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INFORMATION SYSTEM FOR NUCLEAR MATERIALS ASSAY TECHNIQUES

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Work Performed by:

J. R. Beyster (Chief Scientific Investigator)
L. A. Kull (Scientist)

JRB ASSOCIATES
P.O. Box 1393, 1250 Prospect St., La Jolla, California 92037 (714) 459-2601

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* This section is largely taken from a paper presented at the Symposium on Implementing Nuclear Safeguards, October 25-27, 1971 held at Kansas State University, Manhattan, Kansas. Proceedings to be published.

I. INTRODUCTION

During the last several years, Safeguards programs in the U. S. and abroad have generated increasing amounts of information relating to non-destructive measurements and assay techniques for fissionable isotopes. This barrage of data, including ideas for new techniques, prototype design information, operating experience, and critical technical evaluations, has been stimulated by the concerned interest of a number of national and international agencies over the effectiveness of controls for the flow of nuclear materials in the fuel cycle. In anticipation of an international community which will depend increasingly on nuclear energy to satisfy its power requirements, responsible agencies have diverted resources, time, and talent into the development of more accurate measurements techniques -- since good measurements are one of the cornerstones of effective materials control.

This paper concerns itself with a data management problem which is beginning to appear concomitant with the increase in the production rate of Safeguards information.[1] The problem can be broken down into three parts:

1. The collection of all available data.
2. The condensation and arrangement of the data into a general format to form a data base.
3. The development of a retrieval system which provides convenient access to the data base for users with specific information requirements.

This additional effort is required in order that the information being generated can be readily put to use in the variety of tasks for which it was originally intended. Administrators of government research programs, plant operators and engineers, technical people working in the measurement field, and national and international inspectors charged with enforcement of existing Safeguards regulations and agreements — all can easily be shown to benefit from a complete but condensed record of Safeguards experience.

II. DESIGN OF THE PROPOSED INFORMATION SYSTEM

As a start toward the resolution of this problem, the International Atomic Energy Agency has authorized a study to find how one might put together a Safeguards information system. In the initial effort it was decided to limit the range of assay techniques for which data would be collected to those shown to be simple to use, capable of being easily transported, and relatively inexpensive. This was done so that a limited subset of the complete information system would in itself be useful to a selected set of the potential users -- in this case, inspectors charged with the implementation of Agency Safeguards pursuant

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[1] L. Kull, S. Suda, and W. Marcuse, "Generation of a Measurement Catalog for Safeguards," INMM 12th Annaul Meeting, June 1970, (Proceedings to be published).

to agreements with Member States. Table I gives a list of the techniques which were selected within these general guidelines. The list is not considered to be complete, however it was felt that this mix does cover some of the more important techniques now receiving attention in the Safeguards community as promising non-destructive measurement methods and would be representative of the overall data management problem.

Considering the resources available, it was decided to initially collect about forty assay results, arrange the important facets from these results in an acceptable format, and use this as a data base to work from. The format into which each of these individually reported results was fitted is shown in Fig. 1. Each data sheet refers back specifically to a single reference in the published literature, however more than one data sheet may be generated from a single reference if the variety of information presented would be confusing when reduced to a single data sheet.

As the data base grew in size, various indexing systems were tried with the goal of providing convenient access to the system for specific problems. It became apparent that a relatively simple index would suffice for a system containing a small number of techniques and limited data base. However, it was also just as apparent that as the size and coverage of the system grew, more detail and flexibility in the index would be useful for extracting certain information from the data base.

The data base itself is ordered by technique (e.g., Section 1 contains all the gamma spectroscopy data sheets, Section 2 contains passive neutron, etc.), so that questions related to the characteristics of a specific assay technique can be answered by scanning the appropriate section. Each section is preceded by an introduction including the physical principles of the measurement technique, description of the measuring apparatus, and a list of references covering the general application of the technique. The present index, or variations of it, is probably sufficient for a collection of data of catalog or handbook size. The index is keyed to a description of the assay sample and is first of all broken down by elements (uranium, plutonium, thorium) as weel as including a separate section for irradiated or spent fuels (Fig. 2). Each of these sections is further divided according to the physical form of the material (bulk materials, fuel configurations, and scrap and waste). The final breakdown under these classifications is shown in Fig. 3. Notice that scrap and waste is broken down two different ways; first by the amount of material present per container and second by the container size irrespective of content.

TABLE I.

Techniques Selected for Inclusion in the Safeguards Information System

1. Gamma Spectroscopy
2. Passive Neutron Yield
3. Gamma Absorptiometry
4. Active Neutron (Isotopic Sources)
5. X-ray Fluorescence

DATA SHEET ________

TECHNIQUE:

REFERENCES:

FISSILE MATERIAL:

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

ACCURACY SENSITIVITY PRECISION

SNM CHEMICAL & PHYSICAL FORM:

MATRIX MATERIALS:

CONTAINER DESCRIPTION:

HOMOGENEITY:

SELF SHIELDING:

BACKGROUNDS:

MEASURING PROCEDURES & THROUGHPUT:

STANDARDS & CALIBRATION:

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS:

OTHER APPLICABLE REFERENCES:

Fig. 1. Format for each individual result in the data base.

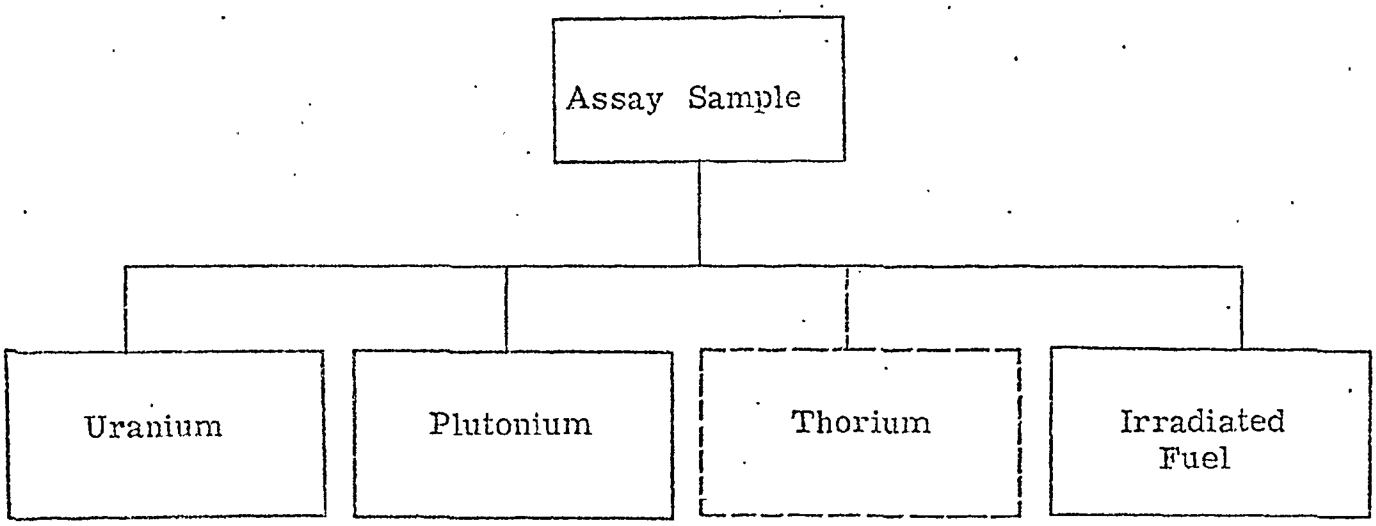

Figure 2. Indexing System — Breakdown by Assay Sample Composition

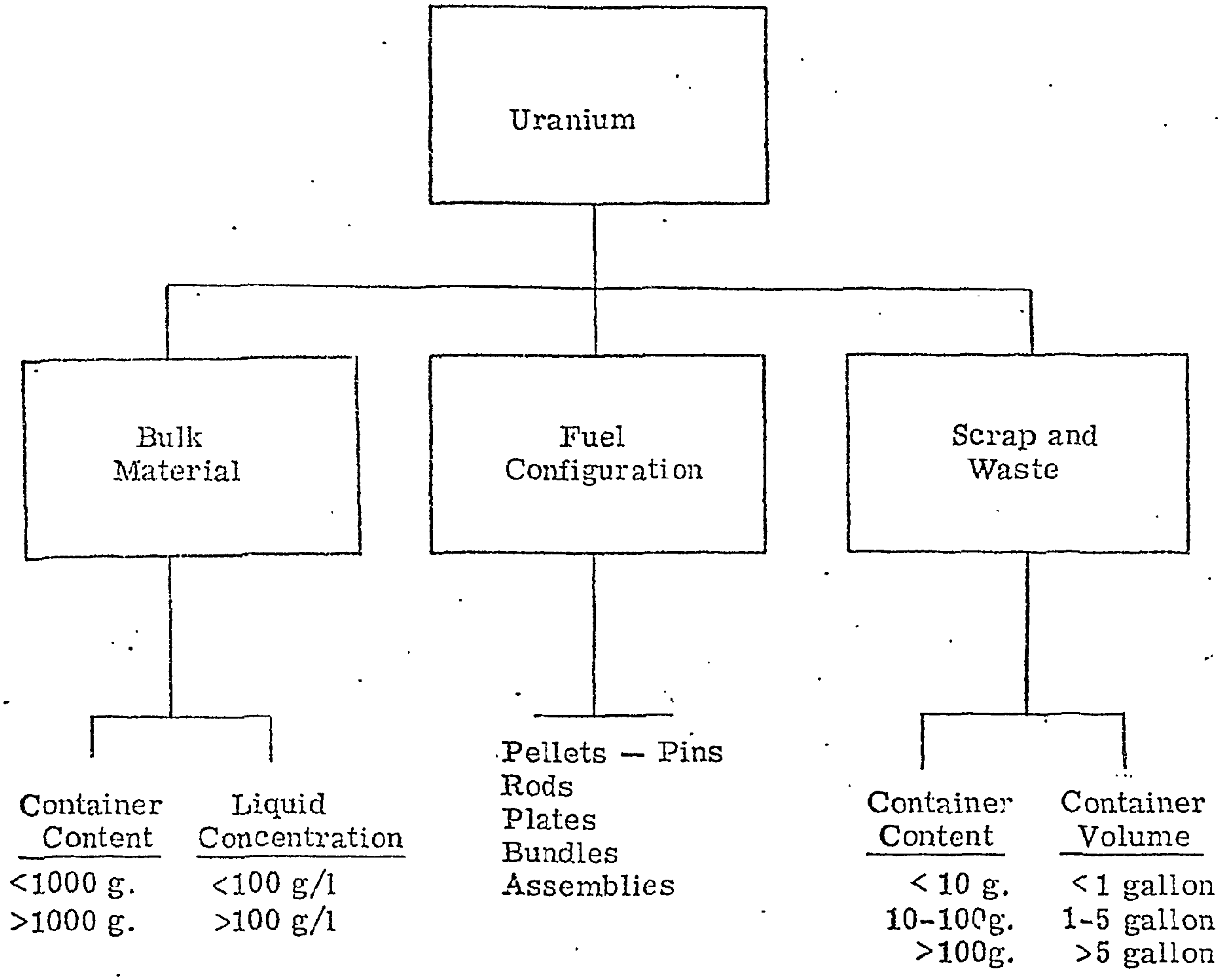

Figure 3. Indexing System — Further Breakdown by Assay Sample Description

As an example of how this index could be used, assume that a particular query pertains to the assay of plutonium waste in 1 gallon containers with an expected material content of several grams per container. With this information, the present limited data base would refer the user to one data sheet on gamma spectroscopy and three data sheets using the method of passive neutron yield. These data sheets in turn contain references to five papers available in the literature for elaboration of the information contained on the data sheets. It is apparent from the above description that the existing Safeguards information system consists of a handbook or catalog containing a limited data base of essential details from published assay results. Convenient access to the information contained therein is attained by both the ordering of the data sheets and the use of an indexing system. The system is simple, but it illustrates the usefulness of the concept.

III. FUTURE NEEDS

It is appropriate now to consider what lies ahead. The first and most immediate need is to expand the data base under the present limited set of techniques and to get this catalog into the field. There it can be improved upon and refined, based on suggestions and complaints from the users. Secondly, the system should broaden out in scope so as to include all important assay techniques now in use or under serious development. Finally, once the coverage of the system has been expanded, a responsibility needs to be established to update the information system on a periodic basis. Once the systems comes "up to speed", i.e., all the relevant existing data has been fed into the system, the reviewers can also spend some time insuring that all the possible pertinent data from newly reported measurements or operating experiences is recorded. There are important omissions in many earlier reports which might possibly have been avoided if the authors had been contacted soon after publishing their data.

As the data base expands and the number of techniques grows, it may be more effective to move from the "catalog" stage and program the information system on a computer. This is especially true if it becomes desirable to query the system in considerably more detail than was presented here. For example, if the same problem were put to the system as was mentioned earlier and 30 data sheets for a variety of techniques were referenced, it might be more effective to refine the problem further by defining the range of accuracy desired, isotopic content, enrichment, etc. As the data base develops, it may also be desirable to subdivide the data under additional major headings such as mixed oxides, burnup determination, etc.

The effort described here is a small, but significant step toward more effective management of the flow of Safeguards measurement data. The problem of collection, evaluation, display and dissemination of this information concerns us all -- even though we work on different facets of the Safeguards problem. Stated very simply, accurate and up-to-date knowledge of our ability to measure forms a vital background for any decisions which are made with the object of bringing about more effective control of nuclear materials.

INDEX - URANIUM*

BULK MATERIAL

(Index by container content)

<1000 g/container

2-07 (238)
4-02
4-07
4-08
5-02 (U)

>1000 g/container

2-07 (238)
4-02
5-02 (U)

(Index for liquids by concentration)

<100 g/l

3-02 (U)
4-01
5-01 (U)
5-04 (U)

>100 g/l

3-01 (U)
3-02 (U)

FUEL CONFIGURATIONS (UNIRRADIATED)

Pellets and Pins

4-05

Plates

3-03 (U)

Rods

1-08 (^{235}U enrichment only)

Bundles and Assemblies

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* Unless denoted otherwise, indices refer to ^{235}U assays; e.g. (238) = ^{238}U, (U) = uranium (no isotopic discrimination).

INDEX - URANIUM*

SCRAP AND CONTAMINATED WASTE

(Index by material content per container)

0 - 10 g/container

4-04

10-100 g/container

4-03
4-04
4-06
4-07

>1000 g/container

1-09 (^{235}U enrichment only)
1-10 (^{235}U enrichment only)
4-02
4-04

(Index by container size)

<1 gal. container (41.)

4-02
4-03
4-04
4-07

1-5 gal. container (4-19 1.)

1-10 (235 enrichment only).

>5 gal. container (>19 1.)

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* Unless denoted otherwise, indices refer to ^{235}U assays; e.g. (238) = ^{238}U, (U) = uranium (no isotopic discrimination).

INDEX-PLUTONIUM*

BULK MATERIAL

(Index by container content)

1000 g/container	1000 g/container
2-08	1-01 2-08

(Index for liquids by concentration)

100 g/l	100 g/l
5-03	1-01

FUEL CONFIGURATION (UNIRRADIATED)

Pellets and Pins

2-01

Plates

1-06
1-11

Rods

4-06

Bundles and Assemblies

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*Unless denoted otherwise, indices refer to plutonium assays in which ^{239}Pu is the predominent isotope; e.g. (238) = ^{238}Pu.

INDEX - PLUTONIUM*

SCRAP AND CONTAMINATED WASTE

(Index by material content per container)

0-10 g/container

1-02
1-03
1-04
1-05
1-07 (238)
1-12
2-02
2-03
2-05
2-09

10-100 g/container

1-03
2-02
2-04
2-05
2-06

100 g/container

2-05

(Index by container size)

<1 gal. container (41).

1-07 (283)
1-12
2-02
2-03
2-04
2-05
2-06

1-5 gal. container (4-19 1.)

1-02
1-07 (238)
1-12

>5 gal. container (19 1.)

1-03
1-04
1-05
1-07 (238)
1-12

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*Unless denoted otherwise, indices refer to plutonium assays in which ^{239}Pu is the predominent isotope; e.g. (238) = ^{238}Pu.

INDEX - IRRADIATED MATERIAL

Fuel Configurations

1-13
1-14
1-15
3-04

Processed Fuel

5-03

Scrap and Waste

TECHNIQUE 1 - GAMMA SPECTROSCOPY

I. PHYSICAL PRINCIPLES OF MEASUREMENT

All nuclear materials and their daughter elements emit relatively small, but measurable amounts of gamma radiation following spontaneous alpha and beta decay. The gamma radiation is emitted at discrete energies, typically in the range of 50 to 2500 keV, and the complete spectrum from a given isotope (the intensities and energies of the emitted gamma rays) is unique to that isotope. Energies and intensities of important gamma rays from common nuclear materials are given in Ref. 1-1. Measurements of the gamma ray spectrum from nuclear materials can be used to identify the isotopic composition and determine the quantities of materials in the sample.

