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MEASUREMENT CONTROL FOR
PLUTONIUM ISOTOPIC MEASUREMENTS
USING GAMMA-RAY SPECTROMETRY

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A B S T R A C T

A measurement control (MC) program should be an integral part of every nondestructive assay measurement system used for the assay of special nuclear materials. This report describes an MC program for plutonium isotopic composition measurements using high-resolution gamma-ray spectroscopy. This MC program emphasizes the standardization of data collection procedures along with the implementation of internal and external measurement control checks to provide the requisite measurement quality assurance. This report also describes the implementation of the MC program in the isotopic analysis code GRPAUT. Recommendations are given concerning the importance and frequency of the various MC checks in order to ensure a successful implementation of the MC procedures for the user's application.

MEASUREMENT CONTROL FOR PLUTONIUM ISOTOPIC MEASUREMENTS USING GAMMA-RAY SPECTROMETRY

John G. Fleissner

INTRODUCTION

A measurement control (MC) program is necessary for plutonium isotopic measurements to identify anomalous measurement results that may be due to instrument failure or operator (procedural) error. A pragmatic MC program involves tradeoffs between operator convenience, cost, throughput and the ability of the program to detect all possible failures. The expertise level of the operator and/or the type of isotopic measurement may also dictate the level of sophistication necessary for the MC program. The MC program should cover all phases of the plutonium isotopic measurement process, from the data collection through the calculation of the isotopic atom ratios. This report describes the philosophy and implementation of MC procedures for plutonium isotopic measurements and how these procedures have been incorporated into the isotopic analysis program GRPAUT.

CONTROL LIMITS AND RANGES

The first step in the implementation of the measurement control (MC) process is the standardization of the data collection procedures for each type of sample or measurement application. To standardize procedures, control limits or ranges should be established for the various data collection parameters such as count time, count rate, system dead time, and counting geometry. These standardized procedures should be documented and available to the instrument operator at all times. These procedures should also include such things as techniques for orienting the sample with respect to the detector(s), absorber selection, multichannel analyzer settings, data acquisition procedures, and spectral storage and archiving. Having established standardized data collection

procedures, a system of internal and external MC techniques or checks is then employed to assure the quality of the collected data and analysis methods.

Internal Techniques and Checks

Internal checks utilize parameters or measurement results from the collected spectral data of the sample item being assayed. These internal measurement control (MC) techniques are important diagnostic tools since they can help in identifying the origin of an equipment malfunction or measurement problem. An important internal check that indicates the overall hardware performance is the system resolution. The resolution of one or more peaks (for example, the 129.26 keV gamma ray of Pu-239) should be monitored on a spectrum-to-spectrum basis. Depending upon the level of sophistication of the MC program, these values could be plotted on a control chart or compared with performance limits by the analysis software. In the latter case, if the system is determined to be out of control, the software could prevent the operator from continuing until remedial action is taken. Other internal checks that may be used are the position of certain spectral peaks, and the precision of the plutonium atom ratios. Again, these parameters could be manually plotted on a control chart or monitored automatically by the measurement analysis software. Additionally, if more than one peak pair is used to obtain a particular atom ratio, the internal consistency of the atom ratios, as determined by the various peak pairs, can indicate interferences that may affect measurement control. For analysis methods that use fitting techniques, the statistical measures of "goodness-of-fit," such as chi-square, can be used with suitable control limits for MC purposes.

External Techniques and Checks

External checks rely on a comparison of isotopic results among replicate gamma-ray spectral measurements or between the spectral measurement and another assay technique. The isotopic assay of standard reference materials and working standards can indicate measurement control (MC) and system benchmarking. Measurements of the same sample on parallel instruments can also be an MC indicator. Other external techniques are comparisons of the gamma-ray results to destructive analysis results, participation in interlaboratory exchange programs, comparisons of the present data with historical or stream average data, and the re-analysis of samples at random.

A successful MC program will employ a combination of these internal and external techniques. Total reliance on an individual technique or check is not recommended. While external checks can indicate the *presence* of measurement problems, they usually cannot identify the *source* of the problem. Internal checks in many cases can provide information on the problem source and can also give an earlier warning when a measurement parameter is approaching an out-of-control state. The simpler measurement checks, such as the monitoring of the system resolution, should probably be performed on a sample-by-sample or daily basis, while other more complex techniques could be performed less frequently. The MC data provided by the internal and external checks can be used for constructing a data base for identifying and monitoring the random and systematic errors associated with the isotopic measurement system.

