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Evaluation of In-Line Nondestructive Assay Systems

July 1—September 30, 1979

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EVALUATION OF IN-LINE NONDESTRUCTIVE ASSAY SYSTEMS

by

D. F. Bowersox, N. Ensslin, S. T. Hsue, S. S. Johnson, L. G. Speir, C. A. Spier, J. W. Tape, G. Walton, and R. B. Walton

ABSTRACT

Specified instruments for in-line non-destructive assay of process streams in spent nuclear fuels will be tested under realistic conditions for use in effective process control and materials counting. Progress in this project from July 1 - September 30, 1979 is described.

I. INTRODUCTION

This report summarizes the progress in our project to design, construct, and operate nondestructive assay systems for characterizing solids and liquids containing plutonium. Considerable effort has been spent on design and construction, and the selected systems appear to be feasible. Successful characterization would mean that automated real-time analysis of nuclear materials could be conducted in-line and at-line in processing facilities with available instruments.

The two systems to be evaluated are for solids and solutions characterizations. These are housed in two glove boxes in the LASL Plutonium Facility. Isotopic analyses, weighing, neutron counting and optional heat treatment are planned for the solids system. Isotopic analysis, rate of flow, neutron counting, X-ray fluorescence, and absorption densitometry are designed into the liquids system. Results will be calculated and reported by a minicomputer. These results will show whether present nondestructive instruments can be combined into systems

to characterize plutonium-containing solids and solutions in process-type operations. The measurements could provide rapid, timely information for process control and materials accounting, thus materially enhancing SNM safeguards.

This study is not part of the DYNAMIC Materials Controls demonstration program (DYMAC) at the Los Alamos Scientific Laboratory (LASL)¹. DYMAC is following the movement of material in and out of given areas by real-time nuclear materials control. In that program a control and accounting system is being installed for the entire LASL Plutonium Facility. We are concerned with the accuracy, precision, and reliability of data from selected, specifically designed instruments under certain programmed conditions, not with an accounting system for the entire facility.

In this project we will develop and test concepts with readily available nuclear materials. Our data will not be needed for balancing material accounts, and out-of-line results will not cause process upsets. By operating separately, we can provide in-depth, trial-and-error evaluation of instruments. For example, by testing the accuracy of a horizontal, high level neutron coincidence counter without, as well as with end plates, we can assess the potential for in-line neutron counting on a process line or conveyor belt. If the geometries cause inaccuracies, no errors will be caused in the overall facility accounting system. Since material in our glove boxes will not be part of a chemical operation, such as the FFTF oxide production line, we can repeat our analyses under a number of conditions.

Our preliminary measurements have been limited to at-line neutron counting and weighings of wet plutonium (III) oxalate cake and calcined plutonium dioxide in a plutonium dioxide production line.² The results indicate that in-line measurements are a promising approach for effective process control and accounting. Most of the instruments that will be evaluated have been described in earlier reports in this series.³⁻⁶ The descriptions include the properties to be measured and the precision and accuracy that we hope to attain in our applications. This present paper is limited to the progress made in this project for the quarter starting July 1 and ending September 30, 1979.

II. DESCRIPTION OF INSTRUMENT SYSTEMS

The layout of the experimental system is diagrammed in Fig. 1. The computer and its accessories are in the northeast corner of the laboratory. There is space for operating the computer system and controlling the experimental

Fig. 1. Schematic of Evaluation Area

program from the Decwriter and Multichannel Analyzer keyboard in this area. The glove boxes are against the east wall. The introductory box, GB-209 has open doors and a strong flow of air inward from the room. This box can be used for preparatory work and as a staging area prior to introducing material through an interlock into the enclosed glove boxes. The shift register for the neutron counter, the read-out section of the electronic balance, the power supply and the programmer for the furnace are located beneath the box. Also (although not shown in the drawing), the manual controls for the neutron counter are located here.

The adjacent glove box, GB-247, is reserved for solids studies. There are a balance and a furnace in the box. A Lexan window is directly above the detectors in the gamma station. The counting chamber in the neutron counter, which is beneath the box, is an integral part of the glove box system. The HEPA filter system passed the ventilation tests and the entire box has been thoroughly leak-tested. It is ready to be placed into radioactive, contaminated service.

The drop box, DB-215, is connected to the glove box system for the entire Plutonium Facility by a series of sealed conveyors. The drop box is reserved for transfers of materials; no experiments are permitted in it.