Nuclear material which has been irradiated, for example, a "hot" fuel element which has been in a reactor, falls in a separate and distinct category. It typically emits considerably larger amounts of gamma radiation, the intensity and spectrum of which is dependent upon the sample's irradiation history. This gamma radiation originates primarily from the decay of fission products; the energy range for important gamma rays is roughly the same as that for "cold" nuclear material (above). After cooling periods of from several weeks to several months, useful information regarding the burnup, irradiation history, and (indirectly) the material content of the irradiated sample can be obtained from measurements of the gamma spectrum from "hot" samples.

Absorption of the gamma rays within the sample is an important consideration in determining the effectiveness of this assay technique. Due to higher gamma attenuation coefficients at the lower energies, materials such as ^{235}U, which emit prominent gamma rays at lower energies (185 keV), are more affected by absorption than materials such as ^{239}Pu, which emit prominent gamma rays at higher energies (385 to 414 keV).

II. APPARATUS

High resolution lithium drifted germanium detectors (Ge(Li)) and lower resolution sodium iodide (NaI) detectors are commonly used to measure the gamma spectrum from assay samples. A block diagram of a typical detector system is shown in Fig. 1.1.

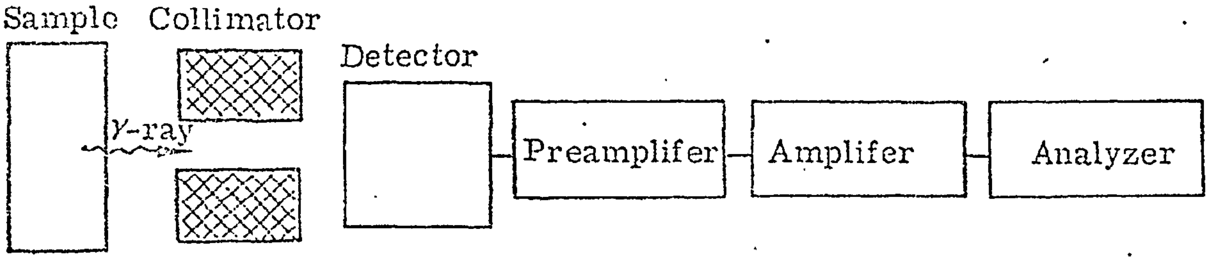

Fig. 1.1. Passive gamma assay system.

Samples can be rotated (drums) or scanned (fuel rods) in order to minimize effects due to non-homogeneous distributions of material within the sample or to obtain data as a function of sample position.

If desired, detectors can be collimated with lead, tungsten, or similar materials to view only a selected portion of the sample. Ge(Li) detectors have excellent energy resolution ($^{\Delta E}/_{E}$) and are useful for discriminating between gamma rays from different materials when their respective gamma lines are close together in energy. NaI detectors have lower energy resolution, but are more efficient. They are most useful for detecting small amounts of material in situations where confusion with contaminants is considered unlikely. The analyzer can be a multichannel device which displays the entire measured spectrum of gamma rays or one or more single channel devices which measure the gamma ray intensity over selected energy intervals of interest.

An important measurement problem for this technique is background gamma rays from other materials which are close in energy to the gamma rays of interest. These can cause erroneous readings if the energy resolution of the system ($^{\Delta E}/_{E}$) is not capable of resolving the separate lines (for example, the weak ^{240}Pu line at 160.3 keV can be masked or confused with the ^{239}Pu line at 161.5 keV). Another common problem is that of excessively high count rates due to a large number of gamma rays at energies lower than the gamma rays of interest. This can be reduced by placing layers of selected absorbing materials between the sample and detector and the use of detector circuitry specifically designed for high counting rates. For large samples, errors in the calculated sample content introduced by unhomogeneities in the distribution of nuclear material, can be minimized through the use of selected collimators and sample rotation. In some cases, data can be taken as a function of position to pinpoint high concentrations of material or to check the homogeneity of the material.

More detailed information on the general use of gamma spectrometers is contained below in references 1-2 through 1-5.

III. REFERENCES

1-1 J. E. Cline, "Gamma Rays Emitted by the Fissionable Nuclides and Associated Isotopes", IN-1448 (1970).

1-2 F. S. Goulding and Yvonne Stone, "Semiconductor Radiation Detectors," Science 170 (1970) 280.

1-3 J. E. Cline, "Studies of Detection Efficiencies and Operating Characteristics of Ge(Li) Detectors," IEEE Trans. Nuc. Science (1968) 198.

1-4 K. Seigbahn (editor), Alpha-, Beta-, and Gamma-Ray Spectroscopy, North Holland, Amsterdam (1968).

1-5 W. J. Price, Nuclear Radiation Detection - Second Edition, McGraw-Hill, New York (1964).

DATA SHEET 1-01

TECHNIQUE: Gamma-Spectroscopy

REFERENCE: J. E. Cline, et. al. - "A Technique for Assay of L-10 Bottles of Plutonium Nitrate", IN-1433 (1970).

FISSILE MATERIAL: Plutonium QUANTITIES PRESENT: 100 to 250 g Pu/l
^{239}Pu/^{240}Pu ratio - 5 to 11
^{239}Pu/^{241}Pu ratio - 15 to 70

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Accuracies	Reproducibility
Total Pu Content - 4 to 5%	<2%
^{239}/241 ratio - ~5%	
^{239}/240 ratio - ~10%	

Several bottles had unexplained differences between measured and quoted values of 7-9% for total Pu, 12-29% for 240, and 5-7% for 241.

SNM CHEMICAL & PHYSICAL FORM: Highly acidic ~5M solutions of plutonium nitrate (chemically separated from spent fuel elements).

MATRIX MATERIALS:

CONTAINER DESCRIPTION: 10 l. polyethylene bottles, (0.1" thick, 5.47" I.D., ~50" height) usually stored in stainless steel cylinders (.075 or .135 inches thick).

HOMOGENEITY: Good

SELF SHIELDING: Once the Pu density of the liquid has been determined by the ratio of the 129.3 and 413.7 KeV lines from ^{239}Pu, a previously calibrated self shielding correction is applied to the measurements of the ratios of the 129.3/148.6 KeV lines and 129.3/160.3 KeV lines to determine the ^{239}/241 and ^{239}/240 ratios respectively. (Separate corrections are applied for stainless steel cylinders of different thicknesses).

BACKGROUNDS: Flat Compton backgrounds were assumed under the peaks of interest and subtracted.

MEASURING PROCEDURES & THROUGHPUT:

Detector - uncollimated, ~2cc high resolution Ge(Li) (0.75 KeV FWHM @ 129 KeV)
Detector to Bottle ℄ - 54 in.
Count Time - 1000 seconds
Throughput - 30 minutes/bottle
Pu density-determined by ratio of 129.3/413.7 KeV lines
240/239 ratio determined by ratio of 129.3/160.3 KeV lines
241/239 ratio determined by ratio of 129.3/148.6 KeV lines

STANDARDS & CALIBRATION: The system was calibrated over the range of 100 to 300 g Pu/l, weight ratio 239/240-5 to 11, and weight ratio 239/241-14 to 54. Standards were analyzed by destructive methods. Uncertainties in the standards were estimated to be:

Total Pu ± 0.5 g/l .
238 ± .008 (weight %)
239 ± .1
240 ± .08
241 ± .05
242 ± .02

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS: A high resolution Ge(Li) detector is required to resolve the weak 160.3 KeV peak (^{240}Pu) from a peak at 161.5 KeV (^{239}Pu). Statistical uncertainities are large for the 239/240 determination due to the weak intensity of the 160.3 KeV peak. If no chemical separations have been performed for one month prior to assay, the strong 208.0 KeV peak from ^{237}U (a ^{241}Pu daughter) can be used to determine the ^{241}/239 ratio.

OTHER APPLICABLE REFERENCES: J. E. Lovett and D. B. James, PIP Progress Report Oct. - Dec. 1970, BHO-69-3.

DATA SHEET 1-02

TECHNIQUE: Gamma-Spectroscopy

REFERENCE: R. A. Deal, J. F. Gettings, D. B. James, "Proceedings-Tenth Annual Meeting, Inst. Nuclear Material Management 61 (1969) p. 62.

FISSILE MATERIAL: Plutonium

QUANTITIES PRESENT: 0.05 to several grams PuO_2 per package

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Statistical Precision: ~3% for 60 mg PuO_2; ~1% for 1 g PuO_2.

SNM CHEMICAL & PHYSICAL FORM: Plutonium oxides.

MATRIX MATERIALS: Solid glove box wastes (PVC bags, cardboard, rubber gloves, graphite and ceramic crucibles, metal turnings and oxides, glass jars).

CONTAINER DESCRIPTION: 1.5 gallon cardboard packages.

HOMOGENEITY: Poor

SELF SHIELDING: A self shielding correction is applied to the results by comparing the response from a Pu source shining through an unknown barrel, with that from a barrel of known matrix content. The correction factor for self attenuation ranged from 0.88 (cardboard) to 1.16 (gloves, glass jars). This correction was relatively effective as compared to a similar measurement on 55 gallon drums.

BACKGROUNDS: It was determined that the most accurate background correction was made by subtracting (1) a flat Compton background and (2) a previously measured environmental background from under the peaks of interest. The background under the Pu " 384 KeV complex" was 890 ± 27 counts/1000 seconds.

MEASURING PROCEDURES & THROUGHPUT:

Detector - collimated 55 cc Ge(Li) (4 KeV FWHM @ 1.33 MeV)
Detector to sample ℄ - 11.5 in.
Sample Rotation - 2 rpm
Counting Period - 800 seconds without exterior Pu source
800 seconds with exterior Pu source

The amount of ^{239}Pu is determined from the measured gamma intensity of the Pu" 384 KeV complex".

STANDARDS & CALIBRATION: Standards were made with known amounts of PuO_2 distributed throughout a 1.5 gallon can containing a PVC bag (1.25 lb.). The calibration range was .06 to 8 g PuO_2. The calibration curve is slightly nonlinear on a log-log plot; the calibration is approximately 10^4 net counts/800 seconds·g PuO_2.

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS:

1. Difference between NaI and Ge(Li) measurements of the same scrap:

   (Ge(Li) - NaI) standard deviation of differences ± 0.18 g (<1g PuO_2); ± 0.68 g (>1 g PuO_2).

   (Ge(Li) - NaI) mean difference .07 g (<1g PuO_2); 0.54g (>1g PuO_2).
2. An 18 mil hafnium absorber was placed between the sample and detector.

OTHER APPLICABLE REFERENCES: J. E. Lovett, D. B. James, PIP Second Quarterly Report, BHO-69-2 (1970).

DATA SHEET 1-03

TECHNIQUE: Gamma-Spectroscopy

REFERENCE: J. Lovett, PIP Quarterly Progress Report, April-June 1970, BHO 69-1.

FISSILE MATERIAL: Plutonium QUANTITIES PRESENT: 1-60 gms Pu per barrel.

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Accuracy	Sensitivity	Precision
Measured Pu count rates can differ by up to 12% depending on the Pu location in the drum.	equivalent to approximately one 0.2 gm pellet of PuO_2	approximately 3.4% (2σ)

SNM CHEMICAL & PHYSICAL FORM: Plutonium oxides.

MATRIX MATERIALS: Solid glove box wastes (PVC bags, Nyo gloves, latex gloves, glass jars).

CONTAINER DESCRIPTION: 55 gallon drum. Each drum held 21 1.5 gallon cardboard scrap containers; the package arrangement was 3 layers of a ring of six with one in the middle.

HOMOGENEITY: Very poor.

SELF SHIELDING: A self shielding correction is applied to the results by comparing the response from a Pu source shining through the unknown drum, with that from a drum containing a know matrix. Relative self shielding factors ranged from .991 to 1.014 for a variety of sample matrices. This correction is considered relatively ineffective due to the fact that the source shines through gaps in the drum contents regardless of their specific composition. It indicates a strong need for a scrap segregation prior to drum loading.

BACKGROUNDS: The background correction subtracted from under the peaks of interest consisted of a flat Compton background plus a previously measured environmental background. The background under the Pu "384 KeV complex" was 1220 ± 44 counts/1000 seconds. (The signal was 1200 counts/sec. g Pu).

MEASURING PROCEDURES & THROUGHPUT:

Detector - 55 cc Ge(Li)
Detector to Barrel ℄ separation - 54 in.
Barrel Rotation Rate - 4 RPM
1000 sec. count of unknown drum.
1000 sec. count of unknown drum with exterior Pu source in position for attenuation correction.

The amount of ^{239}Pu is determined from the measured intensity of the Pu "384 KeV complex".

STANDARDS & CALIBRATION: Four standard packages were made up containing known amounts of PuO_2 and were interspersed in the drum with dummy packages containing typical matrix materials. The calibration range was 0.2 to 19 grams PuO_2. The measured calibration coefficient for the system was 1.12 (g ^{239}Pu)(seconds)/(net count).

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS:

1. Scrap should be segregated before being placed in drums.
2. More accurate results can be obtained by assaying individual packages before they are placed in the drum. See data sheet 1-02.
3. If packages can be screened for average attenuation prior to insertion in drums, assays of drums are considerably more accurate.

OTHER APPLICABLE REFERENCES:

DATA SHEET 1-04

TECHNIQUE: Gamma-Spectroscopy

REFERENCE: J. E. Lovett, D. B. James, PIP Second Quarterly Report, BHO-69-2 (1970).

FISSILE MATERIAL: Plutonium QUANTITIES PRESENT: 1-12 g ^{239}Pu

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Precision: 1.6% (drums containing 2 filters)
9.1% (1 large filter, or 1 filter on end)

SNM CHEMICAL & PHYSICAL FORM: Plutonium oxides - solid grit.

MATRIX MATERIAL: 12 x 12 x 18 in. and 24 x 24 x 24 in. exhaust filters.

CONTAINER DESCRIPTION: 55 gallon drums loaded with filters (described above).

HOMOGENEITY: Fair to poor.

SHIELDING: Since all the filters are of the same construction, attenuation of the gamma radiation was assumed to be equal for all cases. Therefore, no separate attenuation measurements were made.

BACKGROUNDS:

MEASURING PROCEDURES & THROUGHPUT:

Detector - 55 cc Ge(Li)
Detector to Drum ℄ separation - 54 in.
Drum Rotation Rate - 4 rpm
Counting Period - 1000 seconds

The amount of ^{239}Pu is determined from the measured intensity of the Pu "384 KeV complex".

STANDARDS & CALIBRATION: Standards were made by encapsulating one to five g of PuO_2 in plastic bags and randomly distributing them inside virgin filters. The systems were calibrated separately (a) for one 12 x 12 x 18 in. filter/drum or one 24 x 24 x 24 in. filter (0.6 net counts/sec·g ^{239}Pu) and (b) for two 12 x 12 x 18 in. filters/drum (0.85 net counts/sec · g ^{239}Pu). Calibrations were linear over the range 1-12 g ^{239}Pu.

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS:

OTHER APPLICABLE REFERENCES:

DATA SHEET 1-05

TECHNIQUE: Gamma-Spectroscopy

REFERENCE: J. C. Limpert, PIP Quarterly Progress Report, WCAP-7562-3.

FISSILE MATERIAL: Plutonium QUANTITIES PRESENT: 0.5-10 g Pu

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Accuracy: ± 10% from 0.5 to 10 g Pu (segregated scrap)
± 25% from 0.5 to 10 g Pu (non-segregated scrap)

SNM CHEMICAL & PHYSICAL FORM: Plutonium oxides and plutonium-uranium mixed oxides.

MATRIX MATERIAL: Chemical processing - 90% moist paper and plastics, 8% Speedi-Dri absorbent, 2% neoprene gloves.

Fuel fabrication waste - 99% dry paper and plastic

Analytical waste - 30% moist Speedi-Dri and 70% paper and plastic

CONTAINER DESCRIPTION: 55 gallon drum

HOMOGENEITY: Poor

SHIELDING: The ratio of the 414 and 129 KeV ^{239}Pu lines was used to correct for self absorption and absorption in the non-fissile matrix.

BACKGROUNDS:

MEASURING PROCEDURES & THROUGHPUT:

Chemical Processing and Analytical Waste	Fuel Fabrication Waste
Detector - Ge(Li)	Detector - NaI
Counting Period - 1000 seconds	Counting period - 400 seconds

The Pu content is determined from the measured intensity of the 414 keV line.

STANDARDS & CALIBRATION: Two sets of standards were used which were typical of laboratory wastes: (a) 3 w/o PuO_2 - UO_2 mixed oxides and (b) PuO_2. Dummy packages were made up for calibration purposes.

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS:

1. Assay accuracies for individual 1 gallon packages (which are eventually placed in drums) are about a factor of two better than that for 55 gallon drums.

