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SOME TARGET ASSAY UNCERTAINTIES FOR PASSIVE NEUTRON COINCIDENCE COUNTING*

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ABSTRACT

This paper provides some target assay uncertainties for passive neutron coincidence counting of plutonium metal, oxide, mixed oxide, and scrap and waste. The target values are based in part on past user experience and in part on the estimated results from new coincidence counting techniques that are under development. The paper summarizes assay error sources and the new coincidence techniques, and recommends the technique that is likely to yield the lowest assay uncertainty for a given material type. These target assay uncertainties are intended to be useful for NDA instrument selection and assay variance propagation studies for both new and existing facilities.

I. INTRODUCTION

Passive neutron coincidence counting is a versatile technique that is used to assay many forms of plutonium, including metal, oxide, impure oxide, mixed oxide, pyrochemical process materials, and other scrap and waste. Applications of the technique range from confirmatory measurements to accountability assays depending on the attainable accuracy. Recent developments in counter design and data analysis techniques, such as flat efficiency profiles and multiplicity analysis, are expected to reduce assay uncertainties in the future. Consequently, it is important to establish reasonably achievable target assay uncertainties for passive neutron coincidence counting that reflect current abilities and future state of the art. This process has already begun with the work reported in Refs. 1-3.

Estimating assay uncertainties is important for process design, variance propagation analysis, and nondestructive assay (NDA) instrument selection for both new and existing facilities. Uncertainties can be estimated either from past user experience, which is more conservative, or from the work of NDA instrument developers, which takes the new developments into account. This paper will attempt to summarize some current experience from users and developers to obtain a preliminary list of reasonably achievable target values for passive neutron coincidence counters.

II. SOURCES OF ERROR IN PASSIVE NEUTRON COINCIDENCE COUNTING

In neutron coincidence counting, the observed assay uncertainties are dominated by sample-dependent effects, or detector design imperfections, rather than by counting precision, calibration, or measurement control features. (Sample blending or preparation errors are, of course, not present because the entire sample is measured.) The fundamental problem is that the number of sample matrix effects, which includes at least sample mass, self-multiplication, and (α,n) reaction rates, exceeds the number of measured parameters, which are the total and coincident count rates.

Table I lists possible error sources in passive neutron coincidence counting, roughly in the order of decreasing relative magnitude. As mentioned above, sample matrix effects are usually the largest sources of uncertainty. Neutron self-multiplication effects can increase the observed coincidence response by 50% for plutonium oxide and by more than a factor of 10 for plutonium metal. (Alpha,n) reactions yield only random neutrons, but fissions induced by these neutrons cannot be distinguished from spontaneous fission events. The relative error in percent is approximately $200\alpha(M-1)$, where α is the ratio of (α,n) neutrons to spontaneous fission neutrons, and M is the sample leakage multiplication.

Moderating materials can bias the assay by increasing the induced fission rate in the sample and by altering the neutron detection efficiency. Sample moisture, the most commonly found moderator, can also increase the (α,n) production rate by providing additional oxygen.

Because passive coincidence counters respond to the effective ^{240}Pu content of the sample, information on the isotopic composition is required to determine the total plutonium content. This source of error may be only a fraction of a percent, if mass spectroscopy values are available, or between 1% and 2%, if 1-4 h gamma-ray spectroscopy measurements are made, or up to 5% if shorter gamma-ray measurements are made in the field with portable equipment.

Measurement uncertainties caused by variations in the detector efficiency profile across the sample volume can be large, but newer detector designs usually achieve a flat profile between 1% and 3%.

Clearly, measurement bias can always be reduced by increasing the number of representative standards available. In practice, the number of standards is always limited, and it is often necessary to assay samples with variable matrix effects using a single set of calibration standards. To obtain good assay accuracy, data analysis algorithms have been developed to measure, and correct for, sample matrix variations. These techniques are described in the next section.

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Table I. Potential Error Sources in Passive Neutron Coincidence Counting, with their Typical Approximate Magnitudes (1σ RSD)

Error Source	Approx. Relative Magnitude (%)
Sample self-multiplication/density	1-1000
Sample (α,n) reaction rate	1-20
Sample moderating material/moisture	0-10
Isotopic composition measurement	0.3-5
Counting statistics (300 s assay)	0.2-5
Detector efficiency profile variations	1-5
Calibration curve shape	0.5-3
Sample placement	0.3-3
Normalization correction	0.3-1
Container wall materials and thickness	0.1-7
Background determination	0.1-5
Sample neutron poisons	0-5
Emitted neutron energy spectrum variations	0-3
Calibration standards mass	0.0 -0.5
Electronic deadtime correction	0-5
(Alpha,n) yield coefficients	0-5
Neutron multiplicity distribution variations between Pu isotopes	0-1
Spontaneous fission rates in the Pu isotopes	0-0.5

III. PRESENTLY AVAILABLE COINCIDENCE TECHNIQUES

Table II summarizes the passive neutron coincidence assay techniques or data analysis algorithms that are available or will become available.