Measurement Control (MC) Implementation in GRPAUT Software

Many of the above described internal checks have been added to the GRPAUT software package. The GRPAUT plutonium isotopic analysis program^{1,2} is currently in use at many U.S. and international facilities. A very versatile analysis program, it can be used for a wide variety of plutonium product and scrap forms, for example: solutions, metal, oxides, recently reprocessed (nonequilibrium) materials, heterogeneous scrap forms,³ mixed

oxides, and plutonium materials having a wide range of americium content (<50 ppm to >80,000 ppm). It has been in use here at Rocky Flats for about five years. The MC checks in GRPAUT are performed by a module (MCNTRL) that is chained to the isotopic calculation module of GRPAUT. This module (MCNTRL) executes after the spectral analysis has been completed, with no additional operator input required.

The MC software performs five internal checks: (1) spectrum gain and offset, (2) system resolution, (3) reduced chi-square and figure of merit check for each of the peak multiplet fits, (4) system dead time check, and (5) reduced chi-square check for the efficiency curve fit. The MC parameters necessary for these checks are either passed from one of the three previous GRPAUT modules or are calculated directly in the MC module. The MC parameters are then compared with preset performance limits coded into the software. (Most of these preset performance limits have been empirically determined.) Some of the checks have only a pass/fail mode while others are pass/warning/fail. The various checks and modes are shown in Table 1. Failure to pass any one of checks #1, #2, #4 or #5 causes the spectral analysis to fail the MC test. Also shown in Table 1 are the values of the parameter LSTAT associated with each test. LSTAT is the variable whose value determines whether the MC test is passed. If a warning is received for MC check #2 or #5, a value of 1 is added to the LSTAT value. For each failure of MC check #3, a value of 1 is also added to LSTAT. A final value of LSTAT ≥ 5 results in a failure of the measurement control test. Thus, any combination of warnings for checks #2 or #5 and failures of check #3 that produce an LSTAT value ≥ 5 will result in a failure of the MC test.

A hardcopy output of the result of the MC checks is printed following the GRPAUT output page that summarizes the final isotopic results. Examples of this MC output are in the Appendix. The MC output page begins with the spectral file name and a summary of the spectral header information. Following this are the results for each MC check. The results are stated as either having passed, failed, or a warning issued. Below the MC check results, a statement is printed as to whether the overall MC

TABLE 1. Preset Performance Limits for the Internal Checks Employed in GRPAUT Code. Values of parameter LSTAT are in brackets.

MC Check	Mode [LSTAT]		
	Pass	Warning	Fail
#1 Gain: Offset:	<10 Channels < 5 Channels [0]		>10 Channels > 5 Channels [5]
#2 Resolution: (at 129 keV)	<601 keV [0]	>601 keV <630 keV [1]	>631 keV [5]
#3 Chi-square: FOM: (Peak Fits)	<3.6 <3.6% [0]		>3.6 >3.6% [1] per fit
#4 Dead Time:	<30% [0]	>30% <40% [0]	>40% [5]
#5 Chi-square: (Effic. Fits)	<4.0 [0]	>4.0 <6.5 [1]	>6.5 [5]

test for this spectrum and analysis passes, passes provisionally, or fails. If the test fails, a message is printed advising the operator to take immediate remedial action and to heed the warnings printed in the MC check section. Following this statement of the MC status, a section is provided for the analyst/operator to manually enter on the MC output sheet: the count rate, the absorbers used, any relevant comments, and his signature or initials. The requirement of the signature or initials guarantees that the analyst/operator has seen the MC sheet. Finally, a section is printed on the bottom of MC output sheet that lists the various MC parameters and the value of LSTAT that have been used in the

MC checks. This section is useful to the more experienced user to help identify the source of any measurement problems.

RECOMMENDATIONS

All these MC tests, implemented in the GRPAUT analysis code, are internal checks performed on a spectrum-by-spectrum basis. They should be combined with some of the external checks to form a complete MC program. Since software should not change or deteriorate with time, the external checks would assess the presence and magnitude of random errors (outside of the contribution due to counting statistics) and systematic biases, especially those due to incomplete knowledge of the fundamental nuclear constants (half-lives, gamma-ray branching ratios) and to identify interferences. Clearly, these external checks would be done less frequently than the internal checks.

As discussed above, reliance on one particular technique or check is not recommended. The internal checks should always serve as the first line of defense against an out-of-control system. Care should be exercised in the use of "absolute" reference and working standards as MC indicators. A plutonium isotopic measurement is a relative measurement; that is, it measures peak area ratios and does not require an absolute calibration. Due to the nondestructive nature of the measurement, few, if any, of the measurement parameters (such as mass, packaging, chemical form, etc.) can be controlled by the analyst. This dictates that working standards, if used for MC purposes, must resemble the actual process samples as closely as possible. If the reliance of a MC program were based solely on the isotopic measurement results of standards, then applications where a wide variety of plutonium product and scrap forms are encountered would dictate the use of an almost infinite number of reference or working standards. Clearly this is an untenable situation in most production or laboratory environments. The solution to the problem must be an MC program based on a judicious combination of internal and external techniques as applied to the user's particular applications.