2. High resolution Ge(Li) detectors are required for assays of chemical and analytical wastes. Scattered gamma rays from intense lines at 511 KeV (^{106}Ru/^{106}Rh, ^{208}Tℓ) and 624 KeV (^{106}Ru/^{106}Rh) obscure the Pu peaks of interest in lower resolution NaI detectors.

OTHER APPLICABLE REFERENCES: J. C. Limpert, PIP Quarterly Progress Report, WCAP, 7562-4.

DATA SHEET 1-06

TECHNIQUE: Gamma-Spectroscopy

REFERENCE: N. S. Beyer, et. al. Proceedings from Institute of Nuclear Materials Management Tenth Annual Meeting (1969).

FISSILE MATERIAL: Pu QUANTITIES PRESENT: approximately 260 g (^{239}Pu + ^{241}Pu) per fuel plate

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Accuracy - ± 1.00% relative (95% confidence level).

The mean difference between destructive chemical analysis and gamma-spectroscopy was .049 w/o Pu for 344 ternary melts.

SNM CHEMICAL & PHYSICAL FORM: metal alloy fuel plates

Dimensions		Composition	
thickness	0.2 inch	^{239}Pu + ^{241}Pu	25 w/o
width	1.9 inch	Mo	2.5 w/o
length	0.8 to 8.8 inch	U (depleted)	balance

MATRIX MATERIAL:

CONTAINER DESCRIPTION: stainless steel jacket

HOMOGENEITY: Good

SELF SHIELDING: No corrections applied since plates were assumed to be homogeneous.

BACKGROUNDS: Compton background was measured at an energy above the 414 keV peak and subtracted from the area under the peak. Corrections were made for varying backgrounds caused by varying amounts of gamma emitting fission products in plates adjacent to the one being scanned.

MEASURING PROCEDURES & THROUGHPUT:

detector - Ge(Li) 20 cm^3, ($\Delta E/_E$ = 4 KeV @ 1.33 MeV), collimated by 3 inch lead to 1 x 2.5 inch aperture.

plates arranged on scanning table - 9 rows, 6 plates each

plates are scanned on first one side, then the other

scanning table controlled by PDP8 computer, data stored in computer and converted to g ^{239}Pu output.

Throughput - 1000 plates/40 hour work week.

The ^{239}Pu content is determined from the measured intensity of the 414 keV line.

STANDARDS & CALIBRATION: Five standard plates were fabricated, one for each plate size scanned. The standards were calibrated by weighing, chemical analysis and mass spectrographic analysis. The scanning system was calibrated for gain shifts before each run by telling the computer the channel in which the center of the 414 keV peak appeared.

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS:

1. The 267 KeV line (^{237}U) was used to determine the ^{241}Pu content for a set of plates similar to the above but with an additional 25 w/o ^{241}Pu added (displacing depleted U).

2. Although agreement between the gamma scan and chemical techniques was very good in general, two instances occured in which the gamma scan detected anamolies which had gone unobserved in the destructive analysis.

OTHER APPLICABLE REFERENCES: R. B. Perry, et. al., Proceedings of the Ninth Annual Meeting of the Institute of Nuclear Materials Management, (1968) p. 205.

R. W. Branderbury, et. al., Materials Evaluation, 28 N.4 (1970).

DATA SHEET 1-07

TECHNIQUE: Gamma-Spectroscopy

REFERENCE: W. Strohm, Proceedings from Passive Gamma Seminar, Germantown, Maryland (1970)

FISSILE MATERIAL: ^{238}Pu QUANTITIES PRESENT: 0.1-5.0 g ^{238}Pu in cans
0.2-15 g ^{238}Pu in drums

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Accuracies (2σ): cans ± 18%; 55 gallon drums ± 28%

These accuracies are based on comparisons of gamma scan results with results from calorimeter measurements.

SNM CHEMICAL & PHYSICAL FORM: ^{238}Pu contamination on glove box scrap and waste.

MATRIX MATERIAL: metal turnings, plastic, lucite, metal scrap, broken glass lead lined gloves, cloth.

CONTAINER DESCRIPTION: cans - 0.5 gallon cans (13.8 cm dia. x 14.9 cm high) sealed in # 12 cans (15.7 cm dia. x 22.2 cm high).
- cardboard container (17.5 cm dia. x 17.8 cm high).

drums - 30 gallon drums contained in 55 gallon drums.

HOMOGENEITY: Poor

SHIELDING: The attenuation of the 766 KeV ^{238}Pu gamma ray versus can weight has been experimentally measured for sample matrices. Using this data, an attenuation correction is applied to unknown cans based on their measured weight. For drums, the attenuation of an external ^{238}Pu source (766 KeV line) through the unknown drum is used to correct the assay data.

BACKGROUNDS:

MEASURING PROCEDURES & THROUGHPUT:

cans
detector - 3 x 3 inch NaI
detector to can ℄ - 40 cm
count period - 4 minutes
cans are rotated during assay

drums
detector - 3 x 3 inch NaI
detector to can ℄ - 200 cm (occasionally 400 cm for high ^{238}Pu content).
count periods - 15 min. with external ^{238}Pu source (attenuation correction)
- 15 min. without external ^{238}Pu source

^{238}Pu content is determined from the corrected measured intensity of the 766 keV line.

STANDARDS & CALIBRATION: Standards were constructed using representative matrix materials and known amounts of ^{238}Pu.

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS: The gamma assay is checked by making calorimeter measurements on randomly selected samples.

OTHER APPLICABLE REFERENCES:

W. Strohm, Nucl. Appl. 5.3 (1968), p 183.

W. Strohm, et. al., Gamma-Ray Assay of ^{238}Pu in Waste Cans, MLM-1585 (1969).

DATA SHEET 1-08

TECHNIQUE: Gamma-Spectroscopy

REFERENCE: T. D. Reilly, R. B. Walton, Nuclear Safeguards R&D Progress Report LA-4605-MS (1970), p. 37.

FISSILE MATERIAL: ^{235}U **QUANTITIES PRESENT:** 2 1/2 - 5% enrichment

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Statistical Precision: 2-3% (relative) Accuracy: typically 2-3% (relative)
Two rods differed from stated enrichment values by 7-9% (relative), however uncertainties due to the cladding thickness of some rods may contribute to the errors observed.

SNM CHEMICAL & PHYSICAL FORM: uranium oxide pellets in fuel rods

MATRIX MATERIAL:

CONTAINER DESCRIPTION: fuel rod cladding material for rods 2 to 10 feet in length, 0.4 to 2.75 inch O.D.

HOMOGENEITY: Good

SELF SHIELDING: Self shielding of the 185 KeV line from ^{235}U is the basis for determining the enrichment using this method. (See Ref. below). Corrections for shielding due to different or non-uniform cladding were not taken into account in these measurements.

BACKGROUNDS: Two counts were taken for each measurement. One with the window centered on the 185 KeV line and a second count with the window set above 185 KeV to correct for the Compton background.

MEASURING PROCEDURES & THROUGHPUT:

detector - 2" x 1/2" thick NaI detector
portable electronics unit

count periods - 2 counting periods (one for a background correction) took a total of 5 minutes

^{235}U enrichment was determined by the intensity of the 158 keV gamma line

STANDARDS & CALIBRATION: Two rods of known enrichment were used to calibrate the system. The calibration range was 1.8 to 9.9%.

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS:

OTHER APPLICABLE REFERENCES: J. D. Reilly, et al., LA-4605-MS (1970), p. 19.

DATE SHEET 1-09

TECHNIQUE: Gamma-spectroscopy

REFERENCE: J. L. Parker, Nuclear Safeguards R&D Progress Report, LA-4605-MS (1970) p. 36.

FISSILE MATERIAL: ^{235}U QUANTITIES PRESENT: 1-5% enrichment

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Statistical Precision: $\leq 1\%$ Accuracy: Estimated to be ± 5%

SNM CHEMICAL & PHYSICAL FORM: Uranium oxides ($\geq 80\%$ of container contents).

MATRIX MATERIAL: incinerator residues

CONTAINER DESCRIPTION: Can 6.0 inch O. D., 15.0 inch height. Contents bagged inside cans. Cans were sufficiently full so that tumbling would cause only minor mixing.

HOMOGENEITY: Fair to poor - the difference between measurements of the enrichment for the top and bottom of the cans varied from less than 5% (relative) to 700% (relative).

SHELF SHIELDING: Self shielding of the 185 KeV line from ^{235}U is the basis for determining the enrichment using this method. (See reference below).

BACKGROUNDS:

MEASURING PROCEDURES & THROUGHPUT:

detector - Ge(Li)

detector to bottom (top) of can - 12 inches

counting period - 5 minutes

^{235}U enrichment was determined by measurement of the intensity of the 185 KeV gamma line.

STANDARDS & CALIBRATION: Two standard cans of U_3O_8 were prepared to span the calibration range of 0.8% to 10% enrichment.

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS:

OTHER APPLICABLE REFERENCES: J. D. Reilly, et. al., LA-4605-MS (1970), p. 19.

DATA SHEET 1-10

TECHNIQUE: Gamma-Spectroscopy

REFERENCE: J. L. Parker, Nuclear Safeguards R&D Progress Report, LA-4605-MS (1970), p. 37.

FISSILE MATERIAL: ^{235}U QUANTITIES PRESENT: 3-4% enrichment

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Statistical Precision: 1% (1 σ) Accuracy: ~ ± 2% (relative)
± 1% due to variations in the surface area of the containers.
< ± 2% due to imperfect averaging over the pellet mixture due to concentration of low-enrichment pellets.

SNM CHEMICAL & PHYSICAL FORM: reject UO_2 fuel pellets.

MATRIX MATERIAL:

CONTAINER DESCRIPTION: 2 gallon cardboard drums (O.D. 9.9 in.). The drums were filled with containers of 20-40 pellets per container. Each drum held ~1000 pellets (about 11 kg).

HOMOGENEITY: Fair. The entire bottom of the drum was viewed and the drums were tumbled between 3 successive measurements to obtain a good average response from the contents.

SELF SHIELDING: Self shielding of the 185 KeV line is the basis for determining the enrichment using this method. (See reference below.)

BACKGROUNDS:

MEASURING PROCEDURES & THROUGHPUT:

detector - Ge(Li)

detector to bottom of drum - ~26 inches

counting periods - 3-5 minutes counts between which the drums are tumbled.

^{235}U enrichment was determined by measurement of the intensity of the 185 KeV gamma line.

STANDARDS & CALIBRATION: Two drums were sampled and concluded to have pellets of 3.1% and 4.1% enrichment, respectively. It turned out, however, that the 4.1% standard predicted the 3.1 % standard to have an enrichment of 2.8%. Accurate standards were not assembled for these measurements.

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS:

OTHER APPLICABLE REFERENCES: J. D. Reilly, et. al., LA-4605-MS (1970), p. 19.

DATA SHEET 1-11

TECHNIQUE: Gamma-Spectroscopy

REFERENCE: F. Brown, D. R. Terrey, P. T. Good, A. E. Ross, A. Lumatte, T. Otomo, M. Sultan, E. Kerr, Proceedings - Symposium on Safeguards Techniques (1970), IAEA-SM-133/69 p. 125.

FISSILE MATERIAL: Plutonium QUANTITIES PRESENT: 18-70 g Pu

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Statistical Precision: 0.5 - 1.0%

The overall precision including the contributions from instrumental sources was approximately equivalent to the statistical precision.

SNM CHEMICAL & PHYSICAL FORM: Zero power reactor (ZEBRA) fuel plates made from plutonium metal and pins containing mixed plutonium-uranium oxides.

MATRIX MATERIAL:

CONTAINER DESCRIPTION: 13 types of plates, mostly 2 inch square, stainless steel cladding. 3 types of pins containing mixed oxides.

HOMOGENEITY: Good

SHIELDING: Since all samples were assumed to be of uniform size and composition, no shielding or self shielding corrections were made.

BACKGROUNDS: No background was subtracted from the single channel count rate.

MEASURING PROCEDURES & THROUGHPUT:

detector - Pitman model 292 portable gamma spectrometer (collimated)

detector to plate separation - ~25 cm

counting period - 10 seconds

The Pu content was determined from a measurement of the intensity of the 384 keV line.

STANDARDS & CALIBRATION: 13 plate types with ^{239}Pu contents ranging from 18 to 71 gm were used to calibrate the instrument. The entire gamma spectrum of these standards would normally be measured and checked with previous results to insure that they had not been tampered with. Calibration: ~43 cps/gm Pu (metal) and ~42 cps/gm Pu (oxides); the calibration curves were essentially linear over the sample range.

EQUIPMENT STABILITY: Standard plates and a ^{133}Ba source (prominant peak - 360 KeV) would be measured periodically to check for electronic drifts. As an example of long term drifts or inaccuracies uncurred in resetting the instrument controls, the results from a standard were observed to change by more than 5%.

SPECIAL CONSIDERATIONS: The instrument was not successful in carrying out gamma assays of irradiated fuel plates due to interference from fission product gamma rays. Measurements show that it takes a month or more for irradiated plates to return to their pre-irradiation count rate.

OTHER APPLICABLE REFERENCES: S. H. W. Tarrant, D. R. Terry, UKAEA Report COS9 (1969).

R. J. May, S. H. W. Tarrant, IAEA Personnel Dosimetry for Radiation Accidents, Vienna (1965) p. 435.

DATA SHEET 1-12

TECHNIQUE: Gamma-Spectroscopy

REFERENCE: W. H. Zimmer, The Ge(Li) IV ^{239}Pu Package Counter, ARH-1993 (1971).

FISSILE MATERIAL: Plutonium

QUANTITIES PRESENT: Trace quantities up to quantities close to criticality limits.

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Sensitivity: (max.) 70 μg ^{239}Pu per l.

Accuracy: The measured response from Pu in various positions in a 127 l. carton compared to that measured for Pu in the center of the carton varies from + 14% to -55% in the high sensitivity counting position and varies from + 7% to -2% in the low sensitivity counting position.

Statistical Precision: ± 30% at 95% confidence level (70 μg ^{239}Pu/l.)
± 5% at 95% confidence level (7 mg ^{239}Pu/l.)

SNM CHEMICAL & PHYSICAL FORM: plutonium wastes.

MATRIX MATERIAL:

CONTAINER DESCRIPTION: The apparatus is constructed to assay containers ranging from small pasteboard pill boxes to 55 gallon drums. Specific adapters are available for 10 cm dia. x 15 cm high tin cans, 127 l. waste cartons, and 55 gallon drums.

HOMOGENEITY: Fair to Good

SELF SHIELDING: An absorption correction is applied to the data by measuring the transmission of a ^{133}Bu gamma source (355.3 keV) through the sample. A point source is used for small samples and a 25 cm long line source is used for larger packages.

BACKGROUNDS: The Compton background is subtracted from under the 355.3 keV (^{133}Bu) and 413.7 keV (^{239}Pu) peaks.

MEASURING PROCEDURES & THROUGHPUT:

detector - a vertical array of 4-20cc Ge(Li) detectors (FWHM)=3.0 keV @ 355 keV) 32 cm in height (collimated). The combined surface area is >50 cm^2. All detectors are covered by a 1/16 in. lead filter.

detector to sample ℄ - ~ 35 cm (high sensitivity position) for trace amounts up to 20g Pu.
130 cm (low sensitivity position) for more than 20 g Pu.

Samples are rotated at 4 rpm and large samples can be vertically oscillated at a vertical speed of 10 cm/minute.

Counting Period - typically ~1000 seconds.

STANDARDS & CALIBRATION:

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS: Data is read automatically into a small computer which subtracts backgrounds, applies the absorption correction, calculates statistical precisions, and prints out the estimated number of g. ^{239}Pu in the sample.

OTHER APPLICABLE REFERENCES:

W. H. Zimmer, ARH Report 1877 (1970).
W. H. Zimmer, ARH drawings no. SK-2-22152, Ge(Li) IV System Components.

DATA SHEET 1-13

TECHNIQUE: Gamma-Spectroscopy

REFERENCE: R. S. Forsyth, W. H. Blackadder, The Non-Destructive Determination of Burn-up by Means of the ^{144}Pr 2.18 MeV Gamma-Activity, Report AE-187 (1965).

FISSILE MATERIAL: ^{235}U (burn-up) QUANTITIES PRESENT:

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Estimated precision ~5%. Burnup was calculated from the activity of the ^{144}Pr, the $^{95}Zr/^{95}Nb$, and ^{137}Cs lines. An ~8% bias between the ^{144}Pr and $^{95}Zr/^{95}Nb$ results was attributed to inaccuracies in the nuclear data used in the calculations or experimental problems.

SNM CHEMICAL & PHYSICAL FORM: UO_2, irradiation time of 8-23 days, cooling times of 155 to 646 days.

MATRIX MATERIAL:

CONTAINER DESCRIPTION: Zircaloy clad fuel elements from 1.26 to 1.50 cm diameter.