Plans for GB-285, the solutions glove box, are not final, and changes are still being considered. At present, we plan to pump radioactive solutions through a series of loops. Flow meters and two 10-l storage tanks are located

in the glove box. A neutron well counter, an X-ray fluorescence chamber, and a gamma densitometer (of the SRP-type) will be mounted on loops above the box. A Tokai-Mura type gamma solution densitometer and an active source neutron assay device will be placed beneath the box. The neutron assay instrument will be in-line; initially, the densitometer will be at-line. Previous commitments and priorities will delay design and installation of the active neutron assay device, similar to the shuffler.⁷

South of GB-285 are counters, with drawers for storage, and a small sink. Approximately seven feet west of our glove boxes is a train of boxes for recovery of ^{242}Pu . A waste management room is located directly east of the glove box line, separated by the concrete room wall, and our small office is located in a room behind the counter and sink.

The solids glove box is shown in more detail in Fig. 2. This equipment is ready for the initial in-line evaluation with reference materials.

Fig.2. Solids Glove Box

Samples; for example, a tray of mixed oxide; are lowered by elevator from the floor of the glove box into the neutron counter (shown in Fig. 3). The elevator gear then disengages and a chain-driven conveyor moves the tray to the mid-point (or any other selected position) in the counter. The instrument can be controlled either manually or by computer. The poly shielding, neutron detector and the high voltage neutron detectors are located outside of the glove box shell. Fig. 4 is a side view of the instrument tunnel and shielding. The shielding should significantly reduce background from the waste management room (to the east) and the ^{242}Pu line (to the west). Neutron counting with a ^{252}Cf source indicated that this instrument is ready for use.

Fig. 3. Neutron Counter

Fig. 4. Neutron Counter Side View

Details of the gamma detector station are shown in Fig. 5. The Lexan window in the glove box floor permits positioning of trays or samples directly above the detector. The gamma spectra will be transferred to a Canberra Series 80 Multi-channel Analyzer and into a DEC RT-11 computer network for analysis. The spectra may also be stored on disks for further study and calculations. The electronics in the system operated satisfactorily and the isotopic abundances program has been tested with fourteen 2g reference samples of Pu from NBS. The reference values are being corrected for radioactive decay; the calculated values are all within 1% of the uncorrected abundances.

Fig. 5. Gamma Detector Station.
(Glove Box)

Tables 1, 2, and 3 show the data from the gamma analysis of sample 12W58. The Cd peak, used for background corrections, provides a "base-line" for effective analysis. The twelve plutonium peaks are listed in increasing energy, and the areas represent count in each peak region. In Table 2, relative efficiencies, calculated from the spectral data, are given. These will be used by instrument designers; at present they provide a small correction in computing isotopic ratios

The ratio of each isotope to the ^{241}Pu , the fraction (without ^{242}Pu) and the percent abundance (with ^{242}Pu) are shown on the first lines of Table 3. The ^{242}Pu content is calculated by Isotopic Correlation Theory. The expected percent error, mainly due to counting statistics, is given on the second line. For

Table 1
Peak Area Data for Gamma Spectra
Sample: 12W58

LIVE TIME:	23.	CLOCK TIME:	0.	12W58.DAT			
CD PEAK	1 ENERGY=	88.0	AREA=	5986.1	SIGMA=	984.	FRACT SIGMA= 16.43 %
PU PEAK	2 ENERGY=	125.3	AREA=	99888.3	SIGMA=	878.	FRACT SIGMA= 0.88 %
PU PEAK	3 ENERGY=	129.3	AREA=	767702.6	SIGMA=	1167.	FRACT SIGMA= 0.15 %
PU PEAK	4 ENERGY=	148.6	AREA=	214451.0	SIGMA=	768.	FRACT SIGMA= 0.36 %
PU PEAK	5 ENERGY=	152.7	AREA=	5125.3	SIGMA=	463.	FRACT SIGMA= 9.03 %
PU PEAK	6 ENERGY=	160.0	AREA=	29369.8	SIGMA=	521.	FRACT SIGMA= 1.77 %
PU PEAK	7 ENERGY=	161.5	AREA=	18977.7	SIGMA=	482.	FRACT SIGMA= 2.54 %
PU PEAK	8 ENERGY=	164.6	AREA=	57212.7	SIGMA=	644.	FRACT SIGMA= 1.13 %
PU PEAK	9 ENERGY=	332.3	AREA=	44379.3	SIGMA=	299.	FRACT SIGMA= 0.67 %
PU PEAK	10 ENERGY=	335.4	AREA=	12525.0	SIGMA=	223.	FRACT SIGMA= 1.78 %
PU PEAK	11 ENERGY=	345.0	AREA=	29811.5	SIGMA=	235.	FRACT SIGMA= 0.79 %
PU PEAK	12 ENERGY=	375.0	AREA=	70558.8	SIGMA=	321.	FRACT SIGMA= 0.45 %
PU PEAK	13 ENERGY=	413.7	AREA=	55294.7	SIGMA=	258.	FRACT SIGMA= 0.47 %

