LA-UR - 73-859

TITLE: DO'S AND DONT'S OF NONDESTRUCTIVE ASSAY MEASUREMENTS

AUTHOR(S): H. O. Menlove

los alamos
scientific laboratory
of the University of California
LOS ALAMOS, NEW MEXICO 87544

Form No. 836
St. No. 2629
1/73

UNITED STATES
ATOMIC ENERGY COMMISSION
CONTRACT W-7405-ENG. 36

DO'S AND DONT'S OF NONDESTRUCTIVE ASSAY MEASUREMENTS*

by

H. O. Menlove

*Work performed under the auspices of the U. S. Atomic Energy Commission.

TABLE OF CONTENTS

DO'S AND DONT'S OF NONDESTRUCTIVE ASSAY MEASUREMENTS*

by

H. O. Menlove

ABSTRACT

This report is a preliminary guide discussing some of the problem areas and recommended procedures in the application of NDA instrumentation. To limit the scope of the present guide, only radiometric NDA techniques employing neutron and gamma signatures will be considered. Thus, measurement techniques which primarily use alpha particles, beta particles, muonic x rays, heat signatures, etc., are not included.

I. DESCRIPTION

This document on the "do's and dont's of NDA measurements" is intended as a preliminary guide discussing some of the problem areas and recommended procedures in the application of NDA instrumentation. Since many NDA techniques are undergoing further development and improvement and there is only preliminary experience in the application of existing instrumentation, the list of do's and dont's given in the following sections will not be all-inclusive. Also, to limit the scope of the present guide, only radiometric NDA techniques employing neutron and gamma signatures will be considered. Thus measurement techniques which primarily use alpha particles, beta particles, muonic x rays, heat signatures, etc., are not included.

---

*Work performed under the auspices of the U. S. Atomic Energy Commission.

II. PROBLEM AREAS COMMON TO MOST RADIOMETRIC NDA TECHNIQUES

A. Instrument Performance and Stability Checks

1. Make an initial confirmation that instrumentation is performing up to specifications of manufacturer.

2. After setting instrumentation parameters for assay application of interest, determine instrument dead time losses for the maximum expected counting rates and the dependence of dead time upon counting rate.

3. Run several measurement cycles with no sample present to confirm that the backgrounds are constant and the instrumentation has no "gliches" in the electronics. This check should be made periodically during measurement campaigns.

4. Measure one or more "working standard" periodically during the course of the measurement program to check for reproducibility of the system. Both the mean and the standard deviation of the measurement should be examined.

5. When possible, routinely monitor diagnostic information, such as ratios of signal or monitor rates, which should be constant if the system is functioning properly.

B. General Measurement Background Considerations

1. The measured response in NDA instrumentation contains some or all of the following components:

   a. desired signal proportional to isotope(s) of interest,

   b. instrument background or noise,

   c. room and possible radiation source backgrounds,

   d. assay sample backgrounds.

2. The magnitude and constancy of the above backgrounds should be determined prior to the data reduction in any NDA application.

3. It should be noted that the presence of the sample in the instrument can alter some of the measured room and source backgrounds. When this is a problem, a sample mock-up containing no fissionable or radioactive material should be used to determine its magnitude.

4. Include background effects in calculating measurement error. This is especially important when backgrounds are a large fraction of total signal.

C. NDA Standards

1. Determine the sensitivity of the instrument to the following parameters:

a. position of sample in instrument,
b. position of fissionable material in sample container,
c. size and shape of sample,
d. level of fill in sample container,
e. sample cladding,
f. sample matrix material,
g. radiation backgrounds and absorption problems in sample,
h. sample homogeneity or "lumpiness",
i. presence of absorbing or hydrogenous material in sample.

2. If any of the above effects significantly influence the measured NDA response, it is necessary to make a correction based on calculations or measurements. In some cases it is possible to use enough working standards to bracket the variable causing the problem, although this is an expensive and often impractical alternative.

3. To avoid extrapolation of the calibration curve, the physical standards should be chosen to bracket the mass range of the assay samples.

4. A destructive postmortem chemical analysis of a sample that has been through the NDA system is often useful to give credence to the applicability of the working standards. However, for many sample categories, the absolute chemical analysis has its own problems, and the intercomparison only establishes that a discrepancy does or does not exist.

5. Precisely characterized physical standards can be prepared as a check on the calibration of the assay system. The standards used in calibration should possess the same nominal physical properties as the unknowns.

III. PROBLEM AREAS FOR SPECIFIC NDA TECHNIQUES

A. Passive Neutron Methods

1. The gross counting of U or Pu samples consists of neutrons from both spontaneous fission reactions and (α, n) reactions. Since the (α, n) yields are strongly influenced by the presence of light elements such as Li, Be, F, B, C, Al, etc., it is necessary to carefully control these elements in the sample if gross counting is to be used. The primary neutron sources for some common materials is given in the following:

a. low enrichment UO_2--neutron yield dominated by spontaneous fission in ^{238}U; therefore gross counts nominally proportional to ^{238}U weight,

b. high enrichment UO_2--neutron yield dominated by ^{234}U alpha decay reacting with the ^{18}O (0.20% abundant in normal oxygen),

c. UF_6--neutron yield dominated by ^{234}U alpha decay reacting with F.

d. PuO_2--neutron yield roughly split between (α, n) reactions and spontaneous fission reactions from ^{240}Pu (minor contributions from ^{238}Pu and ^{242}Pu),

e. PuF_4--neutron yield dominated by (α, n) reactions with F.

2. Coincidence counting[1, 2] separates the spontaneous fission contribution from the random (α, n) contribution. The following lists some of the problem areas in using a neutron coincidence counter:

a. coincidence background neutrons from cosmic ray induced neutrons in the sample and matrix material--especially troublesome for low level counting of high mass samples (1 kg Pb $\Rightarrow$ 1-2 mg ^{240}Pu depending on altitude),

b. fission coincidence neutrons from multiplication of gross neutron yield in the sample--multiplication efficiency is very dependent on the mass, density, and distribution of the fissionable material in the sample,

c. plutonium isotopic composition different from that assumed in the standards or data reduction,

d. very high accidental coincidence rates resulting in errors in electronic circuitry and/or data analyses,

e. neutron moderation in the sample and matrix material[3] (especially hydrogenous materials) causing an increase or decrease in measured response--fast neutron detectors (e.g., plastic scintillators, ^4He, ZnS) are especially sensitive to these effects since scattered neutrons often drop below the neutron energy threshold of the detector,

f. for gamma sensitive detectors, there can be unwanted coincidences from n plus γ reactions or multiple γ cascades (e.g., ^{233}U and ^{232}Th samples),

g. instrument spatial efficiency variations because of end losses, etc.--(measure efficiency profile),

h. high counting rate dead time losses[2] resulting from the large coincidence time gates required for thermal neutron detectors (new electronic coincidence circuits[4, 5] have largely eliminated this problem). It should be noted that when the background is larger than the net signal, the statistical counting uncertainty can be larger than the net signal.

3. Neutron absorption and multiplication effects can be measured and corrected for using the neutron source addition method[6] (i.e., the perturbation of the assay sample on a known ^{252}Cf source in the detector is measured to determine the sample's perturbation on its own signal). The absorption portion of the correction has much in common with the γ-ray transmission correction in passive γ assay work.

4. Neutron moderation effects in the sample matrix can also be measured using a crude neutron energy spectrometer such as a combination of moderated and bare thermal neutron detectors.[7] The ratio of counts in the bare detectors positioned next to the sample and the normal moderated detectors in the counter gives a measure of the neutron slowing down and thermalization in the sample. This correction method is analogous to the use of internal line ratios to correct for passive gamma-ray absorption.

B. Active Neutron Interrogation

1. The problems connected with neutron interrogation are quite different depending on the incident neutron energy and the position of shielding material, moderators, and absorbing material for low energy neutrons (e.g., Cd, Li, and B).

2. Fast neutron interrogation[8] (0.01 - 14 MeV) is very penetrating and especially useful for large heterogeneous samples, but one should watch out for the following problems:

a. thermal and resonance energy neutrons from room return and scattering--these can be removed by surrounding sample with layer of B_4C and Cd,

b. moisture or other hydrogenous material in the sample or container--the use of a thermal neutron detector (e.g., fission detector, BF_3 tube, etc.) adjacent to the sample monitors this problem and the appropriate correction can be made,

c. the higher the incident neutron energy, the less the sensitivity to hydrogenous effects,

d. 14-MeV neutron interrogation results in delayed neutron yields[9] from oxygen ($^{17}O(n, p)^{17}N$) and beryllium ($^{9}Be(n, p)^{9}Li$)--a Pb moderator (~ 6-in. radius) around the 14-MeV neutron source can be used to lower the interrogation neutron energy below the thresholds for these background reactions.