REFERENCES

1. J. G. Fleissner, "GRPAUT: A Computer Code for Automated Isotopic Analysis of Plutonium Spectra," *Nuclear Materials Management, X* (Proceedings Issue), 1981, pp 461-466.
2. J. G. Fleissner, "GRPAUT: A Program for Pu Isotopic Analysis (A User's Guide)," MLM-2799, Monsanto Research Corporation, January 1981.
3. J. G. Fleissner, "Nondestructive Assay of Plutonium in Isotopically Heterogeneous Salt Residues," MLM-3124 (OP), Presented at ANS-INMM Safeguards Technology Conference, Hilton Head Island, South Carolina, November 1983.

APPENDIX

Examples of MCNTRL Model Output

1. MC Tests Passed
2. MC Tests Passed With Warnings
3. MC Tests Failed

FILENAME : FEB5,24

ANALYSIS # : 608.49
DETECTOR # : 22
START TIME : 10:31:47
START DATE : 2/12/85
SAMPLE : MNC 11931

TEST 1: GAIN/OFFSET CHECK PASSED

TEST 2: RESOLUTION CHECK PASSED

TEST 3: FITTING STATISTICS TEST PASSED

TEST 4: DEAD TIME TEST PASSED

TEST 5: CHI SQUARE CHECK PASSED

***** MEASUREMENT CONTROL TESTS PASSED *****

ENTER:

COUNT RATE : -----

ABSORBERS : -----

COMMENTS : -----

ANALYST : -----

MC PARAMETERS:

DCHAN = 1.4
D129 = 0.9
D413 = 2.3
FWHM 597.3
DTIME = 11.
LTIME = 1597.0
ETIME = 1800.0
LSTAT 0
CHISQ = 2.31

**** MEASUREMENT CONTROL SECTION ****

FILENAME : JAN5,16

ANALYSIS # : 922.53
 DETECTOR # : 22
 START TIME : 14:10:53
 START DATE : 1/13/85
 SAMPLE : MHR40

TEST 1: GAIN/OFFSET CHECK PASSED

TEST 2: RESOLUTION CHECK PASSED

TEST 3: WARNING! FITTING STATS OUT OF RANGE, REGION 1, (124.3 KEV FIT)

TEST 4: DEAD TIME TEST PASSED

TEST 5: CHI SQUARE CHECK PASSED

***** MEASUREMENT CONTROL TESTS PASSED PROVISIONALLY, SEE ABOVE WARNINGS ****

ENTER:

COUNT RATE : -----

ABSORBERS : -----

COMMENTS : -----

ANALYST : -----

MC PARAMETERS:

ICHAN = 0.1
 U129 = 0.4
 U413 = 0.3
 FWHM = 588.4
 QFIT(124.3) = 10.2 FOM(124.3) = 4.8
 DTIME = 12.
 LTIME = 1588.0
 ETIME = 1800.0
 LSTAT = 1
 CHISQ = 2.67

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**** MEASUREMENT CONTROL SECTION ****

FILENAME : FEB5,58

ANALYSIS # : 725.77
DETECTOR # : 33
START TIME : 15:49:29
START DATE : 2/15/85
SAMPLE : NLB 3768

TEST 1: WARNING! CHECK SYSTEM GAIN/OFFSET

TEST 2: WARNING! RESOLUTION AT 129 KEV = 611.9 EV
(MAY BE OK FOR HIGH COUNT RATES)

TEST 3: WARNING! FITTING STATS OUT OF RANGE, REGION 1, (124.3 KEV

TEST 3: WARNING! FITTING STATS OUT OF RANGE, REGION 16, (412.6 KEV

TEST 4: DEAD TIME TEST PASSED

TEST 5: CHI SQUARE CHECK PASSED

***** MEASUREMENT CONTROL TESTS FAILED! REMEDIAL ACTION REQUIRED! *****
***** SEE ABOVE WARNINGS *****

ENTER:

COUNT RATE : -----

ABSORBERS : -----

COMMENTS : -----

ANALYST : -----

MC PARAMETERS:

DCHAN = 9.3
D129 = 0.1
D413 = 9.4
FWHM = 611.9
QFIT(124.3) = 9.5 FOM(124.3) = 5.1
QFIT(412.6) = 15.2 FOM(412.6) = 4.1
DTIME = 3.
LTIME = 1753.0
ELIME = 1800.0
LSIAT = 8
LHISQ = 1.48