Most neutron coincidence counters also record the total neutron count rate. Total neutron counting is not often used for assay because it is more sensitive to sample matrix materials and room backgrounds than coincidence counting. However, this technique is occasionally useful because of its high precision and relative insensitivity to sample self-multiplication.

Conventional coincidence counting, based on the number of two-fold events collected by a shift-register-based electronics circuit,[4] is the most widely used technique. No totals count rate or sample matrix information is used to correct or adjust the observed response.

However, for samples such as plutonium metal or well-characterized oxide, in which it is possible to calculate the sample (α,n) reaction rate from the isotopic composition, a self-multiplication correction can be derived from the ratio of the total and coincident count rates.[5] This technique is designated the "known-α approach."[6]

For samples where the (α,n) reaction rate is not known, but the sample shape and density are uniform, the self-multiplication can be parameterized in terms of the ^{239}Pu mass; the "known-M approach" can then be used to correct the observed coincidence rate.[6] If sample density is not uniform, information about the sample fill height can be used to compute its density and estimate the multiplication.[7,8]

The self-interrogation technique is suitable for samples that emit (α,n) neutrons at a very high rate, so that these neutrons provide an internal source of induced fissions. If the sample shape and density are uniform, the coincidence/totals ratio can be used to determine the fissile mass.[9]

Neutron coincidence counters can be designed to have movable or removable AmLi neutron interrogation sources, so that the net active-minus-passive coincidence response can be used to determine the ^{239}Pu mass directly.[10,11] This technique requires further development to establish its potential.

Neutron multiplicity analysis determines the first three moments of the neutron multiplicity distribution from the sample.[12] The effective ^{240}Pu mass, the sample leakage multiplication, and the sample (α,n) reaction rate can be computed from these three moments. An accurate assay can be obtained without operator-declared data for the (α,n) reaction rate or the chemical composition. Neutron multiplicity counters are

Table II. Currently Available Passive Neutron Coincidence Assay Techniques
Total neutron counting
Conventional coincidence counting (no corrections)
Coincidence counting with the known-α self-multiplication correction
Coincidence counting with the known-M correction factor
Coincidence/totals ratio (self-interrogation technique)
Net active-minus-passive neutron coincidence counting
Neutron multiplicity analysis

designed to be very efficient, and the multiple rings of ^3He tubes used to obtain high detection efficiency also reduce sensitivity to the emitted neutron energy spectrum.[13]

IV. SUMMARY OF ESTIMATED TARGET UNCERTAINTIES

Target assay uncertainties [1σ relative standard deviation (RSD)] for passive neutron coincidence counting of typical plutonium materials are summarized in Table III. Measurement precision is taken as the random uncertainty due to counting statistics for a 300-s counting interval. Past user uncertainty is the total uncertainty achieved in the past using only conventional coincidence counting. Developer target uncertainty is a reasonably achievable total uncertainty using the optimum assay technique for that process material. The assay technique required to achieve this measurement uncertainty is given in the last column and is selected from the list in Table II. The target measurement uncertainties in this table do not include any uncertainty for the determination of the plutonium isotopic composition because this uncertainty can vary widely depending on the source used to obtain the isotopic information, as described in Section II above.

Guardini, et al.,[2] have defined the random, short-term systematic, long-term systematic, bias, and other components of the measurement uncertainty. For most of the materials listed in Table III, there is not yet enough information to segregate the measurement uncertainties in this fashion. The developer target uncertainty given in the fourth column should be considered an overall systematic error that propagates in a correlated fashion from sample to sample, unless the measurement precision estimated in the second column is significant, in which case this component will propagate in an uncorrelated fashion.

In Ref. 2, Guardini has also distinguished user and developer measurement uncertainties on the basis of valid factors such as available count time, standards, and calibration curves. In Table III, the distinction is instead based on past user experience and what we think can be reasonably achieved in the future using new coincidence counter designs and data analysis techniques.

The developer target uncertainties in Table III often fall in the range of 1% to 3% for well-characterized materials and 5% to 10% for scrap and waste, despite the long list of potential error sources given in Table I. These target uncertainties can be met in the future if a well-designed coincidence counter is available, if some good standards are used for calibration, if a careful measurement control program is in place, and if the optimum assay technique recommended in the last column of Table III is used. Other facility-dependent factors that can reduce measurement uncertainties are described in the following section.