Table 2
Relative Efficiencies in Gamma Spectra
Sample: 12W58

ENERGY	RELATIVE EFFICIENCY	12W58
125.3	0.32202E+01	
129.3	0.33044E+01	
148.6	0.37111E+01	
152.7	0.37775E+01	
160.0	0.38964E+01	
161.5	0.39164E+01	
164.6	0.39658E+01	
332.3	0.15313E+01	
335.4	0.15081E+01	
345.0	0.14357E+01	
375.0	0.12097E+01	
413.7	0.10000E+01	

Table 3
Calculated Isotopic Abundances from Gamma Spectra
Sample: 12W58

	ISOTOPE/241	FRACTION	PERCENT	AM-241
PU-238	0.27753E-01	0.12615E-03	0.12605E-01	0.00000E+00
	0.90550E-01	0.91995E=01	0.91795E-01	0.00000E+00
PU-239	0.19995E+03	0.90888E+00	0.90814E+02	0.32703E-02
	0.11499E-01	0.33542E-02	0.33542E-02	0.10452E+00
PU-240	0.19018E+02	0.86445E-01	0.86373E+01	0.00000E+00
	0.36860E-01	0.35258E-01	0.35258E+01	0.00000E+00
PU-241	0.10000E+01	0.45455E-02	0.45417E+00	0.65020E+00
	0.00000E+00	0.10926E-01	0.10926E-01	0.00000E+00
PU-242	0.00000E+00	0.00000E+00	0.32326E-01	0.00000E+00
	0.00000E+00	0.00000E+00	0.32330E-01	0.00000E+00
TOTAL				0.29615E-02
				0.10458E+00

example, the ratio Pu-239/Pu-241 is 199.95, the fraction Pu-239 is .9088, and the atomic percent is 90.81. The expected counting error in the atomic fraction is 0.0034%. The Am-241 fraction is calculated; in this particular sample the fraction was quite small.

The detector and multi-channel analyzer have been used to collect data on reference samples of Cd and Co. The preliminary experiments indicate that the system can be used to characterize plutonium-containing materials. We will move the system into the Plutonium Facility, analyze larger reference samples and prepare to evaluate these instruments. Eventually a second Series 80 Multichannel Analyzer and detector will be placed into the solutions area and tied into this computer.

A line drawing of the solutions glove box is shown in Fig. 6. The 10-l Pyrex tanks and the 0.6 cm stainless steel tubing and valves for loops within the glove box have been installed. Load cells have been replaced with a pressure transducer system such as the one shown in Fig. 7 for determining liquid level in the tanks. Several flow meters have been ordered. A small electrical centrifugal pump was tested, but was unsatisfactory for circulating water through the loops.

Fig. 6. Solutions Glove-Box

Fig. 7. Pressure Transducer System

It will be replaced with a variable flow pump, 0-5 gpm.

The volumes of the 15.2 cm dia. solution tanks have been determined with deionized water by determining liquid levels by a visual marker technique and by a pressure transducer method. In the visual marker system, stainless steel rulers (marked into 0.025 cm divisions) were mounted on the side of each tank. The lower meniscus of the water level was read; the water level was then lowered approximately 5.08 cm by withdrawing water into a beaker. After five minutes, the water level in the column was reread and the withdrawn water weighed. This calibration procedure was repeated three times to obtain the data given in Table 4. The results show that the column diameter is not uniform; that is, there are small variations in volume between marks. Repeatability in reading was excellent, and this method would be satisfactory for volume measurements in this system.