HOMOGENEITY: Fair - Asymetries of up to 9% were measured when viewing a sample pellet from 4 different directions.

SHIELDING: Absorption corrections due to the fuel and container were calculated and applied to correct the measured intensities of all gamma rays of interest.

BACKGROUNDS: Ambient background plus a straight line Compton backgrounds were subtracted from under the peaks of interest.

MEASURING PROCEDURES & THROUGHPUT:

detector - 1 3/4 x 2 in. NaI (collimated) (FWHM = 5.5-6.0% @ 2.2 MeV).

Elements were rotated during the scan to average out asymetries.

Counting Period - several hundred minutes

The burnup was infered from a measurement of the intensity of the ^{144}Pr 2.18 MeV gamma line.

STANDARDS & CALIBRATION: The ^{144}Pr gamma activity was measured from seven different rods and compared with their known burnup. The observed intensity for 155 - 373 days cooling time was approximately 150-2.18 MeV gamma/min.g (UO_2).

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS:

OTHER APPLICABLE REFERENCES:

DATA SHEET 1-14

TECHNIQUE: Gamma-Spectroscopy

REFERENCE: R. S. Forsyth, W. H. Blackadder, "Use of the Fission Product ^{106}Ru Gamma Activity as a Method for Estimating the Relative Number of Fission Events in ^{235}U and ^{239}Pu in Low-Enriched Fuel Elements," Proceedings - IAEA Symposium on Safeguards Techniques (1970), SM-133/4 p. 521.

FISSILE MATERIAL: ^{235}U and ^{239}Pu (burn-up)

QUANTITIES PRESENT: 1.476% enriched uranium fuel pellets

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Reasonable agreement was obtained between calculations plus destructive measurements and the gamma scan results for ^{239}Pu and ^{235}U burn-up in BWR fuel elements. Estimates of the overall precision range from about 3-20%. Several possible systematic errors could account for differences between the measurement and calculations. The calculations were assumed to be accurate to within ± 5%.

SNM CHEMICAL & PHYSICAL FORM: UO_2 pellets (1.476% enrichment) from a BWR reactor (~6600 MWd/t burn-up).

MATRIX MATERIAL:

CONTAINER DESCRIPTION: Pellets were in stringers containing 50 pellets. Pellet size 1.2 cm diameter, 1.6 cm height. The container was made of Zircalloy-2, wall thickness 0.76 mm. and was ~80 cm in length.

HOMOGENEITY: Fair to Poor. Burn-up varied by more than a factor of 2 from one end of the stringer to the other.

SHIELDING: Measured counting rates were corrected for attenuation in the fuel and containers using known dimensions and contents.

BACKGROUNDS: A straight line Compton background was subtracted from under the peaks of interest.

MEASURING PROCEDURES & THROUGHPUT:

detector - Ge(Li)~0.5 cc, with 2 mm lead filter

counting period - 200, 400 and 800 minutes per measurement, measurements were made every 5 to 15 cm along the stringers (80 cm length).

The ^{235}U + ^{239}Pu burn-up was determined from measurements of the intensity of the gamma lines from ^{95}Zr (726, 758 keV), ^{137}Cs (662 keV), and indirectly from ^{134}Cs (605, 796 keV). The ^{239}Pu burn-up was determined from the relative enhancement of the measured intensity of the ^{106}Ru gamma lines at 513 and 624 keV.

STANDARDS & CALIBRATION: Selected pellets from the stringer were analyzed for burn-up using the stable molybdenum isotopes method.(1) The ^{137}Cs (662 keV) gamma scan results were normalized to these results so that a burn-up profile was obtained along the stringer. A computer program, REBUS-2, was used to calculate the percentage of the total fission that occured in ^{239}Pu as a function of burn-up. This was then related to the ^{106}Ru activity and the results were compared with the data. Gamma scan results were also compared with burn-up calculated from the difference in two destructive measurements - stable molybdenum isotopes (^{239}Pu and ^{235}U) and uranium depletion (^{235}U only).

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS: The accuracy and precision of these measurements could be improved by using a larger detector to obtain better statistical data.

OTHER APPLICABLE REFERENCES:

(1) R. S. Forsyth, D. G. Guthrie, A. E. Ross, Proceedings-5th Conf. Analytical Chemistry in Nuclear Reactor Technology, Gatlinburg (1961). TID-7629 p. 166.

DATA SHEET 1-15

TECHNIQUE: Gamma-Spectroscopy

REFERENCE: N. C. Rasmussen, J. A. Sooka, S. A. Mayman, "The Non-destructive Measurement of Burn-up by Gamma-Ray Spectroscopy," IAEA SM-67/45 (1965).

FISSILE MATERIAL: ^{235}U (burn-up) QUANTITIES PRESENT:

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Accuracy estimated to be ~±10%

SNM CHEMICAL & PHYSICAL FORM: Uranium fuel plates.

MATRIX MATERIAL:

CONTAINER DESCRIPTION: Irradiated MITR fuel elements, 18 plates per element. Plate dimensions - 2.8 in width, 24-5/8 in length, 0.6 in thick. Cooling times ranged from 1.5 weeks to 3-1/2 years.

HOMOGENEITY: Fair - up to 50% variations were measured in fission product activity along the longitudinal axis of the fuel plate - 6 to 7% variations were measured in the transverse direction across the fuel plate. (Cooling time - 9 months)

SELF-SHIELDING: Absorption corrections were applied to the measured data using known fuel element dimensions and content.

BACKGROUNDS: An averaged Compton background was subtracted from under peaks of interest. Previous measurements have shown this to introduce a maximum uncertainty of approximately twice the counting statistics, i.e., 2 x (1-2%) ≤5% for the case presented here.

MEASURING PROCEDURES & THROUGHPUT:

Detector - Ge(Li) (collimated); ~0.5 cc (FWHM = 4.7 keV @ 662 keV)
Detector to fuel element separation - ~6 feet.
Fuel elements are under water; an air filled tube extends from the fuel element to the detector.
The ^{137}C(662 keV)/^{134}Cs(605 keV) intensity ratio and the ^{137}Cs (662 keV)/^{95}Zr(724 keV) intensity ratios were used to simulataneously determine the neutron exposure and flux level. The ^{95}Zr (724 keV)/[(^{195}Zr (758 keV) + ^{95}Nb (766 keV)] intensity ratio was used to determine the cooling time. The burnup was calculated from the neutron exposure using previously calculated results for MITR fuel elements.

STANDARDS & CALIBRATION

Burnup, cooling time, and average flux exposure were calculated from fission product gamma ray measurements and compared with recorded data, burnup calculations, and cobalt wire measurements. The above accuracy was determined from these comparisons.

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS: 4 to 6 weeks cooling time are required before a reasonably accurate determination of fission product activity can be made.

A 6-m bent crystal spectrograph was also used to measure the fission product spectra. It was awkward and slow to use, but could measure activity after only 18 hours cooling time. However, the background changed so rapidly for short cooling times that accurate gamma intensity determinations were impossible.

OTHER APPLICABLE REFERENCES:

TECHNIQUE 2 - PASSIVE NEUTRON

I. PHYSICAL PRINCIPLES OF MEASUREMENT

Nuclear materials are somewhat unique in that they emit neutrons directly in spontaneous fission reactions and indirectly through (α, n) reactions following spontaneous alpha decay. Several examples of neutron emission rates from nuclear materials (both in elemental form and in compounds are given in Table 2.1.

TABLE 2.1

NEUTRON EMISSION RATES

Isotope	Neutron Emission Rate n/gm-sec	
	Fission	(α, n)
^{238}Pu	2.6×10^3	
^{239}Pu	3×10^{-2}	
^{240}Pu	1×10^3	
^{242}Pu	1.7×10^3	
^{238}U	1.7×10^{-2}	
^{235}U	7.6×10^{-4}	
^{242}Cm	2.3×10^7	
$^{239}PuF_4$		4.3×10^3
$^{238}PuO_2$		1.4×10^4

Several general conclusions can be drawn from the limited but representative data given in Table 2.1.

(1) Some materials (e.g. uranium) emit significantly fewer neutrons than others, making them difficult to assay in small quantities by measuring their neutron yield. On the other hand, several materials (notably ^{240}Pu and ^{242}Pu) emit sufficient neutrons to make neutron yield measurements a useful technique for determining their presence and quantity in assay samples.

(2) Materials which emit few spontaneous fission neutrons (e.g. ^{239}Pu) can be the source of significant numbers of (α, n) neutrons when the material is present in chemical compounds involving light elements (fluroine, oxygen). Measurement techniques which distinguish between fission and (α, n) neutrons are required when the chemical form of the assay samples is not carefully controlled.

(3) Strong fission neutron emitters can be found in relatively uncommon heavy elements (e.g. ^{242}Cm). These neutrons can be exploited for assay purposes if the relative amount of neutron source material is known, compared to other nuclear materials of interest in the assay sample. Conversely, if these source materials constitute an unknown contaminant, their presence can cause serious errors in the interpretation of the assay data.

(4) In all cases, the relative amount of each important neutron emitter present in the sample must be known in order to infer the quantity of nuclear material present from a measurement of the neutron yield.

II. APPARATUS

The most commonly used neutron detectors for passive assay applications employ moderated BF_3 or He^3 gas proportional counters. These counters can be designed in a variety of moderator configurations, and in general, are relatively efficient over a broad band of neutron energies ranging from thermal to several MeV. This range is adequate to span the energy range of both fission and (α, n) neutron spectra.

Detector logic which is designed to distinguish between (α, n) neutrons and fission neutrons operates on the principle that fission events produce more than one neutron (typically somewhere between 2 and 3) while (α, n) events produce a single neutron. One measurement technique employs a moderated array of proportional counters surrounding the sample. For this configuration, two neutrons will be detected for some fraction of the fission events occuring in the assay sample. These neutron pulses are correlated in time; on the average they will be separated by a time roughly equivalent to the neutron lifetime in the moderator. On the other hand, some pairs of neutrons from two uncorrelated (α, n) events or an (α, n) plus a fission event will accidentally have the same time separation as a true fission event. A much simplified diagram of the detector logic for identifying time correlated fission neutrons from accidentally correlated events is shown below in Fig. 2.1 and is described below:

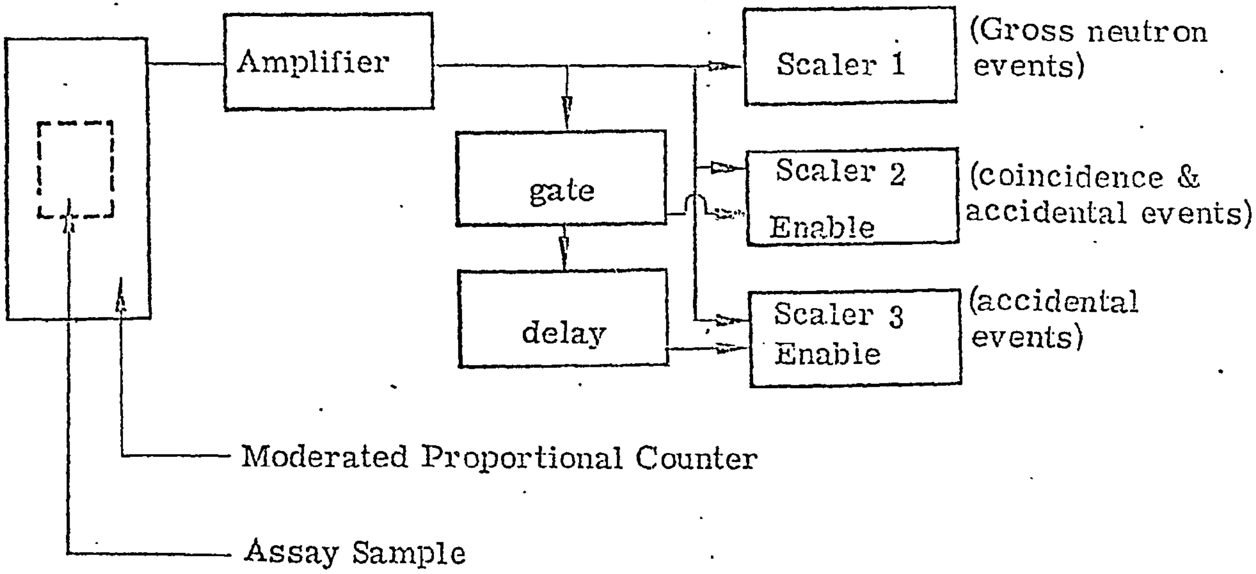

Fig. 2.1. Passive coincident neutron assay system.

Scaler 1 - records all events.

Scaler 2 - records "second" events which occur within a set time interval after a "first" event enables* scaler 2. These are time coincident fission events plus accidental (α, n) or fission events.

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[*] Enable - an electronic pulse is sent to the scaler which allows it to count (the scaler is normally "off").

Scaler 3 - records "second" events which occur in a time interval identical to that in scaler 2, but at a much later time than the lifetime of true coincident neutrons in the detector. These are all accidental events. This scaler is also enabled by the "first" event, but after a fixed time delay. The probability for accidental events to be recorded in scaler 3 is the same as that for scaler 2.

Scaler 2 minus Scaler 3 equals the number of true coincident events (when corrected for deadtime) which can subsequently be related to the amount of material undergoing spontaneous fission within the sample (assuming the isotopic content is known).

Samples with large (α, n) backgrounds and relatively small amounts of spontaneously fissioning material can require unreasonably long counting times to accumulate sufficient statistics for assay information. Considerably more detail on the design, construction, and performance of coincident neutron assay equipment is contained in references 2-1 through 2-4.

It should be noted that the gross neutron count can also be used to determine the amount of fissionable material present in the sample provided the isotopic content <u>and</u> the chemical corporation of the nuclear material is known and controlled. If this is the case, the additional logic for the coincident system is not required.

Neutrons from spontaneous fission or (α, n) events can cause neutron fission reactions in the sample of interest in addition to those from spontaneous fission. This effect, known as multiplication, can cause errors in the assay result for both gross neutron and coincident neutron counting schemes. The importance of this effect increases with the amount of material being assayed; it is also enhanced in samples containing large amounts of hydrogenous material. Neutrons are also moderated and absorbed by non-fissionable materials in the sample. Examples of good absorbers include, hydrogen, boron, lithium and cadmium. In general for passive neutron assay, moderation and absorption is more of a problem for hydrogenous and other materials of low atomic number and less a problem for dense materials of high atomic number. This is exactly the opposite of a similar absorption problem for passive gamma assays.

III. REFERENCES

2-1 C. V. Strain, "Potential and Limitations of Several Neutron Coincidence Equipments", NRL Report 2127 (1970).

2-2 C. V. Strain and R. J. Omohundro, "Coincident Neutron Equipment," NRL Report 2107 (1970).

2-3 S. H. W. Tarrant and D. R. Terry, "Development of a Portable Instrument for the Measurement of Neutrons and Gamma Rays Emitted from Nuclear Materials," AWRE Report COS 9 (1969).

2-4 R.B. Walton "Assay for Pu by Neutron Coincidence Counting", LASL Report LA-4457-MS, p. 26-34.

DATA SHEET 2-01

TECHNIQUE: Passive Neutron

REFERENCE: D. M. Bishop, E. A. Aitken, FIP Quarterly Progress Report, GEAP-12114-5 (1970).

FISSILE MATERIAL: Plutonium

QUANTITIES PRESENT: 2-13 g Pu containing 0.2 to 0.7 g ^{240}Pu equivalent*

* ^{240}Pu equivalent = the total neutron yield including contributions from ^{240}Pu, ^{238}Pu and ^{242}Pu expressed in equivalent g of ^{240}Pu.

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Precision:	±1.2% gross neutron ±0.5% coincident neutron
Accuracy:	±1.8% gross neutron ±1.1% coincident neutron

SNM CHEMICAL & PHYSICAL FORM: Plutonium-uranium mixed oxides fuels; Pu/Pu+U ranged from 1.6 to 24.7%.

MATRIX MATERIAL:

CONTAINER DESCRIPTION: 6 inch active fuel columns.

HOMOGENEITY: Good.

SHIELDING: No observable effect.

BACKGROUNDS:

no shielding	20,000 gross counts 160 real & accidental counts 150 accidental counts	per 5 minute run
Shielding consisting of 0.026 in. Cd Sheet + 2 in. borax + 4 in. polyethylene	5,000 gross counts 10 real & accidental counts 5 accidental counts	per 5 minute run

MEASURING PROCEDURES & THROUGHPUT:

detector - BNL coincident neutron detector

gate length - 2 x 64 microseconds

counting period - 10 minutes

^{240}Pu equivalent content of the samples is determined from the measured neutron activity of the samples. From this quantity, the amount of Pu, U, ^{235}U, and other Pu isotopes are calculated using data based on chemical analyses.