V. FACILITY FACTORS IN REDUCING MEASUREMENT UNCERTAINTIES

In the future, coincidence counting measurement uncertainties may be reduced by facility processing changes as well as by improvements in NDA technology. Process improvements that change material handling procedures may improve assay accuracy as well. Some examples are given in this section.

Two important goals in pyrochemical processing are reduction in the number of process steps and reduction in the radiation dose to the operator. Recent process changes that minimize the use of fluoride and magnesium compounds reduce the neutron dose from (α,n) reactions, and thereby improve assay accuracy. Pulverizing pyrochemical sand and slag materials and segregating crucible parts, also improves assay accuracy, but increases the number of process steps and the operator exposure.

As facilities develop automated processing lines and rely more heavily on remote handling, sample containers will become more standardized, and materials will become more uniform in size, shape, density, and impurity content. This will reduce some of the error sources listed in Table I and reduce measurement uncertainties. Other sample matrix error sources will be reduced by the trend towards more administrative segregation of waste for criticality safety concerns.

VI. APPLICATIONS OF DEVELOPER TARGET UNCERTAINTIES

We believe that the developer target uncertainties listed in Table III are reasonably achievable on a routine basis for well-designed passive neutron coincidence counters that are under measurement control and that have good calibration standards available. These results are achievable if the optimum assay technique recommended in the last column of Table III can be incorporated into the data analysis software.

The developer target uncertainties represent reasonable goals for new facilities that will have access to these techniques in the future, or for existing facilities that plan to upgrade their NDA measurement capabilities. Thus these target

Table III. Summary Table of Estimated Target Assay Uncertainties (1σ RSD) for Passive Neutron Coincidence Counting of Typical Plutonium Materials*

Plutonium Process Material	Measurement Precision in 300 s (%)	Past User Uncertainty (Conv. Coinc.) (%)	Developer Target Uncertainty (%)	Assay Technique Needed to Get Developer Uncertainty
Pure metal	0.3	1-5	1-2	Known-α
Impure metal	0.3	5-10	2-3	Multiplicity
Pure oxide	0.5	1-2	1-2	Known-α
Impure oxide	0.5	3-10	1-3	Multiplicity
MOX canisters	0.5	2	1-3	Known-α
MOX powder	0.5	2	1-3	Known-α
MOX fuel pins	0.5	1	1-2	Known-α
MOX assemblies	0.5	1	1-2	Known-α
DOR salt cake	0.5	5	5	Conventional
DOR spent salt	5	5-10	5-10	Known-α
DOR salt scrub	1-3	5	5	Known-α
DORSS metal	1-3	-	3-5	Multiplicity
DOR metal product	0.3	10	2-3	Multiplicity
MSE spent salt	5	5-10	5-10	All techniques
MSE salt scrub	5	5-20	5-20	Conventional
MSESS metal	1	-	10	Self-interrogation
MSE castings	0.3	2-3	1-2	Known-α
MSE skull, crucible	1-2	20	5	Multiplicity
ER metal input	0.2	5-10	2-4	Multiplicity
ER anode heel	1	-	5	Multiplicity
ER spent salt	5	-	5-10	All techniques
ER salt scrub	5	5	5	Conventional
ERSS metal	1-2	-	3-5	Multiplicity
ER metal product	0.2	1-4	1-2	Known-α
ER skull, crucible	1-2	20	5	Multiplicity
Pu[illegible]	2-5	10-50	5-10	Self-interrogation
Scrap, noncombustible	1-3	5-25	5	Conventional
Scrap, combustible	1-3	5-25	5	Multiplicity
Sweepings	2-5	5	2-5	Multiplicity
Waste, low-α	2-5	5-10	5-10	Conventional
Waste, high-α	5-10	10-100	10-100	Conventional

*The pyrochemical processes referred to in the first column are direct oxide reduction (DOR), molten salt extraction (MSE), and electrorefining (ER), with SS denoting salt scrub materials. User and developer uncertainties are defined in the text, and do not include any uncertainty in determination of plutonium isotopic composition. Data on mixed-oxide (MOX) are from Ref. 14.

uncertainties can be used for error propagation studies to determine the usefulness of neutron coincidence counting as part of the overall materials control and accountability plan for the facility, or to determine whether coincidence counters can provide useful process monitoring information.

The target uncertainties in Table III are not complete, and many of the values are estimates that require further study. They are intended to serve as a starting point for further discussion and data collection, and we welcome the help of others in building and interpreting this data base. At this time, Table III can be used as a goal, and as a way to compare developer numbers with current user experience. Where current experience is substantially different, it will be important to determine whether the target values require revision or whether the choice of a better assay technique can yield results closer to the target values.

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