to any given situation. Comments are requested from personnel operating in the field and in development laboratories for the eventual improvement and refinement of this proposed format.

TABLE I
PROPOSED FORMAT FOR
MEASUREMENT CATALOG

- I. Equipment Description
 - A. Apparatus: sensor, detector electronics, reagents, etc.
 - B. Set-up and arrangement, environment, background conditions, radiation shielding, etc.
 - C. Sample holder and scanning procedures
 - D. Measurement mode
 - E. Data output
- II. Measurement Characteristics
 - A. Properties measured
 - B. Method of measurement
 - C. Constraints, limitations and precautions

- D. Applicability
- E. Reliability of the method

III. Type of Material

- A. Chemical composition
- B. Physical form
- C. Size of lot

IV. Sampling Procedure

- A. Sampling plan
- B. Range of gram quantity per sample
- C. Packaging & containment of sample
- D. Sample matrix (if applicable)
- E. Compositing procedure (if applicable)
- F. Sample dissolution and preparation (if applicable)

V. Calibration & Corrections

- A. Measurement quality control program: standards, background, frequency and procedures
- B. Calibration curves and equations
- C. Limit of error calculations
- D. Corrections applied for drift, background, dead time, etc.

VI. Measurement Procedures

- A. Typical values for raw data and range
- B. Duration and number of determinations per sample
- C. Data recording format

VII. Data Analysis and Results

- A. Calculating equations
- B. Results of data analysis
- C. Limits of error

EXHIBIT 1

SAMPLE ENTRY - MEASUREMENT CATALOG

I. Equipment DescriptionA. Apparatus

The sensor is a neutron well counter consisting of 24 paraffin moderated BF_3 detectors, 10 inches in length, arranged in a cylindrical pattern around the sample well. The sample well is 8 inches in diameter and has a depth of 10 inches. The circuitry is that of the BNL neutron coincidence counter described in Reference 1. A block diagram of the neutron counting electronics is given in Figure 1. A photograph of the apparatus is shown in Fig. 2.*

B. Setup and Equipment Arrangement

The well counter was set up indoors where the normal background rate was about 4000 gross counts per minute or about 1/4 the counting rate of a moderate sized sample. No special precautions were taken to regulate room conditions, but the average temperature and humidity were 22°C and 50% respectively. Additional shielding was introduced consisting of .030 inches of cadmium around the counter body surrounded by 4 inch slabs of polyethylene to form a cube approximately four feet high. The space between the poly cube and the counter body was tamped with borax. This shielding reduced the average background to a net count of 2 in a five minute period. The detectors were unpoisoned.

C. Sample Holder and Scanning Procedures

No special sample holder was used. The samples were centered at the bottom of the bottom of the sample well and were stationary throughout the measurement.

D. Measurement Mode

The neutron coincident counter was run in a coincidence live time mode (rather than a clock time mode). In the live time mode,

* These figures and references are not actually included in this exhibit, but similar references or descriptions would be very useful in actual data entries.

the analysis dead time correction for the net coincidence count is made automatically by the instrument.

E. Data Output

The data from the three scalers was recorded by hand at the completion of each run.

III. Measurement Characteristics

A. Properties Measured

Neutrons are produced in the sample by spontaneous fission and in (α, n) , (γ, n) and other nuclear reactions. It is assumed that the observed coincident neutron events are due to the fissioning of plutonium isotopes present in the sample; most of the true coincidence events are due to the fissioning of ^{240}Pu .

B. Method of Measurement

The method of operation of the coincident neutron detector is given in Ref. 2.* A description of the 3 scalar outputs from the device are given below:

G = gross count - all events detected during the counting period (not corrected for dead time).

R = real + accidental coincidence count - coincidence events from spontaneous fission + accidental coincidences between unrelated events detected during the counting period (corrected for dead time).

A = accidental count - unrelated coincidence events detected during the counting period (corrected for dead time).

The quantity $N = R - A$ is the net number of coincidence events recorded during the counting period and is already corrected for dead time. It is proportional to the number of spontaneous fissions which have occurred in the sample during the counting period.

* Reference 2 is not actually included in this sample illustration.

C. Constraints and Limitations

The grams equivalent ^{240}Pu content and the coincidence count rate is not linear for samples containing greater than 50 grams equivalent ^{240}Pu . In the presence of an appreciable (α, n) count rate the statistical accuracy is degraded since the net fission counts, $N = R - A$ is now the difference between two large numbers.

D. Applicability

Neutron detection methods are in practice applicable only to material containing ^{240}Pu . The isotopic distribution of the plutonium of the standards and the unknown samples is required to calculate the grams of plutonium from the count of true coincidences. The presence of unknown neutron sources, such as any of the curium or californium isotopes, would lead to false results.

E. Reliability of the Method

Routine measurements of mixed oxide scrap and waste were good to 30% random and 10% systematic errors over a wide range of sample sizes. The observed uncertainty in the measurement of product material (both PuO_2 powder and $\text{PuO}_2\text{-UO}_2$ pellets) was about 3% random error and 3% systematic error.