STANDARDS & CALIBRATION: 6 standards were fabricated and the contents determined by weighing, chemical analyses, and mass spectrographic measurements. The calibration range was 2.3 to 13.2 g Pu (0.25 to 1.44 g ^{240}Pu equivalent).

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS: A similar system was tried for scanning fuel rods with little success, although results under improved designs may make scanning feasible (ref. 2 below).

OTHER APPLICABLE REFERENCES:

1. D. M. Bishop, E. A. Aitken, GEAP-12114-3, (1970).
2. J. C. Limpert, WCAP-7562-4 (1971).

DATA SHEET 2-02

TECHNIQUE: Passive Neutron

REFERENCE: D. M. Bishop, E. A. Aitken, PIP Quarterly Progress Report, GEAP-12114-5 (1970).

FISSILE MATERIAL: Plutonium

QUANTITIES PRESENT: 1-40 g Pu (0.1-4.5 g ^{240}Pu equivalent*)

*^{240}Pu equivalent=the total neutron yield including contributions from ^{240}Pu, ^{238}Pu and ^{242}Pu expressed in equivalent g of ^{240}Pu.

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Accuracy: ±4.7% gross neutron
±3.6% coincident neutron

SNM CHEMICAL & PHYSICAL FORM: Plutonium-uranium mixed oxides in sintered pellet form. Pu/Pu+U ranged from 2.6 to 24.7%.

MATRIX MATERIAL:

CONTAINER DESCRIPTION: Standard one-gallon paint can.

HOMOGENEITY:

SHIELDING: No observable effect.

BACKGROUNDS:

no shielding	20,000 gross counts 160 real & accidental counts 150 accidental counts	per 5 minute run
Shielding consisting of 0.026 in Cd Sheet + 2 in borax + 4 in polyethylene	5,000 gross counts 10 real & accidental counts 5 accidental counts	per 5 minute run

MEASURING PROCEDURES & THROUGHPUT:

detector - BNL coincident neutron detector

gate length - 2 x 64 microsecond

counting period - 10 minutes

^{240}Pu equivalent content of the samples is determined from the measured neutron activity of the samples. From this quantity, the amount of Pu, U, ^{235}U and other Pu isotopes are calculated using data based on chemical analysis.

STANDARDS & CALIBRATION: 14 standards were fabricated and the contents determined by weighing, chemical analysis, and mass spectrographic measurements. The calibration range was 1 to 41 g Pu (0.1 to 4.8 g ^{240}Pu equivalent).

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS:

OTHER APPLICABLE REFERENCES:

D. M. Bishop, E. A. Aitken, GEAP-12114-3, (1970).

DATA SHEET 2-03

TECHNIQUE: Passive Neutron

REFERENCE: [illegible]. M. Bishop, E. A. Aitken, PIP Quarterly Progress Report, GEAP-12114-5 (1970).

FISSILE MATERIAL: Plutonium

QUANTITIES PRESENT: 10 to 250 mg Pu (1-20 mg ^{240}Pu equivalent).*

*^{240}Pu equivalent = the total neutron yield including contributions from ^{240}Pu, ^{238}Pu and ^{242}Pu expressed in equivalent g of ^{240}Pu.

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Accuracy: 47% gross neutron**
22% coincident neutron**

**Most of the error here is due to poor statistical data for small amounts of material (1 mg ^{240}Pu equivalent). Omitting these very low-level samples, the accuracies are:
10% gross neutron
2% coincident neutron

SNM CHEMICAL & PHYSICAL FORM: unsintered green powder, mixed uranium-plutonium oxides. Pu/Pu + U ranged from 1.5 to 27.4%.

MATRIX MATERIAL: shredded composite absorbent pads (hydrogenous).

CONTAINER DESCRIPTION: Standard one-gallon paint can.

HOMOGENEITY:

SHIELDING: No observable effect.

BACKGROUNDS:

no shielding	20,000 gross counts 160 real & accidental counts 150 accidental counts	per 5 minute run
Shielding consisting of 0.026 in Cd sheet + 2 in borax + 4 in polyethylene	5,000 gross counts 10 real & accidental counts 5 accidental counts	per 5 minute run

MEASURING PROCEDURES & THROUGHPUT:

detector - BNL coincident neutron detector

gate length - 2 x 64 microseconds

counting period - 10 minutes

^{240}Pu equivalent content of the samples is determined from the measured neutron activity of the samples. From this quantity, the amount of Pu, U, ^{235}U and other Pu isotopes are calculated using data based on chemical analysis.

STANDARDS & CALIBRATION: 10 standards were fabricated and the calibration determined by weighing, chemical analyses, and mass spectrographic measurements. The calibration range was 10 to 250 mg Pu (1-20 mg ^{240}Pu equivalent).

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS: Plutonium uniformly dispersed in containers has a higher specific gross count rate than more localized distributions. This is a result of the difference in detection efficiency for the system as a function of the material location within the container.

OTHER APPLICABLE REFERENCES:

D. M. Bishop, E. A. Aitken, GEAP-12114-3, (1970).

DATA SHEET 2-04

TECHNIQUE: Passive Neutron

REFERENCE: J. C. Limpert, PIP Quarterly Progress Report, WCAP-7562-4

FISSILE MATERIAL: Plutonium QUANTITIES PRESENT: 15-50 g PuO_2 (approximately 0.3% ^{238}Pu, 80% ^{239}Pu, 13% ^{240}Pu, 5% ^{241}Pu, 0.8% ^{242}Pu)

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Precision: 2.5% (1σ) for 20 g PuO_2 sample
Accuracy: ±5%

SNM CHEMICAL & PHYSICAL FORM: PuO_2 hard scrap.

MATRIX MATERIAL: Ashed waste.

CONTAINER DESCRIPTION: Cans

HOMOGENEITY: Fair to good

SHIELDING: No observable effect

BACKGROUNDS:

MEASURING PROCEDURES & THROUGHPUT:

detector - BNL coincident neutron detector

gate length - 2 x 32 microseconds

counting period - 5 minutes

Pu content is related to a measured neutron count rate for samples with known isotopic composition.

STANDARDS & CALIBRATION: Plutonium standards were made up covering the range from 10 to 100 g PuO_2 (9-87 g Pu, ~13% ^{240}Pu). The calibration was not completely linear, but showed some possible high count rate effects for samples containing more than 70 g Pu. Calibration was ~80 net counts/gm Pu·minute.

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS:

1. Six cans of assayed scrap were dissolved, the chemical assay of the dissolver solution indicated that the Pu recovered was 182.3 gm - neutron coincidence measurements of the same samples gave 183.5 gm.
2. Results indicate little problem with absorption for hard scrap - a significant improvement in accuracy over gamma-scan techniques for the same samples.

OTHER APPLICABLE REFERENCES:

DATA SHEET 2-05

TECHNIQUE: Passive Neutron

REFERENCE: H. O. Menlove, B. R. Dennis, and J. H. Menzel, Nuclear Safeguards R&D - Program Status Report, LA-4523-MS p. 30.

FISSILE MATERIAL: Plutonium QUANTITIES PRESENT: 3-500 g Pu.

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Statistical precision: $\leq 2\%$ (1σ)

Accuracy: Heterogeneous samples $\pm 8\%$ (1σ) 3-155 gm Pu
Homogeneous sample $\pm 3\%$ (1σ) 10-500 gm Pu

Bias: Heterogeneous samples +1%
Homogeneous samples +1%

SNM CHEMICAL & PHYSICAL FORM: PuO_2 scrap

MATRIX MATERIALS:

Heterogeneous: insulation, slag, glass, graphite scarfings, ash

Homogeneous: graphite scarfings and ash

CONTAINER DESCRIPTION: 1 gallon polyethylene bottles

HOMOGENEITY: The material was divided into 2 classes; homogeneous and heterogeneous (see Matrix Materials)

SHIELDING: A small ^{252}Cf source of neutrons (10^{-9} gm) is placed beneath the unknown container to correct for the perturbation due to multiplication and matrix effects of the unknown sample. This correction can amount to up to 5% for coincidence counts and up to 20% for gross counts.

BACKGROUNDS:

MEASURING PROCEDURES & THROUGHPUT:

detector - LASL portable neutron coincidence counter

counting period - 1000 seconds (400 seconds for homogeneous samples of more than 25 g Pu)

A second counting period was required while a ^{252}Cf source was placed under the sample to apply a multiplication and matrix effects correction. A dead time correction was also applied to the data.

The Pu content of the sample is determined from the measured coincident neutron count rate for material of known isotopic composition.

STANDARDS & CALIBRATION: 15 heterogeneous standards and 11 homogeneous standards were prepared by the Rocky Flats Chemistry Standards Laboratory and the system was checked for accuracy over the range 3-155 g Pu (heterogeneous) and 10-500 g Pu (homogeneous).

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS:

OTHER APPLICABLE REFERENCES:

H. O. Menlove, R. B. Walton, LA-4457-MS.

DATA SHEET 2-06

TECHNIQUE: Passive Neutron

REFERENCE: H. O. Menlove, R. B. Walton, Nuclear Safeguards R&D Status Report, LA-4457-MS (1970), p. 30.

FISSILE MATERIAL: Plutonium QUANTITIES PRESENT: 10-180 g Pu

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Statistical precision: 6-15%

Accuracy: Results agree to within 5-10% of results obtained from active neutron interrogation.

SNM CHEMICAL & PHYSICAL FORM: Plutonium oxides

MATRIX MATERIAL: Slag and crucibles containing significant amounts of fluorine and magnesium.

CONTAINER DESCRIPTION: 1 gallon can

HOMOGENEITY: Poor

SHIELDING:

BACKGROUNDS: A large (α, n) background of neutrons from the fluorine and magnesium in the slag caused a low coincidence to accidental ratio of ~0.05. This causes the statistics for the net count to be poor due to the large background subtraction. (See special considerations.)

MEASURING PROCEDURES & THROUGHPUT:

detector - LASL portable coincidence neutron counter.

The Pu content of the sample is determined from the measured coincidence neutron count rate and the given material isotopic composition.

STANDARDS & CALIBRATION: A 20 g PuO_2 sample in a can of MgO was used to calibrate the detector.

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS: A passive neutron assay of ^{238}Pu with various (α,n) background contributions showed the accuracy of the assay to be within 3% of a chemical assay for (α,n) contributions to the neutron flux of up to 60%. For (α,n) contributions of 175% and 490%, the accuracy of the passive neutron assay was 8% and 12% respectively.

OTHER APPLICABLE REFERENCES:

DATA SHEET 2-07

TECHNIQUE: Passive Neutron

REFERENCE: J. E. Foley, Nuclear Safeguards R&D Status Report, LA-4605-MS (1970), p. 34.

FISSILE MATERIAL: Uranium

QUANTITIES PRESENT: ^{238}U - 1 to 65 lbs* enriched to 1-5% in ^{235}U

*A minimum of several hundred grams is required for assays using this technique, because of low specific count rates.

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Statistical precision - ±5% (1σ) (200 sec count, ~25 lb U)
±10-15% (1σ) (1000 sec count, <2 lb U)

Accuracy - Results agree to within ~3% of chemical sampling measurements.

SNM CHEMICAL & PHYSICAL FORM: Mostly uranium incinerator oxides

MATRIX MATERIAL:

CONTAINER DESCRIPTION: Can - 6-1/4 in OD, 15-1/4 in length

HOMOGENEITY: Fair to poor

SHIELDING: A small ^{252}Cf source (~10^4n/sec) was placed under the unknown samples and the count rate measured to apply a matrix and multiplication correction to the observed data. Matrix corrections ranged from 0-25%. There is evidence that this technique does not apply a large enough correction for the heaviest cans.

BACKGROUNDS:

MEASURING PROCEDURES & THROUGHPUT:

detector - LASL portable coincidence counter

count period - 200 sec (~25 lb container)
- 1000 sec (~2 lb container)

A second count period is required with the ^{252}Cf source in place for the matrix and multiplication correction. The uranium content of the samples is determined from the measured coincidence count rate and the given material isotopic composition.

STANDARDS & CALIBRATION: A standard for calibrating the system consisted of a small can (4-1/4 OD, 5 in length) containing 2707 g UO_2 (natural enrichment). The coincidence counting rate was 0.40 counts/sec for the standard.

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS: Extreme care must be taken when inserting massive uranium samples into the detector to insure criticality safety if the enrichment is not carefully controlled.

OTHER APPLICABLE REFERENCES:

J. E. Foley, LA-4605-MS (1970), p. 18.

DATA SHEET 2-08

TECHNIQUE: Passive Neutron

REFERENCE: J. E. Lovett, D. B. James, PIP Quarterly Progress Report, BHO-69-3 (1970).

FISSILE MATERIAL: Plutonium

QUANTITIES PRESENT: 100-350 g. PuO_2
(8.5-30 g ^{240}Pu equivalent)*
1100-3500 g PuO^2-UO^2
(22.5-71 g ^{240}Pu equivalent)*

*^{240}Pu equivalent = the total neutron yield including contributions from ^{240}Pu, ^{238}Pu and ^{242}Pu expressed in equivalent g of ^{240}Pu.

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Statistical precision: 2-5% (1σ)

SNM CHEMICAL & PHYSICAL FORM: PuO_2 and 25 w/o PuO_2-UO_2 mixed oxide.

MATRIX MATERIAL:

CONTAINER DESCRIPTION: 1 quart containers - 3-1/2 in OD, 7 in length

HOMOGENEITY: Good

SHIELDING: No effects of the matrix or composition of the oxide (PuO_2 or 25 w/o PuO_2-UO_2) on the gross or coincidence count rate was observed.

BACKGROUNDS: The unpoisoned detector was linear up to about 40 g of ^{240}Pu equivalent while the poisoned detector (0.034 in Cd foil around all the BF_3 tables) was linear up to about 100 g of ^{240}Pu equivalent. These figures only correspond to this particular scrap material. The observed non-linearity is associated with excessively high count rates due to (α, n) background neutrons; non-linearities in calibration begin to appear when the corrected gross neutron count rate is 10^6 counts/minute or larger.

MEASURING PROCEDURES & THROUGHPUT:

detector - BNL neutron coincidence counter

real and accidental gate widths - 32.5, 64.5 and 129 μsec.

counting period - 1 minute

The amount of ^{240}Pu equivalent is determined from the measured coincident neutron count rate. The Pu content is calculated from known isotopic data.

STANDARDS & CALIBRATION: Seven samples containing from 8.5 to 188 gm of ^{240}Pu equivalent and four samples containing 24 to 75 gm ^{240}Pu equivalent were prepared.

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS: The response of the detector at the edges of the well is 14-20% less (relative) than at the center. The response at the top and bottom of the well differs from the vertical center by about 14% (relative).

OTHER APPLICABLE REFERENCES:

J. E. Lovett, D. P. James, BHO-69-2 (1970).

DATA SHEET 2-09

TECHNIQUE: Passive Neutron

REFERENCE: V. W. Scheider, F. Hille, M. Kiy, G. Gmelin, Symposium - Safeguards Techniques (1970), IAEA-SM-133/1, p. 181.

FISSILE MATERIAL: Plutonium QUANTITIES PRESENT: 0.1-10 g Pu (avg. ~1 g per container)

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Accuracy: 6-15%

SNM CHEMICAL & PHYSICAL FORM: plutonium oxides.

MATRIX MATERIAL: Recoverable and non-recoverable waste material.

CONTAINER DESCRIPTION: cans.

HOMOGENEITY:

SHIELDING:

BACKGROUNDS:

MEASURING PROCEDURES & THROUGHPUT:

detector - 10 BF_3 counter (50 cm length) in a paraffin block. Useful counting volume ~5 liters.

STANDARDS & CALIBRATION: 5 standards were fabricated covering the range 0.11 to 12.84 gm Pu. The chemical analysis of the standards was accurate to 0.5%.

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS: The system used here was the simplest form of neutron counting, but it has a demonstrated usefulness for certain limited and controlled applications.

OTHER APPLICABLE REFERENCES:

TECHNIQUE 3 - GAMMA ABSORPTIOMETRY

I. PHYSICAL PRINCIPLES OF MEASUREMENT

Gamma rays exhibit a characteristic exponential absorption in matter, i. e.,

$$I(x) = I_0 e^{-\mu x}$$

$I(x)$ = gamma intensity of a beam of gamma rays with initial intensity I_0 after traversing a thickness x of a given material

μ = attenuation coefficient

For gamma rays in the energy region 50 to 500 KeV, the physical processes responsible for absorption are the photoelectric effect and Compton scattering. The attenuation coefficient, which is related to the probability for the occurrence of both these processes, is strongly dependent on the gamma ray energy and the material composition. To illustrate this effect, the total gamma ray attenuation coefficients for several common materials are given below in Table 3.1. (3-1)

TABLE 3-1

Total Gamma Ray Attenuation Coefficients

	E_γ = 100 KeV	E_γ = 400 KeV
H_2O	.167	.106
Al	.435	.249
Fe	2.70	.722
Pb	60.0	2.36
U	19.8	4.84

It is evident from the above table that by virtue of its high attenuation coefficient, the uranium content of a sample containing water, aluminum, or other materials of low atomic number can be determined by comparing the relative attenuation of a gamma-ray beam through the sample with similar measurements on a set of known standards. It is also evident that materials with high atomic numbers such as lead are also good gamma-ray attenuators and can cause background problems or assay errors if their presence is not accounted for prior to the measurement. All nuclear materials have approximately the same attenuation coefficient so that material identification is not possible using this technique.