The pressure difference between the glove box atmosphere and that of a column of liquid in the tank is proportional to the height of that column. In our system difference was measured by passing compressed air simultaneously into the box and into a dip tube to the bottom of the column (tank). The difference is measured with a pressure transducer, and the resulting signal displayed (in mv) on a nearby instrument. Since our tanks are only 56 cm high, the overall pressure changes are small (3.5 mv) for an entire tank of liquid. Therefore, relatively small changes in the readouts or fluctuations in pressure, will cause relatively large errors in calculation of the level position. Bubble rates were about 60 bubbles per minute for Tank 1 (for easy visual level reading) and 125 bubbles per minute for Tank 2 (for more stable pressure readings). For volume determinations the tank level was adjusted to 1.0 on the marker and the pressure differential read. The water was withdrawn in approximately 2.0-mark increments (equivalent to two inches on the ruler). After five minutes, the withdrawn water was weighed and the pressure recorded. The experiment was complete at mark 21. This procedure was repeated three times. Pressure readings varied up to 5% over a few seconds, probably because of changes in differential pressure during bubble formation. If a computer were used, as we plan to later, an average pressure value will probably be more representative than the "single" values used in this test. In larger, nearly full tanks, the pressure fluctuations would be much smaller percentage of the total pressure differential. The data are summarized in Table 5. The results from Tank 1, where the bubbler rate was very low, were quite variable. However, the variation for the entire column was about 1.6%. Results in Tank 2 were still variable, but the size of the fluctuations decreased.

Table 4
Calibration of Columns by a Visual Marker Method

Marker Readings ¹	Volume, ml	
	Tank 1	Tank 2
1-3	937.2 $\pm$ 5.0	921.1 $\pm$ 0.6
3-5	897.8 $\pm$ 5.5	926.7 $\pm$ 4.2
5-7	941.1 $\pm$ 5.7	920.3 $\pm$ 5.2
7-9	926.9 $\pm$ 9.7	925.3 $\pm$ 10.5
9-11	939.6 $\pm$ 4.1	940.2 $\pm$ 4.5
11-13	932.3 $\pm$ 5.6	883.9 $\pm$ 3.8
13-15	948.8 $\pm$ 7.2	924.7 $\pm$ 5.2
15-17	937.2 $\pm$ 14.8	920.7 $\pm$ 8.0
17-19	939.0 $\pm$ 3.1	929.0 $\pm$ 16.2
19-21	949.1 $\pm$ 4.6	920.3 $\pm$ 6.1
Total	9351 $\pm$ 3.7	9218 $\pm$ 13.6

^aOne marker unit is 2.54 cm

Table 5
Calibration of Columns by a Pressure Differential Method

Marker Reading ^a	Volume/Pressure Change (ml/mv)	
	Tank 1	Tank 2
1-3	3570 $\pm$ 350	3350 $\pm$ 8.6
3-5	3130 $\pm$ 530	3370 $\pm$ 22
5-7	3750 $\pm$ 190	3370 $\pm$ 6.6
5-9	3620 $\pm$ 400	3420 $\pm$ 33
9-11	3290 $\pm$ 770	3420 $\pm$ 20
11-13	3360 $\pm$ 320	3480 $\pm$ 24
13-15	3940 $\pm$ 760	3460 $\pm$ 19
15-17	3430 $\pm$ 330	3450 $\pm$ 30
17-19	3690 $\pm$ 95	3480 $\pm$ 16
19-21	3590 $\pm$ 160	3340 $\pm$ 50
Total	($\pm$ 1.61%)	($\pm$ 0.14%)

Although the visual marker technique was far more precise in the calibration runs, a pressure transducer method, featuring automatic data collection, should be far more effective in any reprocessing operation. Additional methods for determining volumes and levels, such as flow meters and sonar (ultrasonic), should also be examined.

III. TRAVEL TALKS, AND PUBLICATIONS

D. F. Bowersox, N. Ensslin, S. T. Hsue, J. W. Tape and R. B. Walton attended the twentieth annual meeting of the Institute of Nuclear Materials Management July 16-18, 1979, in Albuquerque, N.M. D. F. Bowersox attended the Design Integration Study Design Review Meeting for the Design Integration for Coprocessing and Revised Separated U-Pu Facilities at the Savannah River Laboratory on September 18, 1979. D. F. Bowersox and R. B. Walton discussed this project with G. Hammond, W. Ballard, J. McVey and V. Schneider of DOE in Germantown, Md., on September 20, 1979.

IV. FUTURE WORK

We plan to initially evaluate the instruments in the solids area with sealed reference materials containing plutonium. Design and priority planning for the solutions system will continue. The search for additional funding will also be continued.

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