III. Type of Material

A. Chemical Composition

The samples consisted of $\text{PuO}_2\text{-UO}_2$ oxide. The Pu to metal ratio was 2.94%. The fissile content ($^{239}\text{Pu} + ^{241}\text{Pu}$)/Pu was 80.14%.

B. Physical Form

The samples were pellets about 1/2 inch in diameter and 1/2 inch high. There were 18 pellets including: 6 whole pellets, 6 pellets split in half and 6 pellets were crushed and pulverized. The average pellet weight was about 10 gms.

C. Size of Lot

The measured samples were taken from a lot of approximately 5400 pellets.

IV. Sampling Procedure

A. Sampling Plan

Approximately one pellet sample was drawn per 300 pellets produced.

B. Range of Gram Quantity per Sample

The range of the net weight of the pellets was 9 to 11 grams.

C. Packaging & Containment of Sample

The pellets were encapsulated in a double unit cylindrical pellet holder measuring about 1 inch in diameter and 2 1/2 inches in height; it is shown in Fig. 3. The pellets were held down in the bottom of the cylinder with steel wool packing. (Again, Fig. 3 is not shown in this sample illustration.)

V. Calibration & Corrections

A. Measurement Quality Control Program: Standards, Background, Frequency and Procedures

Four background measurements were made each day that pellet measurements were made. Pellet number 6 was designated as a "standard" pellet and it was measured after each background measurement was made.

B. Calibration Curve and Equations

Standards of known plutonium metal and isotopic content were used to establish the relationship between the coincident counts observed and the grams equivalent of ^{240}Pu . The chemical and isotopic data needed on the standards were:

net weight (grams MO_2)

Pu concentration (Pu/ MO_2)

Isotopic abundance (% Pu-238, % Pu-240,
% Pu-242)

The grams equivalent of ^{240}Pu is calculated by:

$$\text{Pu-240 Eq} = (\text{grams Pu}) f_o$$

where grams Pu and f_o are given by:

$$\text{grams Pu} = (\text{grams MO}_2) (\text{Pu/MO}_2),$$

$$\% f_o = 3.23 (\% \text{ Pu-238}) + 1.00 (\% \text{ Pu-240}) + 1.61 (\% \text{ Pu-242}) .$$

Over the period of several weeks repeated measurements of the standards were made to generate an array of paired observations (X_i, Y_{ij}) . The X_i are the values of the calculated grams equivalent ^{240}Pu where the subscript i denotes the standard number. The Y_{ij} are the corrected coincidence counts where the subscript i denotes the associated standard and j denotes the run number. Regression analysis on the array of paired observations was used to produce the calibration curve:

$$Y = MX + \alpha$$

The slope, M , gives the average relationship between the net counts and gr Pu-240 Eq. The y-intercept, α , is a value very close to zero.

C. Limit of Error Calculations

(Unavailable at present.)

D. Corrections for Drift, Background, Dead Time, etc.

Drift

Standard sample 6 was counted 4 times a day to monitor the instrument output. No observable drift in the equipment was observed.

Background

The average background values observed for a 20 minute counting period were:

G (gross events)	10,030
R (fissions and accidentals)	26
A (accidentals only)	14
Net coincidence	12

Dead Time

When operating in the live time mode the counting period is refined by a fixed number of clock pulses. Clock pulses are inhibited during the amplifier dead time, D, and gate time, S. This gives corrected values for the R, A and N counts. However, it gives an uncorrected number for G. For these samples, only coincidence count data was used.

VI. Measurement Procedures

A. Typical Values of Raw Data and Range

	Min.	Max.	Typical
Clock time	20' 2.7"	20' 7.1"	20' 3.6"
G (gross events)	28,229	35,797	33,000
R (fissions + accidentals)	1,499	1,909	1,750
A (accidental only)	96	127	115
N (Net coincidence)	1,403	1,782	1,635

B. Duration and Number of Determinations per Sample

Twelve assays per pellet were made. The counting intervals were 20 minutes of live clock time and were taken during two 2-hour counting periods. The 2-hour counting periods were run on separate days.

C. Data Recording Format

In addition to the data shown above in VI.A, the sample number, run number and date were recorded.

VII. Data Analysis and Results

A. Calculating Equations

Grams equivalent ^{240}Pu :

$$\text{Pu-240 Eq} = \frac{N - B - \alpha}{m}$$

where

N = net coincident count

B = background

α = Y intercept

m = slope (net counts/gr Pu-240 Eq.)

Grams plutonium:

$$\text{g Pu} = \text{Pu-240 Eq}/f$$

where

f = correction for Pu-238, Pu-239 and Pu-242 in sample
 = 3.23 (% Pu-238) + 1.00 (% Pu-240) + 1.61 (% Pu-242)

B. Results of Data Analysis

(Unavailable at present.)

C. Limits of Error

(Unavailable at present.)