The technique is useful for determining nuclear material concentration when the contents and dimension of the sample are closely controlled. Contaminants are impossible to detect using conventional techniques, although techniques employing two sources with different gamma-ray energies can be used to discriminate between some contaminant effects.

II. APPARATUS

An absorptimetry measurement requires a source, sample holder, and gamma-ray detector. Collimation of the source and detector may be desirable depending on the application. Figure 2.1 below depicts a typical arrangement.

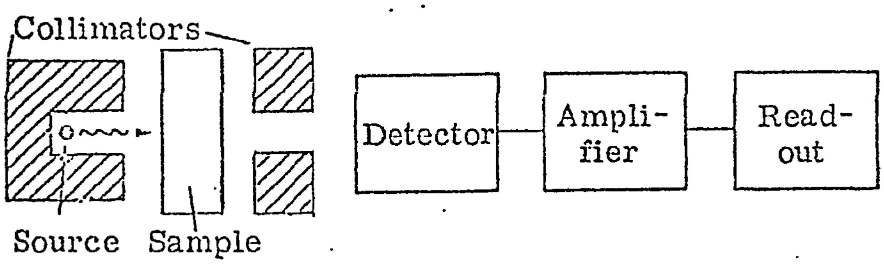

Fig. 3.1. Gamma absorptimetry.

Commonly used sources range in gamma ray energy from ^{241}Am (E_γ = 60 keV) to ^{22}Na (E_γ = 511 keV). Higher energy sources are more penetrating for thicker or denser samples, however the sensitivity between nuclear materials and other low-z matrix materials decreases with increasing gamma-ray energy. Both these factors must be evaluated in the source selection. The detector can either be energy insensitive such as a Geiger-Mueller tube or an energy sensitive device such as a scintillation detector or gas proportional counter can be used. The use of an energy sensitive detector allows one to minimize the effects due to scattered radiation within the sample and to reduce the measured ambient background. Data readout devices include scalers, count rate meters, and electrometers (both differentiating and integrating).

The simplicity of the technique and the associated apparatus makes it useful for on-line measurements and other monitoring functions although, as noted previously, the interpretation of the instrument's response can be ambiguous.

III. REFERENCES

3-1 G. W. Grodstein, NBS Circular 583 (1957) Chap. 1-3.

DATA SHEET 3-01

TECHNIQUE: Gamma-Absorption

REFERENCE: F. Brown, D. R. Terrey, J. B. Hornsby, R. G. Monk, F. Morgan, J. Herrington, P. T. Good, K. C. Steed, V. M. Sinclair, Symposium - Safeguards Techniques, Vol. II (1970), IAEA-SM-133/70, p. 125.

FISSILE MATERIAL: Uranium (no fissile discrimination) QUANTITIES PRESENT: 250 to 350 g/l U

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

SENSITIVITY - 1% changes in concentration are detectable.

SNM CHEMICAL & PHYSICAL FORM: Uranyl nitrate (3N in nitric acid)

MATRIX MATERIAL:

CONTAINER DESCRIPTION: Annular cell, absorption path length 1 cm.

HOMOGENEITY: Good

SHIELDING: The method is slightly sensitive to changes in nitric acid concentration; going from a 3N to 5N solution produced a measurable effect.

BACKGROUNDS: Exposure to a 65 gm Pu metal plate caused a small shift in response. If applied to Pu solutions, it would be sensitive to the different background radiation levels of different samples.

MEASURING PROCEDURES & THROUGHPUT:

source - 250 mCi ^{241}Am (strong line at 60 KeV) modulated by spinning it inside tungsten shield with collimation slots. Frequency of modulation - 200 cps.

detector - plastic scintillator ($\sim 4\pi$) viewed by 5 inch dia. PM tube operating in the current mode. The output signal is therefore AC.

The concentration is determined from the attenuation of the source gamma rays by the sample.

STANDARDS & CALIBRATION: The calibration was carried out over the range 250 to 350 g/l uranium in 3N nitric acid. The calibration was not linear.

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS: A special purpose absorptiometer was also devised for determining the uranium in annular uranium-aluminum alloy pieces. The response is linear over the range 0-470 mg/cm^2. A relative standard deviation of 0.8% was observed with a bias of less than 0.2%.

OTHER APPLICABLE REFERENCES:

DATA SHEET 3-02

TECHNIQUE: Gamma-Absorption

REFERENCE: S. J. Broderick, J. C. Whitmer, "Gamma-Ray Absorptiometer for Determination of Uranium in Aqueous and Organic Solvent Solution", Anal. Chem. 33, (1961), p. 1314.

FISSILE MATERIAL: Uranium (no fissile discrimination) QUANTITIES PRESENT: 1-550 g uranium/l

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Accuracy: ≤0.25% (for a series of measurements)

Precision: <1%

Sensitivity: 0.5 g U/l for the range 0-25 g U/l
<1% for the range 300-550 g U/l

SNM CHEMICAL & PHYSICAL FORM: Uranyl nitrate solution

MATRIX MATERIAL:

CONTAINER DESCRIPTION: Sample tubes were 15.2 mm and 22 mm in diameter. The larger tube was used to increase the sensitivity of the system for the larger concentrations.

HOMOGENEITY: Good

SHIELDING:

BACKGROUNDS: The following compounds are listed with their equivalence in g/l U per g/l impurity for the range 350-550 g/l U.

Na_2SO_4	0-0.15	$Pb(C_2H_3O_2)_2 \cdot 3H_2O$	2.09 - 2.13
H_3PO_4	0	$Fe(NO_3) \cdot 9H_2O$	0.29-0.33
NaCl	0.08-0.14	(additional compounds are listed in the reference).	
$CaCl_2$	0.24		

MEASURING PROCEDURES & THROUGHPUT:

source - ^{241}Am (250 mg) 60 KeV

detector - Xenon gas proportional counter (575 mm Hg)

Readings are taken instantaneously-they can be permanently recorded on a strip chart recorder.

The concentration is determined from the attenuation of the source gamma ray by sample.

STANDARDS & CALIBRATION: Standard solutions were prepared over the range 0-550 g U/l for uranyl nitrate solution. The calibration on semi-log paper was nonlinear. The uranyl nitrate salt was chemically analyzed before dissolution.

EQUIPMENT STABILITY: Standard solutions run several times a day over a period of 18 days. The system was stable to within 1%. However, there was an obvious increase in recorder readings with time.

SPECIAL CONSIDERATIONS: Influence of collimation slit width between the gamma source and sample tube are given in ref. 1 below.

OTHER APPLICABLE REFERENCES:

S. J. Broderick, Gamma Ray Absorptiometer for Determination of Uranium in Aqueous and Organic Solvent Solutions, Anal. Chem. 34, No. 2 (1962), p. 295.

DATA SHEET 3-03

TECHNIQUE: Gamma-Absorption

REFERENCE: L. C. Nelson, Gamma-Ray Absorptiometry Determines Total Uranium in Flat Fuel Elements, Nucleonics 21, No. 6 (1963), p. 88.

FISSILE MATERIAL: Uranium (no fissile discrimination) **QUANTITIES PRESENT:** Uranium plate 0.03 to 0.85 mm thick

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Precision and accuracy -	~1% (thinner foils)
(plate thickness)	~3% (thicker foils)

SNM CHEMICAL & PHYSICAL FORM: Uranium metal

MATRIX MATERIAL:

CONTAINER DESCRIPTION: Fuel plates, clad with zirconium and stainless steel; nominal cladding thickness 0.026 mm.

HOMOGENEITY: Good

SHIELDING:

BACKGROUNDS: The source energy was selected so that it was greater than the prominent ^{235}U gamma-ray at 185 KeV.

MEASURING PROCEDURES & THROUGHPUT:

source - $^{152,154}Eu$ (~1.3 μCi) $E_{\gamma} = 340$ keV.

detector - 1-3/4 x 2 in NaI (collimated).

The thickness of the uranium metal plate is determined by the measured attenuation of the source gamma rays through the fuel plate and measured thickness of the total fuel plate.

STANDARDS & CALIBRATIONS: The uranium sample thicknesses were calibrated from 0.03 to 0.85 mm, the zirconium from 0.66 to 3.2 mm, and the stainless steel from 0.66 to 2.7 mm.

EQUIPMENT STABILITY: Several measurements which fell outside the quoted range of accuracy were thought to be caused by electronic drifts of the single channel analyzer.

SPECIAL CONSIDERATIONS: A stronger source was recommended for thicker samples to improve counting statistics.

OTHER APPLICABLE REFERENCES:

DATA SHEET 3-04

TECHNIQUE: Gamma-Absorption

REFERENCE: P. J. Kreyger, R. J. S. Harry, H. Krockel, Non-destructive Burn-up Determination by 145 KeV Gamma-Ray Absorption.

FISSILE MATERIAL: Uranium (no fissile discrimination)

QUANTITIES PRESENT: ~130 g uranium (90% enriched) per element

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

1. Fabrication tolerances and fuel non-homogeneities can introduce ~5% deviation per fuel plate. A number of measurements are required for each element for better accuracy.
2. The precision of the assay varies from ~1% for zero burnup to about 4.5% for 100% burnup; this variation is due to uncertainties in the absorption coefficients.
3. The results for an HFR element agree to within 3-6% of burnup calculations and within 0.8-1.5% with results obtained from other measurements of the isotopic composition.

SNM CHEMICAL & PHYSICAL FORM:

MATRIX MATERIAL:

CONTAINER DESCRIPTION: High flux reactor (Petten, Netherlands) MTR type uranium fuel elements.

HOMOGENEITY: Fair to poor. Latheral displacements along the element had ~2% (relative) higher burnup than that measured along the longitudinal axis. The longitudinal variation was measured to vary in a regular manner by as much as 40% (relative) along the axis.

SHIELDING: The method depends on the accuracy to which an average fission product absorption coefficient which is calculated on the basis of measurements and previous data on the estimated detailed fission product content of the elements.

BACKGROUNDS: See Special Considerations.

MEASURING PROCEDURES & THROUGHPUT:

source - HFR irradiated fuel element containing the fission product gamma emitter, ^{141}Ce (145 KeV).

detector - Ge(Li) collimated, ~10 cc (FWHM = 2.5 KeV @ $E_\gamma < 1$ MeV)

Elements are scanned underwater through an aperture in a reactor storage pool wall.

Counting period - ~30 minutes per point; points were located several cm apart along a 60 cm length.

The burnup was determined by measuring the transmission of the ^{141}Cs 145 KeV gamma ray through the fuel element. Changes in absorption are related to the decrease in U and increase in fission product through burnup.

STANDARDS & CALIBRATION: A standard HFR fuel element of known composition was used to calibrate the system.

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS:

1. The method requires careful shielding from strong radiation, good detector energy resolution, and excellent homogeneity of the sensitive detector area (to minimize changes in detector response due to small spatial displacements.
2. Pulse pile-up accounted for ~1% counting losses and was corrected for.

OTHER APPLICABLE REFERENCES:

TECHNIQUE 4 - ACTIVE NEUTRON

I. PHYSICAL PRINCIPLES OF MEASUREMENT

An external source of neutrons can be used to induce fissions in nuclear material through the (n, fission) reaction. A nucleus which undergoes fission simultaneously emits approximately 7 gamma-rays and 2 to 3 neutrons; following the fission event the fission products continue to emit approximately 7 gamma-rays and .01 neutrons over periods of seconds to minutes thereafter. The fission radiation differs in several important respects from the incident neutron radiation, namely in the type of radiation, its energy spectrum, the multiplicity of emissions, and the time of emission. These differences can be exploited for purposes of assay by designing a detection system which is sensitive to induced fission radiation and insensitive to the background of incident neutron radiation. The measured response of the detector to the neutron induced fission radiation can be related to the amount of fissionable nuclear material in the irradiated sample.

There are differences in the fission radiation characteristics between isotopes, but these are relatively small and are difficult to use for practical isotopic discrimination. The distinct differences in threshold energy* for neutron fission, however, can be used to discriminate between fertile and fissile materials. (For example, ^{235}U (fissile) has a neutron fission threshold of approximately zero energy while ^{238}U

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*Neutron fission threshold energy - minimum neutron energy required to induce fission.

(fertile) has a threshold of ~1.0 MeV. Therefore, incident neutrons with energy less than 1 MeV would cause fissions in ^{235}U, but not in the ^{238}U contained in a sample.) This phenomenon places considerable importance on the selection of a neutron source with an energy spectrum suited to the particular application. The neutron spectrum of any given source can be modified, within limits, for a given application through the use of moderating and absorbing materials.

Attenuation problems for active neutron systems can be divided into two parts: (1) attenuation of the incident neutrons between the source and fissionable material, and (2) attenuation of the induced fission radiation between the fissionable material and the detector. Attenuation of the incident neutron beam is strongly dependent on the incident energy spectrum. Thermal neutrons are readily absorbed by fissile materials, and self absorption can be a serious problem for samples which higher enrichments of fissile material. Higher energy neutrons are not as readily absorbed, but the energy spectrum of neutrons is shifted toward lower energies as the neutron scatter and lose energy in the sample material (moderation). Since the probability for fission to occur is dependent on the neutron energy (especially for sub-MeV neutrons), the response from fissionable material may change as a function of the material's position in the sample. This is especially important if hydrogenous matrix materials are present in the sample as they are extremely efficient in moderating the neutron energy spectrum.

The induced fission radiation is subject to the same attenuation problems encountered with passive neutron and gamma assays. Fission neutrons can scatter and lose sufficient energy to fall below high energy neutron detector thresholds. They can also cause significant multiplication effects in samples with high concentrations of fissionable material. Prompt fission gamma-rays have energies that are as high as 7 MeV, but the majority are emitted with energies below about 1.0 MeV. They are therefore in a similar energy range as those measured in passive gamma assays and therefore are subject to similar attenuation problems.

II. APPARATUS

All active neutron systems consist of an external neutron source, sample holder, and detection system (Fig. 4.1.). Isotopic neutron sources which have been used include fission (^{252}Cf), alpha-n (Pu-Be), and photoneutron (Sb-Be). These sources can be moderated to reduce the effective energy of the neutron spectrum. Unmoderated, the fission sources emit neutrons in the energy range from 0 to 8 MeV, average energy ~1 MeV. The alpha-n sources typically emit neutrons in the range from 0 to 11 MeV, average energy ~4 to 5 MeV. The photoneutron sources have energies typically less than 1 MeV. A review of isotopic neutron sources for Safeguards applications is contained in Ref. 4-1.

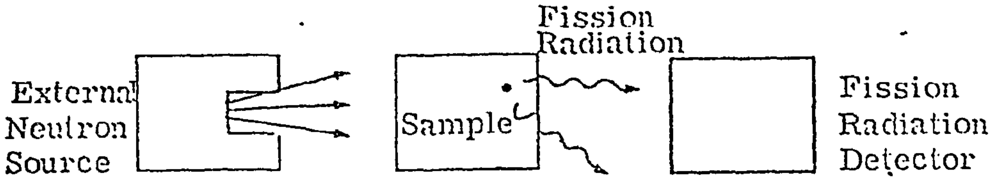

Fig. 4.1. Active neutron assay configuration.

Samples may be rotated and scanned to reduce effects due to inhomogenieties and to provide information or spacial distributions of material. Detector systems include conventional gamma ray and neutron detectors, but are especifically designed to detect radiation which is characteristic to fission while at the same time remaining insensitive to the incident neutron radiation used to induce fission. Systems which have been used to date include the following:

A. A neutron source with average energy below that of fission neutrons is used to induce fission; neutron detectors are biased to detect neutrons above the incident neutron energy and hence are predominantly sensitive to fission events.

B. Neutron induced fission gamma-rays and/or neutrons are detected with fast coincidence logic to pick out fission events. The high multiplicity associated with gamma-rays and neutron emission in fission distinguishes fission events from other processes such as inelastic neutron scattering.

C. Induced fission neutrons are detected in correlated pairs in a moderated neutron detector (similar to the passive fission neutron detector). The multiplicity of neutron emission in fission is used to distinguish fission events from the background of single events due to source neutrons.

D. Delayed fission radiation is detected by transporting the sample from the irradiation position to an isolated counting position. Shielding between the source and counting position prevents the incident radiation from interfering with detection of the delayed fission radiation. Because the delayed radiation (neutron and gamma) is emitted from seconds to minutes after fission has taken place, transport by mechanical means is a reasonable possibility for certain types of samples.