3. Resonance energy interrogation (0.3 - 1000 eV) will show considerable self-shielding effects in the fissile materials. The use of any radioactive source together with its moderator[10, 11] and shield will have many neutrons in this energy region unless special precautions are taken (e.g., use of Li or B liner around sample in addition to Cd). The following problems may arise when using neutrons in this energy region:

a. neutron thermalization by hydrogenous materials (a little worse than for the fast neutron case above),

b. self-absorption effects in the fissile materials[12]--especially pronounced for ^{239}Pu because of the 0.3-eV resonance (Cd can be used to remove neutrons below ~ 0.4 eV),

c. selective resonance absorption by nonfissile isotopes such as ^{240}Pu causing a reduction in the observed fission rate, caused by removal of neutrons from the slowing-down process.

4. Thermal neutron interrogation (<0.1 eV) will have a large amount of self-shielding in the fissile material, cladding, and matrix, and thus it is primarily applicable to low enrichment material or uniform density fissile material. The following problems are encountered when using thermal neutron interrogation:

a. the presence of thermal neutron poisons such as Gd_2O_3 makes the measurement impossible or difficult to interpret,

b. only the surface layer is sampled in the measurement of high enrichment material,

c. the results for beads[13] (e.g., HTGR fuel) or other lumpy material will be very dependent on the size, shape, and density (e.g., U/Th ratic) of the fissile material,

d. the presence of unknown amounts of thermal neutron absorbing material such as fission products affect the measurement,

e. since the interrogation neutrons are already thermalized, the results are less sensitive to the presence of moisture or other hydrogenous material in the sample,

f. the assay results for LWR-type fuel rods and pellets are dependent on the diameter (~ 0.03%/mil), length (~ 0.1%/% length change), density (~ 0.08%/% density change), and cladding (~ 0.3%/mil S.S. and ~ 0.03%/mil zircaloy).[14]

5. For any active interrogation system, the induced fission rate is measured by counting the associated neutrons and γ rays (prompt and delayed), and these emissions are subject to the same matrix problems (absorption, etc.) as for passive assay methods. Energy sensitive neutron detectors will be more sensitive to sample matrix material than flat efficiency detectors.[3]

6. A change in the ratios or types of fissionable isotopes in the sample will change the observed response, and thus a careful selection of interrogation energies, measurable signatures, and calibration standards must be made to assay samples containing mixtures of fissionable materials.

7. When using an accelerator as a neutron source, the sample response must be normalized to a neutron flux monitor, rather than some secondary parameter such as integrated target current.

8. For pulsed neutron sources, precautions must be taken to ensure the detector electronics have fully recovered from the intense burst of interrogation radiation.

C. Passive Gamma Ray

1. Since passive γ rays and x rays are emitted by many isotopes in addition to fissionable isotopes, it is generally necessary to use γ-ray spectroscopy to isolate the signatures from the backgrounds (i.e., a G-M tube is not generally useful for quantitative isotopic measurements).

2. For applications where the backgrounds are intense or there are interfering γ-ray energies, use high resolution detectors (e.g., GeLi or intrinsic Ge) to improve the signal/background ratio.

3. For many applications, the backgrounds are dependent on the history and age since chemical separation (e.g., ^{234m}Pa in UO_2 and ^{241}Am in PuO_2). Thus the appropriate background should be measured for each sample at nominally the same time as the assay measurement.

4. When using the 185-keV γ ray for the assay of ^{235}U, the following problems should be checked:

a. cladding variations between samples and working standards,

b. inhomogeneous (layered or lumpy) distribution of uranium,

c. other γ-ray absorbing material in the container (e.g., screws, nuts, wrenches, etc.),

d. interference from ^{237}U (208 keV) in reprocessed uranium less than about 60 days old (this background peak can be resolved from the 185-keV peak using a GeLi detector),

e. because of low penetration, the 185-keV γ ray samples only the surface enrichment of the uranium,

f. variable x-ray and Compton scattering levels below the 185-keV peak.

5. In order for the "enrichment meter"[15] principle to be applicable, the following conditions should be met:

a. the fissile material in the sample must fill all of the viewing solid angle projected by the detector (i.e., the collimation area

must be less than the smallest assay sample and the sample must not have "holes"),

b. the parameters

$$\frac{\mu_m}{\mu_u}\,\frac{\rho_m}{\rho_u} \leq 0.1$$

where

μ_u, μ_m = 185-keV mass absorption coefficients of uranium and matrix, and

ρ_u, ρ_m = density of uranium and matrix

in order for the matrix correction to be less than 10%*,

c. the sample must be opaque to 185-keV γ rays,

d. the γ-ray absorption of the container wall must be known,

e. since only the surface area is sampled, the enrichment must be uniform throughout the entire sample.

6. In the measurement of enrichment in UF_6 cylinders, errors can result from the plating out of protactinium on the cylinder wall and from the presence of hex heels in the cylinders.

7. When counting the 185-keV γ ray from reactor fuel rods, pellets, or other dense samples using a γ detector (e.g., a NaI through-hole or well detector), the response corresponds to the enrichment times the surface area viewed by the detector; thus a change in the pellet dimensions will change the measurement even though the enrichment is constant. Also a change in pellet density will not affect the measurement, but the fissile content will have changed.

---

*If the absorption coefficients and densities are known, the 185-keV count rate can be corrected by the factor[15]

$$F = 1 + \frac{\mu_m\,\rho_m}{\mu_u\,\rho_u}$$

to give the enrichment of the sample.

8. Gamma-ray absorption corrections can be made using the "line ratio"[16] technique and/or the "external source transmission"[15] technique. The line ratio method consists of measuring two or more γ lines from the isotope of interest having different energies; then a change in their relative intensities corresponds to a change in the γ-ray absorption in the sample. The 129- and 414-keV γ rays in ^{239}Pu samples have been used for this purpose. The following problems can arise in using this technique:

a. complete attenuation of both γ rays by high-Z material in the sample,

b. nonuniform distribution of the fissile material (e.g., a lump of Pu in an otherwise dilute Pu sample would result in an insufficient line ratio correction),

c. high resolution detectors (e.g., GeLi) must be used to isolate the lines of interest in the ratio measurement from interfering background γ rays,

d. convenient line ratio γ rays are not available for uranium samples,

e. must know effective Z of the matrix and the proper interpretation of the line ratio can be difficult.

9. Attenuation corrections based on transmission measurements are possible if the sample can be divided by proper collimation into zones that have uniform transmission. Then the measured transmission in each zone can be used to correct the γ-ray response from that zone. Sources commonly used for the external source transmission method are ^{235}U (185 keV) and ^{169}Yb (177 and 198 keV) for ^{235}U samples and ^{239}Pu (100 - 400 keV), ^{75}Se (401 keV) and ^{137}Cs (662 keV) for ^{239}Pu samples. The following conditions will cause problems in the application of the transmission correction:

a. inhomogeneous sample material in the "slice" of the transmission collimation (i.e., any absorbing lump such as fissionable material or high-Z matrix must uniformly fill the transmission "slice"),

b. too much attenuation of the transmission source by the sample causing excessive error in the correction,

c. the use of improperly matched transmission γ-ray energy and source γ-ray energy--especially in the low energy region where the mass attenuation coefficients vary rapidly with energy and Z.

10. The "rotation collimation" technique[17] (i.e., collimating and rotating to view the central region of the sample for a longer time than the outside regions to compensate for the higher attenuation from the central region) can be used to give equal weighting to the various radial positions in the sample.

D. Active Gamma Ray (Photofission)

1. Photo-induced fission reactions can be obtained by interrogating a sample using high energy bremsstrahlung radiation from a Linac or electron microtron. The amount of fissionable material is determined by counting prompt and/or delayed neutrons induced by the photofission reactions.[18] Some of the problems encountered in bremsstrahlung interrogation follow:

a. in addition to the prompt-neutron (γ, f) signal, contributions from certain light elements (e.g., ^{2}D, ^{9}B, ^{13}C) with (γ, n) thresholds below the interrogation energy will introduce an unknown sample dependent background,

b. precautions must be taken to ensure the detector electronics have fully recovered from the intense "flash" of interrogation radiation,

c. uncertainties in the backgrounds make it difficult to use the lower intensity delayed neutron signal,

d. to measure the fissile content of uranium, the enrichment must be accurately known,

e. attenuation of the interrogation radiation (γ rays) by high-Z materials and induced signal radiation (prompt neutrons) by hydrogenous materials,

f. instrument instabilities caused by magnetic field drifts for the electron microtron,[19]

g. general complexity of accelerator operation.

IV. GENERAL RECOMMENDATIONS

1. Obtain a listing of "if statements" (i.e., conditions for valid operation of instrument) in the manual describing the assay instrument.

2. Periodically read out diagnostic information from NDA instrumentation to establish internal consistency and credence in the measurements.

3. Segregate material and use "standard" containers as much as possible to reduce the number of standards required for calibration and to decrease the measurement error.

4. Obtain NDA instrumentation that is as independent as possible of sample size, density, distribution, matrix, cladding, etc., to reduce the number of standards and uncertainty in the assay. The money saved in standards preparation and data interpretation will often more than compensate for the more sophisticated instrumentation.

5. Treat the uncertainties in the assay results in a statistically acceptable manner (i.e., including the statistical errors in the measurement and the calibration curve used to transform the measurement into an assay).

6. Reduce the chance for human errors, particularly in the case of large numbers of samples, by using automated data collection and analysis.

V. ACKNOWLEDGMENTS

The author wishes to express his appreciation to the staff of the Nuclear Analysis Research Group at the Los Alamos Scientific Laboratory for their numerous suggestions and contributions in this report.

REFERENCES

1. G. R. Keepin, "Los Alamos Scientific Laboratory Safeguards Research and Development Program," WASH-1147, p. 114, Proceedings of the AEC Symposium on Safeguards Research and Development, Los Alamos, NM (1969).

2. R. Sher, "Operating Characteristics of Neutron Well Coincidence Counters," BNL 50332, TSO, Brookhaven National Laboratory (1972).

3. H. O. Menlove, "Matrix-Material Effects on Fission-Neutron Counting Using Thermal- and Fast-Neutron Detectors," LA-4994-PR, p. 3, Los Alamos Scientific Laboratory (1972).

4. L. V. East and J. E. Foley, "An Improved Thermal-Neutron Coincidence Technique," LA-5197-PR, p. 23, Los Alamos Scientific Laboratory (1972).

5. K. Böhnel, "Two New Instruments for Nondestructive Fuel Assay with Neutrons," Trans. Am. Nucl. Soc. 15, No. 2, p. 671 (1972).

6. H. O. Menlove, R. H. Augustson, L. V. East, A. E. Evans, and G. R. Keepin, "Neutron Interrogation Techniques for Fissionable Material Assay," Proc. of Eleventh Annual Meeting of INMM, p. 316, Gatlinburg, TN (1970).

7. J. E. Foley, "4π-Neutron Counter for 55-Gallon Barrels," LA-4605-MS, p. 25, Los Alamos Scientific Laboratory (1970).

8. H. O. Menlove, R. H. Augustson, and Darryl B. Smith, "A Multi-Spectra Neutron Irradiation Technique for the Nondestructive Assay of Fissionable Materials," Nucl. Technology 10, 366 (1971).

REFERENCES (cont.)

9. H. O. Menlove, R. H. Augustson, and C. N. Henry, "Cross Section for the Delayed-Neutron Yield from the $^{17}O(n,p)^{17}N$ Reaction at 14.1 MeV," Nucl. Sci. Eng. 40, No. 1, 136 (1970).

10. D. B. Smith, "^{252}Cf Spectrum-Tailoring," LA-4457-MS, p. 16, Los Alamos Scientific Laboratory (1970).

11. R. A. Forster, H. O. Menlove, and J. L. Parker, "Fast Neutron ^{252}Cf Assay Systems," LA-4883-PR, p. 7, Los Alamos Scientific Laboratory (1971).

12. H. O. Menlove, C. D. Tesche, M. M. Thorpe, and R. B. Walton, "A Resonance Self-Indication Technique for Isotopic Assay of Fissile Materials," Nucl. Applications 6, 401 (1969).

13. H. O. Menlove, A. E. Evans, and R. A. Forster, "Assay of HTGR Fuel Rods--Fast- and Thermal-Neutron Interrogation," LA-5197-PR, p. 2, Los Alamos Scientific Laboratory (1972).

14. R. A. Forster, D. B. Smith, and H. O. Menlove, "Error Analysis of a ^{252}Cf Fuel Rod Assay System," to be published as an LA Report, Los Alamos Scientific Laboratory (1973).

15. J. L. Parker, T. D. Reilly, J. E. Foley, R. B. Walton, and L. V. East, "Passive Assay-Innovations and Applications," Proc. of Twelfth Annual Meeting of INMM, Vol. II, p. 515 (1971).

16. J. E. Cline, "A Relatively Simple and Precise Technique for the Assay of Plutonium Waste," ANCR-1055, Aerojet Nuclear Company, Idaho Falls, ID (1972).

REFERENCES (cont.)

17. J. L. Parker, R. B. Walton, and T. D. Reilly, "Gamma-Ray Scanning System for Assay of Fifty-Five-Gallon Drums," LA-4457-MS, p. 20, Los Alamos Scientific Laboratory (1970).

18. R. L. Bramblett et al., "Application of Photoinduced Reactions to Nuclear Materials Safeguards Problems," Project Summary Report, A 10826, Gulf Radiation Technology (1971).

19. L. A. Kull, M. S. Zucker, A. S. Thompson, H. R. Froelich, "Tests of an Electron Microtron for Nuclear Materials Assay Applications," Trans. Am. Nucl. Soc. 15, No. 2, 676 (1972).