III. REFERENCES

4-1 J. R. Beyster and L. A. Kull, "Safeguards Applications for Isotopic Neutron Sources", BNL Report 50267 (T-596) (1970).

4-2 W. J. Gallagher, "Evaluation of Isotopic Source Assay System Under Commercial Operating Conditions" BHO-67-4 (1971).

4-3 R. A. Foster, H. O. Menlove, R. H. Auguston, A. E. Evans, R. B. Walton, "Radioactive Neutron Sources for Nondestructive Assay Applications," LA-4605-MS (1970) p. 8.

4-4 R. L. Beanblett, et. al., "Applications of Photoinduced Reactions to Nuclear Material Safeguards Problems," GA-10272 (1970) p. 55.

DATA SHEET 4-01

TECHNIQUE: Active Neutron

REFERENCE: G. L. Booman, J. P. Morgan, W. J. Maeck, H. R. Beard, F. L. Lisman, J. E. Rein, "ICPP Isotope Monitor", IN-1211 (1968).

FISSILE MATERIAL: ^{235}U QUANTITIES PRESENT: 0.001 to 1.0 gm/liter

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY: Measured sensitivity better than 0.001 gm/l of ^{235}U. More optimum shielding configuration indicates a factor of 10 better sensitivity is possible.

SNM CHEMICAL & PHYSICAL FORM: Liquids containing fissile material.

MATRIX MATERIAL: The absorption properties of water and H_3BO_3 solution were investigated. A 0.1 M H_3BO_3 solution reduced the delayed neutron counting rate by about a factor of 2 as compared to pure water.

CONTAINER DESCRIPTION: Thin cylindrical counting chambers surrounding the neutron irradiation source and the neutron detector.

HOMOGENEITY: Good

SELF SHIELDING: See Matrix description.

BACKGROUNDS:

MEASURING PROCEDURES & THROUGHPUT:

source - PuBe (> 3 x 10^8 n/sec) moderated by heavy water

detector - 10 BF_3 tubes moderated by heavy water

source to detector separation - 7 feet (water shielding)

source to detector liquid transit time - 2 to 5 seconds

The fissile material content is determined from the measured delayed neutron count rate.

STANDARDS & CALIBRATION: A prototype system was calibrated over the range 0.001 to 1 gm/l ^{235}U. The response was essentially linear over this range.

EQUIPMENT STABILITY: For 10% variation in flow rate, the neutron counting rate depended linearly with flow rate for transit times in the 2 to 5 second range.

SPECIAL CONSIDERATIONS: The delayed neutron background from

$$^{17}O\,(n,p)\,^{17}N \rightarrow {}^{17}O + \beta^- \rightarrow {}^{16}O + n$$

corresponds to a 1 μg/ml ^{235}U bias in the prototype apparatus.

No mention is made of backgrounds from fertile materials which may be present.

OTHER APPLICABLE REFERENCES:

DATA SHEET 4-02

TECHNIQUE: Active Neutron

REFERENCE: W. J. Gallagher, "Evaluation of Isotopic Source Assay System (ISAS) under Commercial Operating Conditions," BHO-67-4 (1971).

FISSILE MATERIAL: ^{235}U

QUANTITIES PRESENT:

Sample 1. 50-800 g ^{235}U
2. 500-3000
3. 5-900
4. 100-4250

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Precision:	0.7 to 1.7% for large quantities (several hundred g or more) of ^{235}U.
Sensitivity:	~5g ^{235}U (dry samples)
Accuracy:	1.9% for samples whose enrichment differed from standards by ~5% 4.3% for samples whose enrichment differed from standards by ~16%

SNM CHEMICAL & PHYSICAL FORM:

		Enrichment (%)
Sample 1.	U_3O_8 - Al_2O_3 heterogeneous oxide	97.5
2.	UO_2 oxide	97.6
3.	Impure U_3O_8	86.7
4.	U_3O_8 final product	92.7

MATRIX MATERIAL: Sample 1. ~3Kg Al_2O_3 Sample 3. Fe, Cr, Ni oxides
2. -- 4. --

CONTAINER DESCRIPTION: Sample 1. 5 x 6-3/4 in steel cans
2. 5-3/4 x 11 in poly bottles
3. 5 x 6-3/4 in steel cans
4. 5 x 6-3/4 in steel cans

HOMOGENEITY: Good to fair

SHIELDING: Moderation of the incident neutron flux absorption of both incident neutrons and emitted fission gamma rays and neutrons, and multiplication effects are best accounted for by using standards which closely resemble the unknown samples. Varying amounts of hydrogenous materials in the samples can cause significant differences in the system response due to moderation of the incident neutron flux.

BACKGROUNDS: The background coincident rate is not sensitive to the background sample material. 3 Kg of Al_2O_3 or 13 Kg of PbO_2 increase the measured background by ~10% as compared with no sample present.

MEASURING PROCEDURES & THROUGHPUT:

source - ~24 μg ^{252}Cf (5.6 x 10^7 n/sec)
detector - 4 plastic scintillators (ISAS fission multiplicity detector)
scanning rate - 1.75 in/minute (vertical) @ 28 rpm
throughput - 200 second normalization run with standard before and after unknown sample
8 to 10 minute scan of unknown sample and background sample
Total time for single assay - 25 minutes
For a batch of 6 similar samples, the average time per assay is about 15 min.

The uranium or ^{235}U content of the sample is determined from the measured neutron coincidence count rate and the known ^{235}U enrichment.

STANDARDS & CALIBRATION:

Sample	No. of Standards	Range of Standards (g ^{235}U)	Remarks
1	8	50- 800	approx. linear to 200 g ^{235}U
2	6	500-3000	linear
3	7	0-1000	non-linear
4	8	100-4250	non-linear

EQUIPMENT STABILITY: Data available suggest that 3 to 5% fluctuation in the response from a calibrated standard will be observed over periods of a few days to several weeks.

SPECIAL CONSIDERATIONS:

1. Abrupt changes in temperature (e.g., a 20°F temperature drop in 20 seconds) can cause the system's response to change by about 5%. Temperature changes over longer periods of time will be compensated for by periodic measurements of the standards.
2. 3 to 5% changes in count rate can be observed for the range of available polyethylene bottle thicknesses. It is advisable therefore to standardize on a particular type of container.

OTHER APPLICABLE REFERENCES:

DATA SHEET 4-03

TECHNIQUE: Active Neutron

REFERENCE: W. J. Gallagher,"Evaluation of Isotopic Source Assay System (ISAS) under Commercial Operating Conditions," BHO-67-4 (1971).

FISSILE MATERIAL: ^{235}U (low-enrichment) QUANTITIES PRESENT:
Sample 1. 5-100 g ^{235}U
2. 5-100 g ^{235}U

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY: The measured calibration points can differ from the smooth calibration curve by up to 5 or 7% for sample 1 and from to 20% for sample 2.

SNM CHEMICAL & PHYSICAL FORM: Sample 1. UO_2 oxide 3.2% enrichment
2. UO_2 pellets 2.0% enrichment

MATRIX MATERIAL:

CONTAINER DESCRIPTION: Sample 1. 5 x 6-3/4 in steel can
2. 5 x 6-3/4 in steel can

HOMOGENEITY: Good to fair

SHIELDING: Moderation of the incident neutron flux absorption of both incident neutrons and emitted fission gamma rays and neutrons, and multiplication effects are best accounted for by using standards which closely resemble the unknown samples. Varying amounts of hydrogenous materials in the samples can cause significant differences in the system response due to moderation of the incident neutron flux.

BACKGROUNDS: The background coincident rate is not sensitive to the background sample material. 3 Kg of Al_2O_3 or 13 Kg of PbO_2 increase the measured background by ~10% as compared with no sample present.

MEASURING PROCEDURES & THROUGHPUT:

source - ~24 μg ^{252}Cf (5.6 x 10^7 n/sec)
detector - 4 plastic scintillators (ISAS fission multiplicity detector)
scanning rate - 1.75 in/minute (vertical) @ 28 rpm
throughput - 200 second normalization run with standard before and after unknown sample
8 minute scan of unknown sample and background sample
Total time for single assay - 25 minutes
For a batch of 6 similar samples, the average time per assay is about 15 min.

The uranium or ^{235}U content of the sample is determined from the measured neutron coincidence count rate and the previously known ^{235}U enrichment.

STANDARDS & CALIBRATION:

Sample	No. of Standards	Range of Standards (g ^{235}U)	Remarks
1	6	5-100	slightly non-linear
2	6	5-100	non-linear

EQUIPMENT STABILITY: Data available suggest that 3 to 5% fluctuations in the response from a calibrated standard will be observed over periods of a few days to several weeks.

SPECIAL CONSIDERATIONS: 1. Abrupt changes in temperature (e.g., a 20°F temperature drop in 20 seconds) can cause the system's response to change by about 5%. Temperature changes over longer periods of time will be compensated for by periodic measurements of the standards.

2. 3 to 5% changes in count rate can be observed for the range of available polyethylene bottle thicknesses. It is advisable therefore to standardize on a particular type of container.

OTHER APPLICABLE REFERENCES:

DATA SHEET 4-04

TECHNIQUE: Active Neutron

REFERENCE: W. J. Gallagher, "Evaluation of Isotopic Source Assay System (ISAS) under Commercial Operating Conditions," BHO-67-4 (1971).

FISSILE MATERIAL: ^{235}U

QUANTITIES PRESENT:

Sample 1. 5-200 g ^{235}U
2. 5-300
3. 10-400
4. 25-100 g ^{235}U
5. 5-200
6. 5-190

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY: Precisions are limited by counting statistics for smaller amounts of uranium. Typical figures for samples of this type are 50-30% (5-10 g ^{235}U) and 8-4% (50-100 g ^{235}U).

SNM CHEMICAL & PHYSICAL FORM:

Sample		Enrichment
1.	Impure ammonium diuranate	97.0
2.	Acid insoluble residue	92.6
3.	Fluid bed reactor cleanout	97.6
4.	Uranium compounds	97.0
5.	Uranium oxides	86.7
6.	Uranium nitrate solution	93.0

MATRIX MATERIALS:

Sample 1. Fe, Cr, Ni and possibly a small amount of H_2O
2. ~3.6 Kg sand, $Al(OH)_3$, ZrO_2, filter aid
3. $NbOCl_3$
4. ~4.5 Kg of zirconia (crucible pieces)
5. Calcined ash (bomb slag)
6. —

CONTAINER DESCRIPTION:

Sample 1. 5 x 6-3/4 in steel can
2. 5-3/4 x 11 in poly bottle
3. 5 x 6-3/4 in steel can
Sample 4. 5-3/8 x 11 in poly bottle
5. 5-3/4 x 11 in poly bottle
6. 1 gallon plastic bottles

HOMOGENEITY: Fair to poor

SHIELDING: Moderation of the incident neutron flux absorption of both incident neutrons and emitted fission gamma rays and neutrons, and multiplication effects are best accounted for by using standards which closely resemble the unknown samples. Varying amounts of hydrogenous materials in the samples can cause significant differences in the system response due to moderation of the incident neutron flux.

BACKGROUNDS: The background coincident rate is not sensitive to the background sample material. 3 Kg of Al_2O_3 or 13 Kg of PbO_2 increase the measured background by ~10% as compared with no sample present.

MEASURING PROCEDURES & THROUGHPUT: source - ~24 μg ^{252}Cf (5.6×10^7 n/sec)
detector - 4 plastic scintillators (ISAS fission multiplicity detector)
scanning rate - 1.75 in/minute (vertical) @ 28 rpm
throughput - 200 second normalization run with standard before and after unknown sample
8 minute scan of unknown sample and background sample
Total time for single assay - 25 minutes
For a batch of 6 similar samples, the average time per assay is about 15 min.
The uranium or ^{235}U content of the sample is determined from the measured neutron coincidence count rate and the previously known ^{235}U enrichment.

STANDARDS & CALIBRATION:

Sample	No. of Standards	Range (g ^{235}U)	Remarks
1	6	5-200	non-linear
2	7	5-300	non-linear
3	23	10-400	linear up to ~200 g ^{235}U
4	4	25-100	linear
5	8	5-200	linear up to ~75 g ^{235}U
6	7	5-190	non-linear

EQUIPMENT STABILITY: Data available suggest that 3 to 5% fluctuation in the response from a calibrated standard will be observed over periods of a few days to several weeks.

SPECIAL CONSIDERATIONS:

1. Abrupt changes in temperature (e.g., a 20°F temperature drop in 20 seconds) can cause the system's response to change by about 5%. Temperature changes over longer periods of time will be compensated for by periodic measurements of the standards.
2. 3 to 5% changes in count rate can be observed for the range of available polyethylene bottle thicknesses. It is advisable therefore to standardize on a particular type of container.

OTHER APPLICABLE REFERENCES:

DATA SHEET 4-05

TECHNIQUE: Active Neutron

REFERENCE: H. O. Menlove, J. L. Parker, H. A. Walter, Nuclear Safeguards R&D Status Report, LA-4523-MS (1970), p. 14.

FISSILE MATERIAL: ^{235}U QUANTITIES PRESENT: 6-14 gm U (~93% ^{235}U)

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY: Results agree to within 1-2% of nominal values obtained using gamma scan.

SNM CHEMICAL & PHYSICAL FORM: Enriched uranium (93%) beads coated with graphite.

MATRIX MATERIAL:

CONTAINER DESCRIPTION: UHTREX (Ultra High Temperature Reactor Experiment) fuel rods, 6 inch length, 1 inch diameter.

HOMOGENEITY: Good

SHIELDING:

BACKGROUNDS: Four of the rods had been irradiated for 107 MWD. Dose rate at the rod surface was ~10^3 R/hour. The ^{235}U fission chamber used as a neutron detector was insensitive to this high γ-flux. The count rate for a typical sample was about 2.3 times the background count rate (due primarily to detection of ^{252}Cf source neutrons).

MEASURING PROCEDURES & THROUGHPUT:

source - moderated ^{252}Cf (1 mg)

detector - ^{238}U fission chamber

The net prompt fission neutron counting rate is used to determine the ^{235}U content of the sample.

STANDARDS & CALIBRATION: Three cold rods were measured to calibrate the prototype instrument. The calibration curve was slightly non-linear, but the count rate was about 9 counts/sec.gm U.

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS: The burnup for the hot fuel rods measured here was about 1-2%. Fully spent rods (~30% burnup) would be difficult to assay accurately due to the presence of significant amounts of fission product poisons.

OTHER APPLICABLE REFERENCES:

R. A. Forster, H. O. Menlove, LA-4523-MS (1970) p. 11.

DATA SHEET 4-06

TECHNIQUE: Active Neutron

REFERENCE: H. O. Menlove, R. A. Forster, D. B. Smith, Nuclear Safeguards R&D Report LA-4705-MS (1971), p. 6.

FISSILE MATERIAL: ^{235}U and Pu   QUANTITIES PRESENT: Sample 1. ~1 g ^{235}U/in
2. 0.5 g Pu/in

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY: Overall precision - 0.67% (1σ)
Statistical precision - 0.6% (1σ)

SNM CHEMICAL & PHYSICAL FORM:

Sample 1. UO_2 (2.34% enrichment in ^{235}U)
2. UO_2-PuO_2 mixed oxide

MATRIX MATERIAL:

CONTAINER DESCRIPTION: Sample 1. BWR fuel rod. Sample 2. Pu fuel rod.

HOMOGENEITY: Good

SHIELDING:

BACKGROUNDS: No increase in background was observed when Pu or BWR fuel rods were inserted in the detector with the source removed.

MEASURING PROCEDURES & THROUGHPUT:

source - 100 μg ^{252}Cf (moderated)

detector - He^4 gas recoil (LASL Reactor Fuel Rod Assay System)

scan rate - ~5 ft/min which implies ~30 seconds/rod. 6 rods can be scanned together at any one time.

The net prompt fission neutron counting rate is used to determine the fissile material content of the sample.

STANDARDS & CALIBRATION: The following counting rates were measured:
~1200 cps (BWR fuel rod)
~1400 cps (Pu fuel rod)

EQUIPMENT STABILITY: A constant background from high energy ^{252}Cf source neutrons which reach the detectors are utilized in a feedback loop to automatically stabilize the system response.

SPECIAL CONSIDERATIONS: 1. A NaI detector has been set up on the same scanning apparatus to measure delayed gamma rays from the neutron activated rods. Attempts will be made to further develop this technique to provide pellet-to-pellet scan information.

2. A neutron detector for delayed neutrons is being tested on the same scanning apparatus. Further investigation is in progress to determine if the prompt/delayed ratio from the two detectors can be used to discriminate between ^{239}Pu and ^{235}U.

3. Data is processed and read out on a programmed calculator.

OTHER APPLICABLE REFERENCES:

J. E. Foley, M. M. Thorpe, LA-4705-MS (1971) p. 9.

DATA SHEET 4-07

TECHNIQUE: Active Neutron

REFERENCE: J. E. Foley, M. M. Thorpe, Nuclear Safeguards R&D Report LA-4705-MS (1971), p. 9.

FISSILE MATERIAL: ^{235}U QUANTITIES PRESENT: 125 - 200 g ^{235}U

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Accuracy - 4 to 10%*

*The system used in these measurements was not completely developed, but was assembled to demonstrate the feasibility of the technique.

SNM CHEMICAL & PHYSICAL FORM: U_3O_8 (10% enrichment)

MATRIX MATERIAL: One sample contained incinerator ash.

CONTAINER DESCRIPTION: 2 l. polyethylene bottles

HOMOGENEITY: Good to fair

SHIELDING:

BACKGROUNDS: The present measurements were troubled by the coincident neutron background produced by the ^{240}Pu content in the PuLi neutron source. This problem will be eliminated in future development through the use of a PoLi neutron source.

SCANNING PROCEDURES & THROUGHPUT:

neutron source - $^{239}PuLi$ 600 n/sec (PoLi to be used in the future, $\sim 10^3$ n/sec)

detector - neutron coincidence counter (estimated required efficiency - 40%, dieaway time and gate width, 25 μsec)

counting period - ~1 hour for present measurements (estimated ~400 sec for future systems)

The ^{235}U content of the sample is determined from the measured coincident prompt fission neutron counting rate. The coincident neutrons are produced in fission events induced by an exterior neutron source.

STANDARDS & CALIBRATION:

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS:

OTHER APPLICABLE REFERENCES:

H. O. Menlove, R. A. Forster, D. B. Smith, LA-4705-MS (1971) p. 6.

DATA SHEET 4-08

TECHNIQUE: Active Neutron

REFERENCE: W. W. Beyer, J. N. Lewis, G. L. Stukenbroeker, The Determination of ^{235}U by Neutron Activation Using a Ra-Be Source, TID-7568 (Part 2), (1958), p. 158.

FISSILE MATERIAL: ^{235}U

QUANTITIES PRESENT: 0.5 g uranium oxide
0.7-1.3 w/o ^{235}U

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Precision: ~0.6% (relative) at the 95% confidence level.

SNM CHEMICAL & PHYSICAL FORM: U_3O_8 powder

MATRIX MATERIAL:

CONTAINER DESCRIPTION: 1-3/4 in diameter plastic vials

HOMOGENEITY: Good

SELF SHIELDING: As the sample size is increased, the slope of the count rate vs. sample weight curve begins to decrease due to self absorption effects for U_3O_8 samples larger than 4 g. and for uranium metal samples with thicknesses ≥5 mm. Shielding effects due to neutron absorbers such as Cd, B, Sm, and Gd decrease the calibrated counting rate by more than ~5% for concentrations ranging from 0.05 to 1.0%.

BACKGROUNDS: Plastic vials were found to have the lowest background, compared to similar containers of pyrex, brass, aluminum and boron-free glass. Backgrounds due to Cd and Eu gamma rays following irradiation were not measured separately, but are included in the measured shielding effect noted above.

MEASURING PROCEDURES & THROUGHPUT:

source - 2.5 g Ra-Be moderated (~4.5 x 10^5 thermal n/sec)

detector - NaI well counter

counting sequence - measure natural background of sample (3 min)
- irradiate sample (5 min)
- cool sample (1/2 min)
- count sample (3 min)

One technician can process ~16 assays (4 samples per assay) per day. The ^{235}U content of the sample is determined from the integrated measured fission product gamma activity of the sample after irradiation by thermal neutrons.

STANDARDS & CALIBRATION: Five samples of known ^{235}U content were used to calibrate the system over the range 0.7 to 1.3 w/o ^{235}U. The calibration is linear with slope ~2.3 x 10^4 cpm/w/o ^{235}U.

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS: Accuracy of the timing for irradiation, cooling, and count cycles significantly affects the precision of the results. An automatic timing setup for activation improved the measured precision for a series of assays by a factor of two.

OTHER APPLICABLE REFERENCES:

TECHNIQUE 5 - X-RAY FLUORESCENCE

I. PHYSICAL PRINCIPLES OF MEASUREMENT

X-rays of sufficient energy incident on an assay sample can interact with and excite the atoms of the material by freeing bound electrons from their atomic orbits. The atom reacts by allowing electrons from higher energy orbits to fill the lower vacancy created by the x-ray interaction; the excess energy is carried off with the emission of an x-ray. This x-ray has a well defined energy characteristic of the atom it was emitted from. For example, if uranium is irradiated with a "white spectrum" of x-rays with maximum energy greater than 116 KeV, a characteristic series of x-rays (K series) with well defined energies ranging from approximately 94 to 114 KeV will be emitted from the sample; a second characteristic series (L series) is also emitted with energies ranging from approximately 13 to 20 KeV.

Assay samples can therefore be irradiated with an external x-ray source and x-ray detectors can be set up to detect characteristic emissions from the stimulated sample. Under favorable conditions, the characteristic x-ray lines can be used to determine both the identity and concentration of an element, however the technique is not sensitive enough to determine isotopic differences in the emission spectra.

The technique of x-ray fluorescence is only useful in Safeguards assay work for samples with uniform composition. This is due to the fact that the fluorescent x-rays for fissionable material have energies less than 121 and 22 KeV respectively for the K and L series of lines. These x-rays are readily absorbed in intervening material so that only the surface area is sampled using this technique.

II. APPARATUS

X-ray fluorescence apparatus consists of a source, sample, and detector configuration (Fig. 5.1). X-ray sources include radioisotopes at the low end of the intensity scale and x-ray tubes at the upper end. Radioisotope sources are maintainence free, simple to use, and completely stable in operation; however, the x-ray tube offers considerably more intensity, more flexibility for optimizing the fluorescent excitation from a given element, and it can be turned off when necessary.

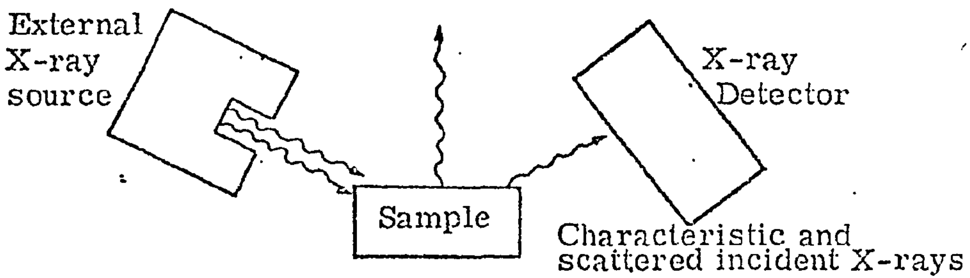

Fig. 5.1. X-ray fluorescent assay configuration.

Detector systems are divided into two general classes: dispersive and non-dispersive. Dispersive systems employ a crystal spectrometer which is made up of an analyzing crystal, collimators, and an x-ray detector. The principle of Bragg diffraction is used to analyze the sample's emission spectra; the system is only capable of measuring a single energy-point on the x-ray spectrum at any one time. Searches for contaminants and signal to background measurements can require considerable lengths of time. The energy resolution of typical systems is extremely good, but the signal intensity loss is large. Strong sources are therefore required to adequately excite small concentrations of material.

Non-dispersive systems use solid state, scintillation, or gas proportional counters to measure the entire or selected portions of the emission spectrum in a single measurement. Thus, contaminants can be located and identified and the background determined, in addition to measuring the X-ray line of interest all at the same time. The energy resolution of these detectors is less than that which is obtained with dispersive systems, however the signal intensity is much larger so that smaller X-ray sources can be used to excite the sample. The decision concerning the design of a system for use with a particular assay sample depends entirely on the composition of the sample and conditions under which the measurement will be made.

Literature which describes the technique of X-ray fluorescence in more detail are given in references 5-1 through 5-3.

III. REFERENCES

5-1 G. H. Morrison, (Editor), Trace Analysis-Physical Methods Interscience Publishers, New York (1965) p. 271.

5-2 J. C. Russ, (Editor), Energy Dispersion X-ray Analysis: X-ray Probe and Electron Probe Analysis, American Soc. for Testing and Materials, Special Technical Publication 485 (1971).

5-3 F. S. Goulding, J. M. Jaklevic, "Trace Element Analysis by X-ray Fluorescence", UCRL Report 20625, TID-4500.

DATA SHEET 5-01

TECHNIQUE: X-ray Fluorescence

REFERENCE: Nucleonics Data Systems, Huntington Beach, Calif., Test Report, 26 March 1971.

FISSILE MATERIAL: Uranium (no fissile discrimination) QUANTITIES PRESENT: Approximately 1% solution of uranium

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Statistical precision of about 0.3% relative for 1% solution

SNM CHEMICAL & PHYSICAL FORM: UO_2 and U_3O_8 dissolved in HNO_3 and diluted with H_2O to ~1 w/o.

MATRIX MATERIAL:

CONTAINER DESCRIPTION: No container between detector and solution.

HOMOGENEITY: Good

SHIELDING: Saturation thick samples are used (~1 inch) of liquid to minimize the effects of self shielding.

BACKGROUNDS: A pure water sample (1500 cps) corresponds to a 0.55 w/o uranium solution.

MEASURING PROCEDURES & THROUGHPUT:

source - 30 mCi ^{241}Am (prominent gamma line at 60 keV)

detector - Kr/CO_2 proportional counter (both are incorporated in the NDS Model 100 Portable XRF Analyzer)

counting period - 100 seconds

The uranium concentration is determined from the measured X-ray intensity in the 10-20 KeV region which includes the L_α (13.6 KeV) and L_β (17.2 KeV) fluorescent X-rays from uranium.

STANDARDS & CALIBRATION: Standard solutions were prepared in the range of 0.84 to 0.88% uranium. The calibration is approximately 2680 net counts per second/1% concentration of U.

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS:

OTHER APPLICABLE REFERENCES:

DATA SHEET 5-02

TECHNIQUE: X-Ray Fluorescence

REFERENCE: Nucleonics Data Systems, Huntington Beach, Calif. Test Report, 26 March 1971.

FISSILE MATERIAL: Uranium (no fissile discrimination)

QUANTITIES PRESENT: Uranium oxide powdered mixtures, 0 to 88 w/o uranium

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Statistical precision: approximately 0.3% (relative) over the range of concentration 3 to 88 w/o uranium.

SNM CHEMICAL & PHYSICAL FORM: UO_2, UO_3, U_3O_8 powders

MATRIX MATERIAL: graphite and binders for some samples, K_2O_3 for others.

CONTAINER DESCRIPTION: No container used.

HOMOGENEITY: Good

SELF SHIELDING: The samples were of saturation thickness (>1/8 inch) so that self attenuation effects are minimal.

BACKGROUNDS: A pure K_2CO_3 sample corresponds to about an 8% concentration of uranium.

MEASURING PROCEDURES & THROUGHPUT:

source - 30 mCi ^{241}Am (prominent gamma line at 60 keV)

detector - Kr/CO_2 proportional counter (both are incorporated in the NDS Model 100 Portable XRF Analyzer)

counting period - ~30 seconds

The uranium concentration is determined from the measured X-ray intensity in the 10-20 KeV region which includes the L_α (13.6 KeV) and L_β (17.2 KeV) fluorescent X-rays from uranium.

STANDARDS & CALIBRATION: Seven standards were prepared to cover the range of 0-88 w/o concentration of uranium. The calibration curve is exponential over this range, however a linear approximation can be made for the range of 60-90% concentration.

EQUIPMENT STABILITY:

SPECIAL CONDITIONS: Readout for the device can be obtained directly in terms of w/o concentration.

OTHER APPLICABLE REFERENCES:

DATA SHEET 5-03

TECHNIQUE: X-Ray Fluorescence

REFERENCE: A. von Baeckmann, Determination of U and Pu in Solutions of Spent Nuclear Fuels by X-ray Fluorescence Analysis, IAEA-SM-133 81, p. 45.

FISSILE MATERIAL: Plutonium and uranium (does not discriminate between uranium isotopes)

QUANTITIES PRESENT: U 0.05 - 1.2 mg g solution
Pu 0.05 - 1.2 mg g solution
U:Pu ratios of ≥300:1 have been assayed.

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

Uranium: Precision - 0.45% (1σ)
Accuracy - 0.6-0.7% (1σ) with systematic deviation of -0.09 to +0.25%

Plutonium: Accuracy - 2.1% (1σ) with systematic deviation of -0.39%*

*If 3 samples are omitted, the accuracy is 0.95% (1σ) with a systematic deviation of -0. 2

SNM CHEMICAL & PHYSICAL FORM: Uranium and plutonium in dissolver solution with activities of 400-700 Ci/l.

MATRIX MATERIAL:

CONTAINER DESCRIPTION: 3-5 ml. samples

HOMOGENEITY: Good

SHIELDING:

BACKGROUNDS: Separate calibrations were made for each type of sample analyzed. High concentrations of heavy elements besides U and Pu contribute a background which is dependent on the composition of each sample. Background was measured at points adjacent to the peak of interest. (For example, the Pu L_α line was at 35.436° and the background was taken as the mean of two measurements at 34.936° and 35.936°.)

MEASURING PROCEDURES & THROUGHPUT:

source/detector - Siemens SRS-1 X-ray Spectrometer

typical operating parameter - 55 KV, 32 ma, LiF 110 analyzer crystal and a 0.15° Soller slit

counting period - ~1 minute per peak for concentration of ~1 mg/ml
~4-5 minutes per peak for concentration of 100 μg/ml

The U and Pu concentrations are determined from the intensity of the $L\alpha$, fluorescent X-rays c U and Pu as compared to the $L\alpha$, line from an internal standard, Th.

STANDARDS & CALIBRATION: An internal standard, thorium is used to calibrate the device. Separate calibrations were made for all sample types due to the different backgrounds from high concentration of other heavy elements in the sample.

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS: The sample changer was modified and a glovebox was coupled to the spectrometer for safe handling of the radioactive samples. Additional lead was also positioned to shield the detector from hard X-rays emitted by fission products.

OTHER APPLICABLE REFERENCES:

P. A. Pella, A. Von Bueckmann, Analytica Chim. Acta 47 (1969) p. 431.

DATA SHEET 5-04

TECHNIQUE: X-Ray Fluorescence

REFERENCE: W. C. Stoecker, "Continuous Stream Analysis of Uranium Solutions by X-ray Fluorescence," Mallinckrodt Chemical Works Report MCW-1477 (1963).

FISSILE MATERIAL: Uranium (no fissile discrimination) QUANTITIES PRESENT: 0.1 to 1.8 g/l

PRECISION, ACCURACY, REPRODUCIBILITY, SENSITIVITY:

SNM CHEMICAL & PHYSICAL FORM: Uranyl nitrate aqueous solution and uranyl nitrate in raffinate slurry.

MATRIX MATERIAL:

CONTAINER DESCRIPTION: Plexiglass irradiation/counting cell

HOMOGENEITY: Good to fair

SHIELDING: Fe is commonly found in raffinate in varying amounts. It acts as an absorber for U L_α X-rays. In off-line measurements, an internal standard Sr is added to measure the effect of the Fe absorption. For on-line methods, the U peak/background could be used to compensate for absorbing matrix materials. Tests show a decrease of ~20% in the U/background ratio correlated with a 30% decrease in the U signal as the Fe concentration is raised from 0 to 75 g/1.

BACKGROUNDS: The peak to background ratio was ~2.0. Maximal variations of ~10% were measured for changes in X-ray tube current of 24 to 48 ma and in X-ray voltage of 30 to 56 KV. For changes in background with changes in absorbing material, see Shielding above.

MEASURING PROCEDURES & THROUGHPUT:

source/detector - Phillips Electronics X-ray Spectrograph #52260 (tungsten target, FA-60 X-ray tube, LiF crystal, scintillation counter detector)

typical operating parameter - 45 KV, 38 ma

response time - several seconds after the system concentration was changed were required for the strip recorder to indicate a change. About 20-25 seconds were then required for the recorder to reach a stable value.

The concentration of U is determined from a measurement of the intensity of the U-L_α line stimulated by an X-ray source.

STANDARDS & CALIBRATION: Eighteen separate uranyl nitrate aqueous solutions were used to calibrate the system between 0.1 and 1.8 g U/l. A similar set was made up for the raffinate slurry samples. The calibration curves were linear. The calibration for both samples was ~3500 cps in the U-L_α peak.

EQUIPMENT STABILITY:

SPECIAL CONSIDERATIONS:

1. Maintaining a constant separation between X-ray tube and sample window is a critical for reproducible measurements.
2. At a flow rate of 600 ml/min there was no tendency for the slurry to settle in the system.

OTHER APPLICABLE REFERENCES: