

*Evaluation of the Integrated Holdup
Measurement System with the M³CA for
Assay of Uranium and Plutonium Holdup*

Los Alamos
NATIONAL LABORATORY

*Los Alamos National Laboratory is operated by the University of California
for the United States Department of Energy under contract W-7405-ENG-36.*

*This work was supported by the US Department of Energy,
Office of Safeguards and Security, through the Nonproliferation
and National Security Institute.*

*Edited by Jeff Skiby, Group CIC-1.
Prepared by Celina M. Ortiz, Group NIS-5.*

An Affirmative Action/Equal Opportunity Employer

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither The Regents of the University of California, the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by The Regents of the University of California, the United States Government, or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of The Regents of the University of California, the United States Government, or any agency thereof. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.

*Evaluation of the Integrated Holdup
Measurement System with the M³CA for
Assay of Uranium and Plutonium Holdup*

*P. A. Russo
J. K. Sprinke, Jr.
C. W. Bjork
T. O. McKown
G. A. Sheppard
S. E. Smith*
J. F. Harris**

**Lockheed-Martin Energy Systems, Oak Ridge Y-12 Plant, Oak Ridge, TN 37830.*

CONTENTS

ABSTRACT	1
I. INTRODUCTION.....	2
II. EQUIPMENT AND PROCEDURES.....	2
A. Uranium	3
B. Plutonium	6
III. RESULTS	9
A. Uranium	9
B. Plutonium	15
IV. DISCUSSION	18
A. Conclusions.....	18
B. Follow-on Work.....	19
REFERENCES.....	19

EVALUATION OF THE INTEGRATED HOLDUP MEASUREMENT SYSTEM WITH THE M³CA FOR ASSAY OF URANIUM AND PLUTONIUM HOLDUP

by

P. A. Russo, J. K. Sprinkle, Jr., C. W. Bjork, T. O. McKown, G. A. Sheppard,
S. E. Smith, and J. F. Harris

ABSTRACT

Uranium and plutonium holdup that has been simulated by insertion of a variety of sealed, reference samples into pipes, ducts, and other hardware has been measured over a period of six years with an integrated holdup measurement system.^{1,2} The result is a systematic evaluation of the generalized-geometry holdup (GGH) formalism applied to portable gamma-ray holdup measurements with low-resolution detectors. The extended exercise was carried out both with and without automation of the measurements, data reduction/analysis, and holdup evaluation. Automation was accomplished by the software Version 2 for the Holdup Measurement System (HMS2).³ The purpose of the exercise was to establish reliable benchmarks for GGH measurements and to document the advantages of the automation with actual measurement results. The results presented below demonstrate a factor of 2 improvement in the quantitative reliability of the holdup assay automated by HMS2. The automated results are otherwise identical to the manual measurements. These and similar exercises also show that automation can decrease by a factor of 20 or more the time required to execute a holdup measurement campaign and obtain the holdup quantities for the facility using an integrated holdup measurement system, and that only one person, rather than two, is required to perform the measurements. Enhanced implementation of the integrated holdup measurement system with new software, corrections for systematic effects, and improved room-temperature gamma-ray detectors is planned.

I. INTRODUCTION

The testing of the integrated holdup measurement system^{1,2} has been completed. The automated system was developed to address quantitative holdup measurement needs that are ongoing (bimonthly, quarterly, semiannually, etc.) and require a large number (hundreds to many thousands) of individual measurements. It uses the tools of portable gamma-ray spectroscopy and can be implemented with any type of portable gamma-ray spectroscopy detector. It stores process equipment information in a database that is also used to manage the holdup assay data, archive the results in time, and track the spatial and time distribution of special nuclear material (SNM) inventory as holdup. It analyzes the spectroscopic data according to the generalized-geometry holdup (GGH) formalism³ and reads out results for SNM mass at specific measurement locations, in individual pieces of equipment, or for extended process areas or buildings.

In order to establish reliable benchmark data for GGH measurements and to document the advantages of the automation, holdup data for uranium and plutonium have been obtained over extended time periods with both the portable automated system and by conventional portable holdup measurements performed manually under circumstances that are otherwise the same. This report documents the methods and results of the extended study.

II. EQUIPMENT AND PROCEDURES

Manual holdup measurements were performed with the compact NaI detector⁴ and the Davidson Corporation Portable Multichannel Analyzer (PMCA). The systematic evaluation of the integrated holdup measurement system was carried out with the same compact NaI and both the Los Alamos Miniature Modular Multichannel Analyzer (M³CA) and the PMCA. The integrated holdup measurement system uses the Holdup Measurement System (HMS) software, HMS2³, a DOS program written in Microsoft FoxPro® for DOS v 2.6. The HMS2 software automates the data acquisition; the reduction and analysis of gamma-ray spectra; and the evaluation, logging, and tracking of holdup quantities. The software was developed to “manage” the immense scope of routine, plant-wide holdup measurements of uranium. It was modified after the earlier tests to address the general routine holdup measurement needs, including those for plutonium. The HMS2-automated measurements also use a palm-size, programmable bar-code reader for operator interface with the hardware; automated setup and control of the portable gamma-ray spectroscopy system (M³CA or PMCA); and automated logging of the bar code and reduced measurement data associated with each measurement location.

Figure 1 illustrates the equipment for the integrated holdup measurement system (including M³CA) in an application of automated portable measurements of holdup by a single user. The procedure for the automated measurements requires entry of the bar code on the label attached to the surface of the process equipment at the measurement location. This first step is performed using the built-in laser scanner or pencil wand associated with the data-logging unit (in user’s left hand in Fig. 1a and 1b). In response to the scan, an audible signal (beep) is issued by the data logger, indicating recognition of the coded measurement location. The user aims the detector at the measurement spot and listens for a second beep that indicates the start of the count. At the end of the count, there is a final beep indicating completion. Because measurement control and reduction and storage of data are automated, the user steps to the next measurement location at the sound of the final beep and immediately scans the new bar code to initiate the next

count within seconds. The combination of bar-coded measurement locations; reliable and compact equipment; and automation of the numerous control, reduction, and storage functions required for the spectroscopic assay optimizes the use of time. This is because the largest portion of operation time with the portable system is spent counting. Automation also makes large numbers of very short counts (for improved sampling) or reduced count times (to accommodate demands for timeliness) feasible.



(b)

(a)

Fig. 1(a) and (b). Photographs of a user performing holdup measurements on (a) a large-diameter duct and (b) a narrow pipe with the integrated holdup measurement system hardware. In both (a) and (b), the M³CA is in a leather pouch hanging from the shoulder of the user, the compact NaI detector is held in the user's left hand and the portable controller/data logger with bar-code reader [different models are shown in (a) and (b)] is in (strapped to the wrist of) the user's right hand. In (a), the operator is scanning the bar code on the label attached to the large duct. In (b), the operator is performing a count with the collimated detector aimed at the holdup deposit marked by the bar code on the narrow vertical pipe.

A. Uranium

Simulated uranium holdup was created for nine equipment setups. The six that are illustrated in Fig. 2 were used in the evaluation of the integrated holdup system applied to uranium assay. Details of the equipment dimensions, uranium reference materials and their holdup reference values, and the alternative generalized geometries applicable to the GGH assay of the simulated deposits are given in Table I.

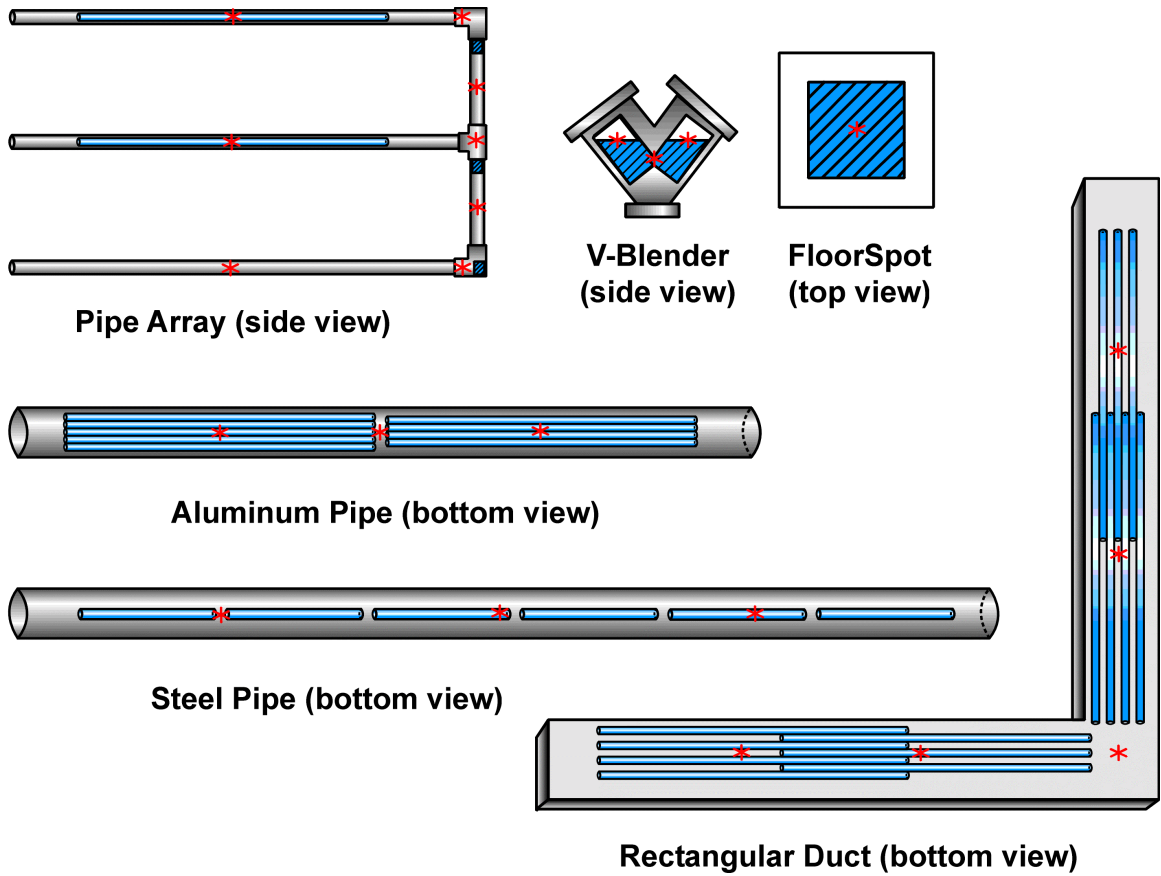


Fig. 2. The six pieces of simulated process equipment are shown approximately to scale with the uranium reference materials positioned within. The asterisks are the locations of the bar codes attached to the external surfaces of the equipment that are scanned prior to each HMS2-automated holdup measurement, and that indicate the measurement position to the user. Table I gives the dimensions of each piece of equipment as well as details of the reference materials.

TABLE I. Equipment and SNM for Uranium Holdup Measurements.

Process Equipment	Equipment Dimensions* and Geometry	Description of SNM Loading	SNM Reference Value (g ²³⁵U)	Alternative Generalized Geometries
Pipe Array, Steel	diameter = 2.8 cm thickness = 0.3-0.6 cm length = 540 cm 3 parallel, confluent lines 2 right angles, 1 T	2 LEU fuel rods, 3 vials HEU oxide	17.63	point, line, area
V-Blender, Lucite (C₅H₈O₂)	diameter = 18 cm thickness = 0.7 cm length = 55 cm symmetric V cylinder	2 bottles LEU oxide in graphite matrix	9.76	point, area
Pipe, Aluminum	diameter = 14 cm thickness = 0.3 cm length = 240 cm straight cylinder	9 LEU fuel rods	16.83	line
Pipe, Steel	diameter = 11 cm thickness = 0.3 cm length = 320 cm straight cylinder	6 long sheets of HEU metal	45.44	line
Floor Spot, Lucite (C₅H₈O₂)	length = 45 cm width = 45 cm thickness = 0.4 cm thin square laminate	1 square sheet of HEU metal	38.47	area, point
Rectangular Duct, Steel	width = 25 cm height = 8 cm thickness = 0.1 cm length = 400 cm 1 right angle bend	14 LEU fuel rods	26.17	line, area

* The inner dimensions of equipment cavities are quoted.

LEU is low-enriched uranium.

HEU is highly enriched uranium.

The loading of the uranium reference materials into the simulated process equipment for the holdup measurement tests with the GGH procedures was done once yearly for three consecutive years. Although the same reference materials were inserted each year, the precise locations within each piece of equipment changed. This is in contrast to the bar-coded measurement locations for the HMS2-automated holdup measurements, which were not moved from one year to the next.

The measurement procedures for the holdup measurement tests applied to uranium evolved from those used in the Los Alamos Department of Energy (DOE)-sponsored seminar on measurements of holdup. Two people (and often only one for the HMS2-automated measurements, which can be carried out in all phases by only one person) comprised each measurement team. Approximately 12 teams in a given year performed the holdup measurements in the manual mode (using the PMCA with manual equipment setup, data acquisition, measurement control, selection of measurement geometry, record keeping, data analysis, and computation of holdup in the extended equipment). These activities involved approximately 1.5 full working days (12 hours), excluding the time required to calibrate, and rarely did a measurement team complete all of the measurement exercises in this time period. Following the manual exercises, each measurement team (and in some cases, individual team members alone because the automated system is a one-user system) performed the same holdup measurements automated by the HMS2 software (using the M³CA primarily, with automated equipment setup, data acquisition, measurement control, selection of measurement geometry, record keeping, data analysis, and computation of holdup in the extended equipment). Both the number of measurements and the count times (~60 s for the manual measurements and 30 s for the automated measurements) were comparable for the manual and automated measurements in the uranium exercises. The complete automated exercise required a total of approximately 20 min for setup, measurements, analysis and computation with printed reports of measurement log information, and equipment holdup results. The time spent in the manual (with at least two users required) and automated (with only one user needed) modes differs by more than a factor of 20.

B. Plutonium

Simulated plutonium holdup was created for seven equipment setups. The six that are illustrated in Fig. 3 were used in the evaluation of the integrated holdup system applied to plutonium assay. Details of the equipment dimensions, uranium reference materials and their holdup reference values, and the alternative generalized geometries applicable to the GGH assay of the simulated deposits are given in Table II.

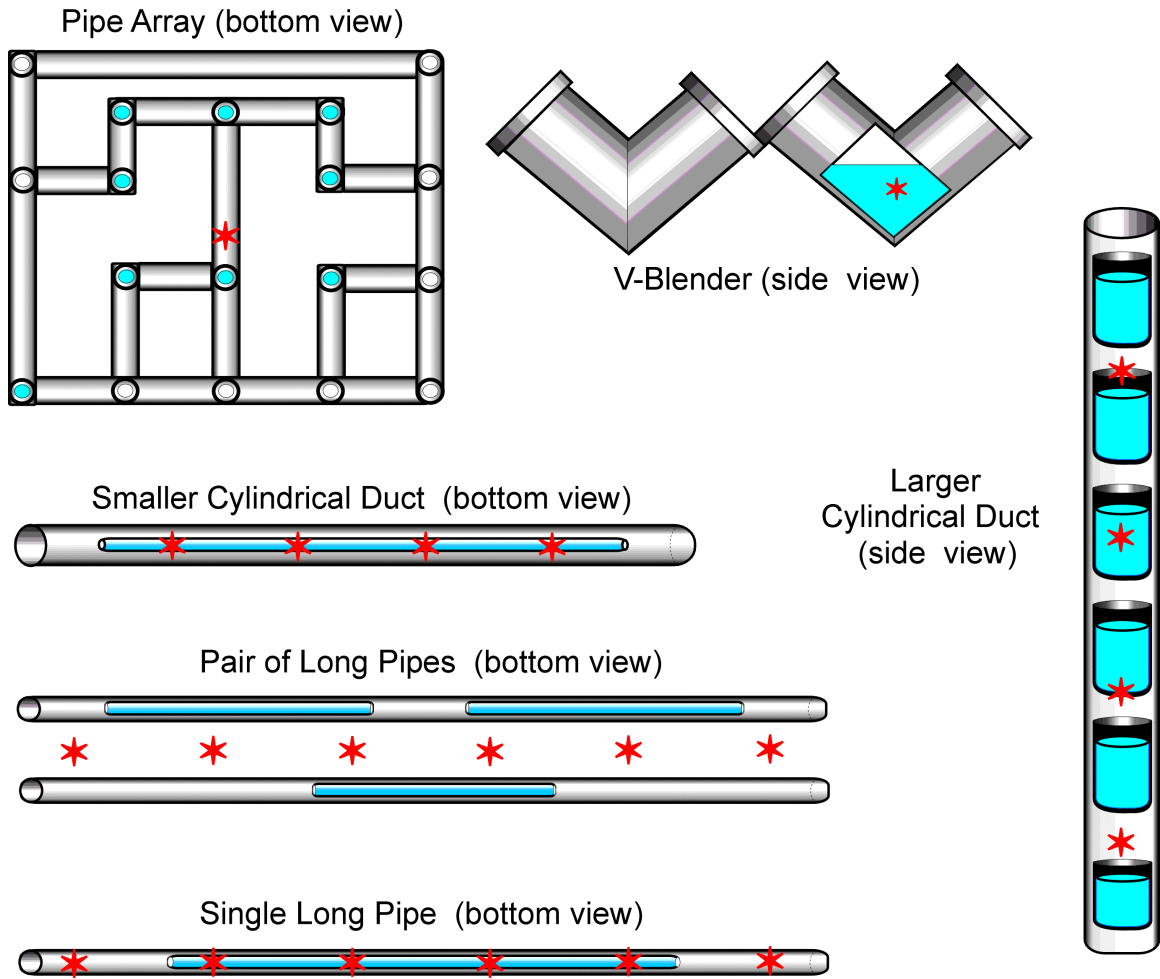


Fig. 3. The six pieces of simulated process equipment are shown approximately to scale with the plutonium reference materials positioned within. The asterisks are the locations of the bar codes attached to the external surfaces of the equipment that is scanned prior to each HMS2-automated holdup measurement, and that indicate the measurement position to the user. Table II gives the dimensions of each piece of equipment as well as details of the reference materials.

TABLE II. Equipment and SNM for Plutonium Holdup Measurements.

Process Equipment	Equipment Dimensions* and Geometry	Description of SNM Loading	SNM Reference Value (g ²³⁹Pu)	Alternative Generalized Geometries
Pipe Array, PVC (C₂H₃Cl)	diameter = 5.1 cm thickness = 0.5 cm length = 140 cm width = 110 cm one rectangular array 10 right angles, 8 Ts	9 LBU samples (plutonium oxide and metal) disk geometries	70	point, line, area
V-Blender Lucite (C₅H₈O₂)	diameter = 10 cm thickness = 0.5 cm length = 110 cm double-V cylinder	1 can LBU ash	17	point, area
Larger Cylindrical Duct, Aluminum	diameter = 16 cm thickness = 0.6 cm length = 244 cm straight cylinder	6 cans LBU oxide (dilute) in matrix of diatomaceous earth	177	line, point
Smaller Cylindrical Duct, Aluminum	diameter = 10 cm thickness = 0.6 cm length = 244 cm straight cylinder	6 LBU nitride fuel pins	65	line
Pair of Long Pipes, Aluminum	diameter = 3 cm thickness = 0.3 cm length = 2 x 270 cm straight cylinder	3 MBU carbide fuel pins	25	line
Single Long Pipe, Aluminum	diameter = 3 cm thickness = 0.3 cm length = 270 cm straight cylinder	2 MBU carbide fuel pins	17	line

* The inner dimensions of equipment cavities are quoted.

LBU is low-burnup plutonium (approximately 93% ²³⁹Pu).

MBU is medium-burnup plutonium (78% ²³⁹Pu in this case).

The loading of the plutonium reference materials into the simulated process equipment for the holdup measurement tests with the GGH procedures performed in both the manual and automated modes was done once. A later version of the HMS2 software was used that addressed the needs of routine assay of holdup (including plutonium) more generally.

The measurement procedures for the plutonium holdup measurement tests evolved from those used in the Los Alamos DOE-sponsored seminar on measurements of holdup, as described above for the uranium measurements. The manual measurements used count times of approximately 60 s. A need for more frequent measurements of room background spectra in the plutonium measurements arises from the higher gamma-ray energy region (~400 keV) used for the ^{239}Pu assay compared with that for the ^{235}U assay, which is based on the 186-keV gamma ray. Therefore, the count time for the automated assays was reduced to 10 s (compared to 30 s, which was used in the uranium tests) in order to complete the automated exercises promptly (within the allotted 20-min time period). A decrease in the time required to complete the total holdup measurement task by reducing the count times is not practical in the manual mode because the gain is not significant. This is because the duty factor for counting with 60-s counts is less than 10% in the manual mode. (More than 90% of the time spent in the manual measurements is allotted to other than counting.) With the adjusted count time for the automated measurements, the total time spent on the manual measurements (including equipment setup, data acquisition, measurement control, selection of measurement geometry, record keeping, data analysis, and computation of holdup in the extended equipment) with two or more users exceeded that spent on the same HMS2-automated activities carried out by one user by more than a factor of 20.

III. RESULTS

A. Uranium

Figures 4 and 5 are graphs for three years of results of uranium holdup measurements obtained by each of 12 groups of users of the portable spectroscopy equipment per year. These were primarily users who were inexperienced in the operation of spectroscopy equipment and in measurements of holdup. Plots of the individual assay results for both the manual (Fig. 4) and automated (Fig. 5) measurements show reasonably good agreement, on average, with the reference values. This is also shown in Tables III–V by values consistent with unity for the ratios of the average assay ($^{235}\text{U}_{\text{mass}}$) result to the reference value for each piece of equipment, and the average (of averages) for all equipment.

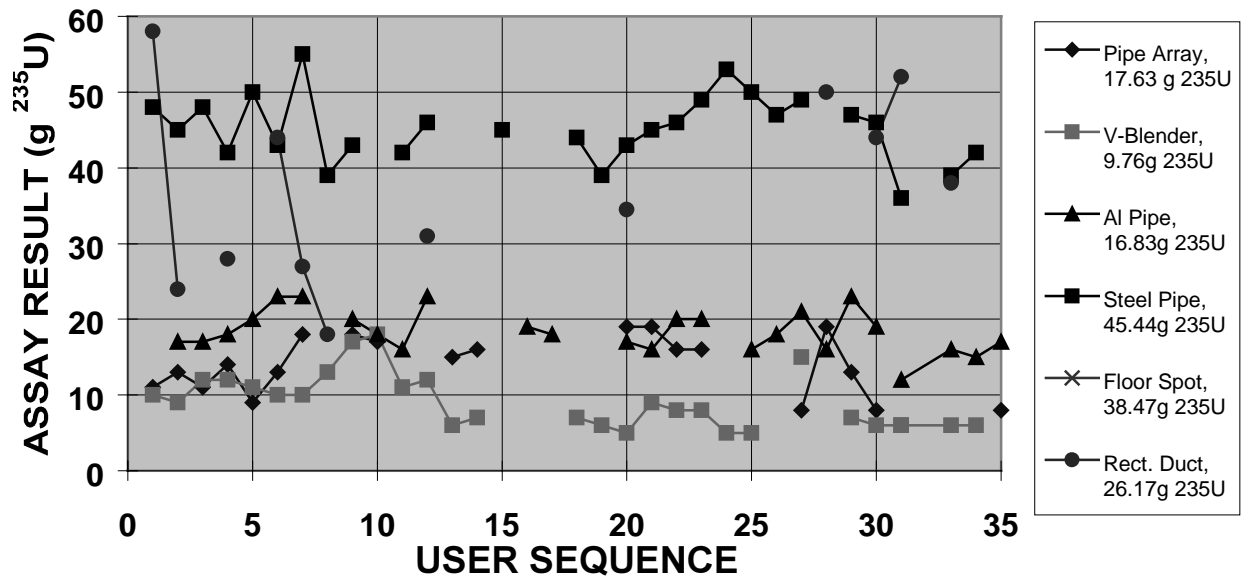


Fig. 4. Portable uranium holdup measurements (no automation). The GGH assay result for ^{235}U mass obtained by manual holdup measurements for each of the six holdup simulations by each measurement team involved in the measurements during the three-year test period is plotted in chronological sequence.

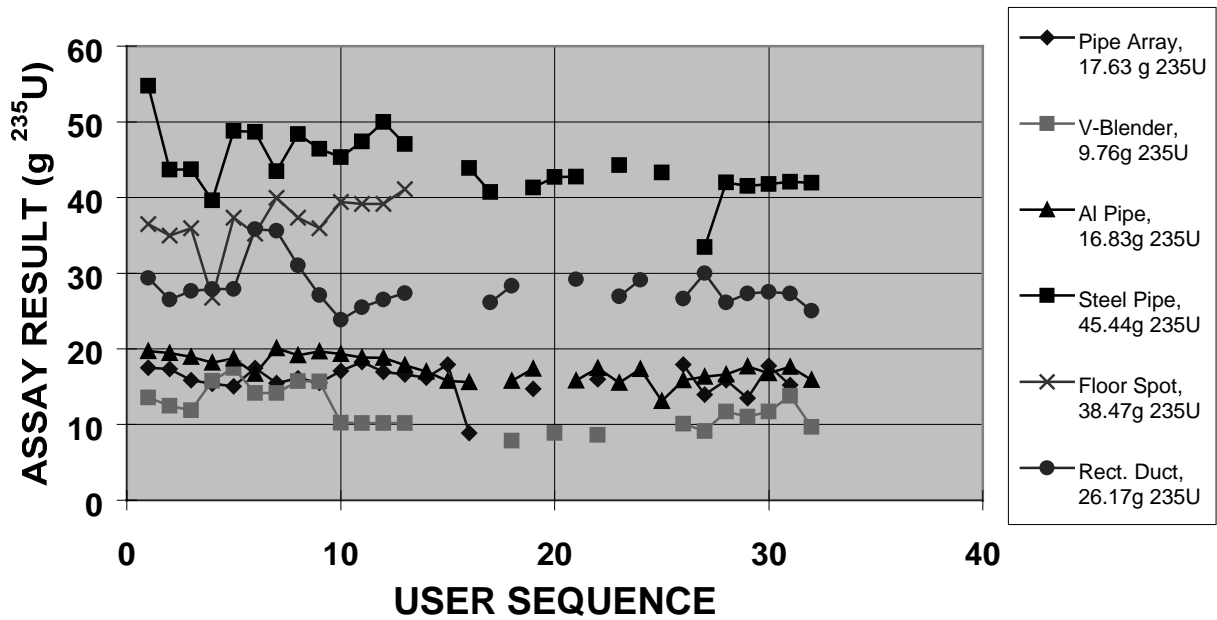


Fig. 5. Portable uranium holdup measurements (automated). The GGH assay result for ^{235}U mass obtained with the automated integrated holdup measurement system for each of the six holdup simulations by each measurement team involved in the measurements during the three-year test period is plotted in chronological sequence.

The standard deviation in the ratio of assay to reference values obtained in the manual measurements of ^{235}U for each piece of equipment is greater than that for the automated measurements, despite the use of comparable or somewhat longer count times in the manual measurements. This is most apparent in the dramatically larger fluctuations in the data for each piece of equipment in Fig. 4 compared with Fig. 5. It is also observed in the tabulated results. Table III includes the results obtained in the manual mode and Tables IV and V are results from the automated measurements with the integrated system. Comparison of average assay-to-reference ratio for each piece of equipment and for all equipment in the three tables show comparable results for manual (Table III) and automated (Tables IV and V) measurements. However, the standard deviation in the ratio for each piece of equipment and for all equipment is twice as large for the manual (Table III) compared with the automated (Tables IV and V) measurements.

TABLE III. Portable Holdup Assay Results (g ²³⁵U) Obtained Without Automation.

Process Equipment, Holdup Reference Value		Pipe Array, 17.63 g ²³⁵ U	V-Blender, 9.76g ²³⁵ U	Al Pipe, 16.83g ²³⁵ U	Steel Pipe, 45.44g ²³⁵ U	Floor Spot, 38.47g ²³⁵ U	Rect. Duct, 26.17g ²³⁵ U	Average Std. Dev.
Year, MCA	User*							
'92, PMCA	Group 1	11	10		48		58	
	Group 2	13	9	17	45		24	
	Group 3	11	12	17	48			
	Group 4	14	12	18	42		28	
	Group 5	9	11	20	50			
	Group 6	13	10	23	43		44	
	Group 7	18	10	23	55		27	
	Group 8		13		39		18	
	Group 9	18	17	20	43			
	Group 10	17	18	18				
	Group 11		11	16	42			
	Group 12		12	23	46		31	
('92, PMCA _{all} Avg)/ ²³⁵ U _{ref}		0.78	1.24	1.16	1.00	na	1.26	1.09
('92, PMCA _{all} 1 σ)/ ²³⁵ U _{ref}		0.19	0.28	0.16	0.10	na	0.52	0.25
'93, PMCA	Group 13	15	6					
	Group 14	16	7					
	Group 15				45			
	Group 16			19				
	Group 17			18				
	Group 18		7		44			
	Group 19		6		39			
	Group 20	19	5	17	43		34.5	
	Group 21	19	9	16	45			
	Group 22	16	8	20	46			
	Group 23	16	8	20	49			
	Group 24		5		53			
('93, PMCA _{all} Avg)/ ²³⁵ U _{ref}		0.95	0.69	1.09	1.00	na	1.32	1.01
('93, PMCA _{all} 1 σ)/ ²³⁵ U _{ref}		0.10	0.14	0.10	0.09	na	na	0.11
'94, PMCA	Group 25		5	16	50			
	Group 26			18	47			
	Group 27	8	15	21	49			
	Group 28	19		16			50	
	Group 29	13	7	23	47			
	Group 30	8	6	19	46		44	
	Group 31		6		36		52	
	Group 32		6	12				
	Group 33		6	16	39		38	
	Group 34		6	15	42			
	Group 35	8		17				
	Group 36							
('94, PMCA _{all} Avg)/ ²³⁵ U _{ref}		0.64	0.73	1.03	0.98	na	1.76	1.03
('94, PMCA _{all} 1 σ)/ ²³⁵ U _{ref}		0.28	0.33	0.19	0.11	na	0.24	0.23
('92-4, PMCA _{all} Avg)/ ²³⁵ U _{ref}		0.80	0.93	1.09	1.00	na	1.43	1.05
('92-4, PMCA _{all} 1 σ)/ ²³⁵ U _{ref}		0.22	0.37	0.16	0.10	na	0.47	0.26

* student groups of 2

TABLE IV. Portable Replicate HMS2-Automated Holdup Assay Results (g ²³⁵U).

Process Equipment, Holdup Reference Value		Pipe Array, 17.63 g ²³⁵ U	V-Blender, 9.76g ²³⁵ U	Al Pipe, 16.83g ²³⁵ U	Steel Pipe, 45.44g ²³⁵ U	Floor Spot, 38.47g ²³⁵ U	Rect. Duct, 26.17g ²³⁵ U	Average Std. Dev.
Year, MCA	User*							
'92, PMCA	JKS	17.6	13.7	19.5	49.2	36.0	30.2	
		15.4	13.4	18.6	51.0	36.9	29.7	
		19.1	15.2	19.3	42.9	38.5	23.7	
		16.2	14.9	16.8	51.3	43.8	26.8	
		17.5	13.6	19.7	54.8	36.5	29.4	
(⁹², PMCA_{JKS} Avg)/²³⁵U_{ref}		0.97	1.45	1.12	1.10	1.00	1.07	1.12
(⁹², PMCA_{JKS} 1 σ)/²³⁵U_{ref}		0.08	0.08	0.07	0.10	0.08	0.10	0.17
'92, PMCA	SES	15.0	13.6	19.9	47.0	35.3	31.2	
		16.5	14.9	19.2	47.9	34.0	29.2	
		16.0	13.4	19.9	49.6	36.0	28.5	
		17.0	14.6	15.8	54.3	35.7	25.7	
		17.3	12.5	19.4	43.7	35.0	26.5	
(⁹², PMCA_{SES} Avg)/²³⁵U_{ref}		0.93	1.41	1.12	1.07	0.91	1.08	1.09
(⁹², PMCA_{SES} 1 σ)/²³⁵U_{ref}		0.05	0.10	0.10	0.09	0.02	0.08	0.18

* experienced individual users

TABLE V. Portable HMS2-Automated Holdup Assay Results (g ²³⁵U).

Process Equipment, Holdup Reference Value		Pipe Array, 17.63 g ²³⁵ U	V-Blender, 9.76g ²³⁵ U	Al Pipe, 16.83g ²³⁵ U	Steel Pipe, 45.44g ²³⁵ U	Floor Spot, 38.47g ²³⁵ U	Rect. Duct, 26.17g ²³⁵ U	Average Std. Dev.
Year, MCA	User							
'92, PMCA	JKS***	17.5	13.6	19.7	54.8	36.5	29.4	
	SES***	17.3	12.5	19.4	43.7	35.0	26.5	
	PAR***	15.9	11.9	18.9	43.7	36.0	27.7	
	GAS***	15.4	15.8	18.2	39.6	26.8	27.9	
	MNH***	15.1	17.4	18.7	48.8	37.4	27.9	
	RWL***	17.4	14.2	16.7	48.7	35.3	35.8	
	TKL***	15.5	14.2	20.1	43.5	40.0	35.6	
	JKH***	16.1	15.7	19.1	48.4	37.4	31.1	
	LLP***	15.5	15.7	19.7	46.4	36.0	27.1	
('92, PMCA _{all} Avg)/ ²³⁵ U _{ref}		0.92	1.49	1.13	1.02	0.92	1.14	1.10
('92, PMCA _{all} 1 σ)/ ²³⁵ U _{ref}		0.06	0.18	0.06	0.10	0.09	0.14	0.21
'93, M ³ CA	Group A**	17.1	10.2	19.3	45.3	39.4	23.9	
	Group B**	18.2	10.2	18.8	47.4	39.2	25.5	
	Group C**	16.9	10.2	18.8	50.0	39.2	26.5	
	Group D**	16.6	10.2	17.8	47.1	41.1	27.3	
('93, M ³ CA _{all} Avg)/ ²³⁵ U _{ref}		0.98	1.05	1.11	1.04	1.03	0.99	1.03
('93, M ³ CA _{all} 1 σ)/ ²³⁵ U _{ref}		0.04	0.00	0.04	0.04	0.02	0.06	0.05
'94, M ³ CA	Group a*	16.2		17.0				
	Group b*	17.9		15.8				
	Group c*	8.9		15.6	43.9			
	Group d*				40.7		26.1	
	Group e*		7.9	15.7			28.3	
	Group f*	14.7		17.4	41.3			
	Group g*		8.9		42.7			
	Group h*			15.8	42.8		29.2	
	Group i*	16.0	8.6	17.4				
	Group j*			15.5	44.3		26.9	
	Group k*			17.3			29.1	
	Group l*			13.1	43.3			
	Group m*	17.9	10.1	15.8			26.6	
	Group n*	13.9	9.1	16.3	33.5		30.0	
	Group o*	15.9	11.7	16.6	42.0		26.1	
	Group p*	13.5	11.0	17.7	41.5		27.3	
	Group q*	17.8	11.7	16.8	41.8		27.5	
Group r*	15.2	13.8	17.6	42.1		27.3		
Group s*		9.7	15.9	42.0		25.0		
('94, M ³ CA _{all} Avg)/ ²³⁵ U _{ref}		0.87	1.05	0.97	0.92	na	1.05	0.97
('94, M ³ CA _{all} 1 σ)/ ²³⁵ U _{ref}		0.15	0.18	0.07	0.06	na	0.06	0.08
('92-4, MCAs _{all} Avg)/ ²³⁵ U _{ref}		0.90	1.22	1.03	0.97	0.96	1.07	1.03
('92-4, MCAs _{all} 1 σ)/ ²³⁵ U _{ref}		0.11	0.27	0.10	0.09	0.09	0.11	0.11

* student groups of 1-2

** student groups of 6 plus instructor

*** experienced and inexperienced individual users

Table IV contains the results of a special series of automated measurements performed in repeated sequences by two users who are experienced in the operation of spectroscopy equipment and measurements of holdup. In general, results obtained in the manual mode by such users are significantly better than those obtained by inexperienced users who, for example, are more likely to control the measurement less effectively, select biased sampling plans, and use the available time less optimally. In the automated mode, however, neither the ratio of the average assay-to-reference value for each piece of equipment and for all equipment nor the standard deviation in this ratio differs significantly between measurements performed by experienced and inexperienced users.

B. Plutonium

Figures 6 and 7 are graphs of the results of plutonium holdup measurements obtained in a single year of these exercises. The groups of users of the portable spectroscopy equipment also consisted primarily of those who were inexperienced in the operation of this equipment and in measurements of holdup. There are more measurement results for automated than for manual measurements because most groups were unable to complete half of the measurement exercises in the manual mode. Plots of the individual assay results for both the manual (Fig. 6) and automated (Fig. 7) measurements show reasonably good agreement, on average, with the reference values. This is also shown in Tables VI and VII by values consistent with unity for the ratios of the average assay ($^{239}\text{Pu}_{\text{mass}}$) result to the reference value for each piece of equipment, and the average (of averages) for all equipment.

The standard deviation in the ratio of assay to reference values obtained in the manual measurements of ^{239}Pu for each piece of equipment is comparable with that for the automated measurements, although the count times for the manual measurements were approximately six times those used for the automated measurements. This comparable result is apparent in the fluctuations in the data for each piece of equipment in Fig. 5 compared with Fig. 6. It is also observed in the tabulated results for the standard deviations in Tables VI and VII.

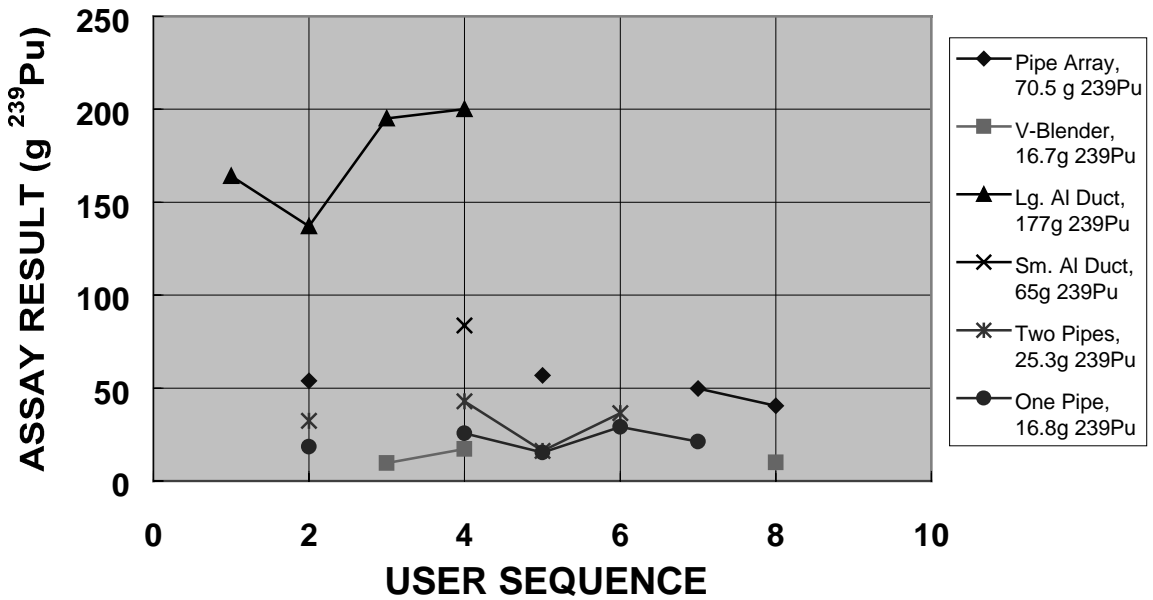


Fig. 6. Portable plutonium holdup measurements (no automation). The GGH assay result for ^{239}Pu mass obtained in 60-s counts by manual holdup measurements for each of the six holdup simulations by each measurement team involved in the measurements during the test period is plotted in chronological sequence.

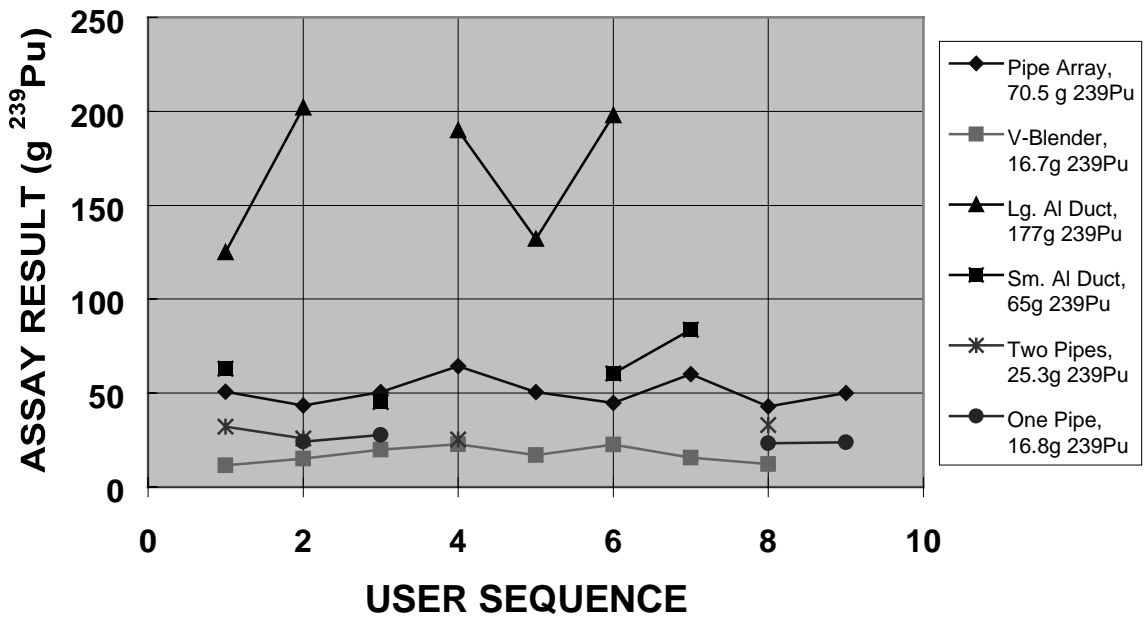


Fig. 7. Portable plutonium holdup measurements (automated). The GGH assay result for ^{239}Pu mass obtained in 60-s counts with the automated integrated holdup measurement system for each of the six holdup simulations by each measurement team involved in the measurements during the test period is plotted in chronological sequence.

TABLE VI. Portable Holdup Assay Results (g ²³⁹Pu) Obtained Without Automation.

Process Equipment, Holdup Reference Value		Pipe Array, 70.5 g ²³⁹ Pu	V-Blender, 16.7g ²³⁹ Pu	Lg. Al Duct, 177g ²³⁹ Pu	Sm. Al Duct, 65g ²³⁹ Pu	Two Pipes, 25.3g ²³⁹ Pu	One Pipe, 16.8g ²³⁹ Pu	Average Std. Dev.
Year, MCA	User*							
'96, PMCA	Group i	53.9	9.6	164	83.7	32.4	18.4	
	Group ii			137				
	Group iii			195				
	Group iv	56.9	17.3	200	42.8	25.7		
	Group v			16.2				
	Group vi	49.8	10			36.5	29.1	
	Group vii					21.1		
	Group viii					40.4		
	Group ix							
('96, PMCA _{all} Avg)/ ²³⁹ Pu _{ref}		0.71	0.74	0.98	1.29	1.26	1.30	1.05
('96, PMCA _{all} 1 σ)/ ²³⁹ Pu _{ref}		0.10	0.26	0.17	N.A.	0.45	0.33	0.28

*student groups of 2-3

TABLE VII. Portable HMS2-Automated Holdup Assay Results (g ²³⁹Pu).

Process Equipment, Holdup Reference Value		Pipe Array, 70.5 g ²³⁹ Pu	V-Blender, 16.7g ²³⁹ Pu	Lg. Al Duct, 177g ²³⁹ Pu	Sm. Al Duct, 65g ²³⁹ Pu	Two Pipes, 25.3g ²³⁹ Pu	One Pipe, 16.8g ²³⁹ Pu	Average Std. Dev.		
Year, MCA	User*									
'96, PMCA	Group I	50.7	11.5	125.0	62.9	32.2	24.2			
	Group II			202.0						
	Group III			190.0						
	Group IV	50.6	17.0	132.0	45.3	25.3	27.7			
	Group V			198.0						
	Group VI	44.7	22.7	198.0	60.5	83.7	33.1		23.3	
	Group VII									23.8
	Group VIII									42.9
	Group IX	50.1								
('96, PMCA _{all} Avg)/ ²³⁹ Pu _{ref}		0.72	1.02	0.96	0.97	1.15	1.47	1.05		
('96, PMCA _{all} 1 σ)/ ²³⁹ Pu _{ref}		0.10	0.26	0.21	0.24	0.16	0.12	0.25		

*student groups of 2-3

IV. DISCUSSION

A. Conclusions

Benefits to the facility operator of reduced holdup assay uncertainty, equivalent accuracy, and increased cost effectiveness observed with the automated integrated holdup measurement system are most apparent in the results of these extended tests of the GGH capability with and without automation.

1. A factor of 2 improvement in the overall uncertainty of the quantitative assay of ^{235}U holdup is observed for results obtained in a three-year period with the integrated holdup measurement system automated by the HMS2 software compared with the precision obtained in the conventional measurements without automation performed under otherwise identical circumstances. The overall uncertainty of the quantitative assay of ^{239}Pu achieved with the integrated holdup measurement system automated by the HMS2 software using 10-s counts is comparable with the precision obtained in the conventional measurements performed in 30- to 60-s counts without automation (under otherwise identical circumstances). The automated system has several advantages that bring about the improved results.
 - The automation controls the measurement quality by compensating for gain drift and verifying accuracy.
 - The manner in which the GGH procedures are applied is the same for all users of the automated (but not the manual) system.
 - The automated system employs optimized choices for the reduction and analysis of the GGH data, compared with those made in the manual mode (despite guidance from written instruction).
 - The automated system eliminates most of the possible errors in manual transcription of data and manual calculation of results.
2. For most of the quantitative measurements, the accuracy of the assay is equivalent for both automated and manual approaches. This is because the accuracy is based on the generalized-geometry calibration of the holdup measurement in both cases.
3. For holdup measurements of both uranium and plutonium, the automated system reduces the time required to obtain the holdup measurement results by a factor of 20. Furthermore, because the automation is combined with highly portable equipment, only one person rather than two is required to perform the measurements. Thus, the cost effectiveness of routine measurements of holdup (for which automated system was designed originally) is greatly increased over that provided by the manual measurements.

B. Follow-on Work

New software has been written to replace the HMS2 software that automates the data acquisition, reduction, and analysis of gamma-ray spectra; and evaluation, logging, and tracking of holdup quantities. Version 1.0 of the HMS3 software,⁶ written in Microsoft Visual Basic® Professional v 4.0 under Microsoft Windows, is now available. It uses Microsoft Access® database files. This transitional version of HMS3 is equivalent to HMS2 with enhanced spectroscopic functions and Windows advantages that include a graphical user interface. Testing of HMS3 v 1.0 is underway.

Plans for the subsequent version of HMS3 include the implementation of corrections that are not currently performed by the automated system. The HMS2 software automates the subtraction of room background and the point-by-point correction for attenuation of gamma-ray intensities by the process equipment. The HMS3 software has retained the point-by-point correction capability for equipment attenuation effects and has additional flexibility in the approach to subtraction of room background. Corrections for finite source dimensions and gamma-ray self-attenuation effects in the framework of the GGH formalism⁷ will be implemented in the next version of HMS3.

Efforts are under way to implement measurements of uranium holdup with the integrated system using the new commercial prototype coplanar-grid CdZnTe detector with resolution that is better than that of NaI. For assay of ²³⁵U, the new room-temperature detector with a thickness of only 0.5 cm will provide approximately 35% of the detection efficiency of the compact NaI detector at 186 keV. The performance goal is to achieve an energy resolution that is more than a factor of 2 better at this energy. A thickness of 1.5 cm achieves a similar relative detection efficiency at 400 keV to address the ²³⁹Pu assay needs with a resolution that is more than twice as good as that of NaI. For certain uranium holdup circumstances, biases from gamma-ray interferences that may be a concern with the compact NaI detectors will be diminished because of the improved resolution. Similarly, variations in plutonium isotopic composition or americium content that introduce biases in the assay of ²³⁹Pu with NaI detectors have relatively little effect of the assay with the improved-resolution room-temperature gamma-ray detectors.⁸ The use of bulky, liquid-nitrogen-cooled HPGe detectors addresses the bias problem but eliminates the benefits of rapid measurements and operation by a single user that are otherwise provided by the integrated system. The solid state devices are more compact (with shielded weights that are 90% of the weights of the compact NaI detector for uranium measurements and only 60% of those for plutonium measurements) and rugged than the scintillator/photomultiplier tube combinations, and operate with lower power needs, all of which benefit the portable applications. Implementation of these new room-temperature gamma-ray detectors with the integrated holdup measurement system, which is fully compatible with these devices, is currently underway.

REFERENCES

1. P. A. Russo, J. K. Sprinkle, Jr., J. K. Halbig, S. F. Klosterbuer, and S. E. Smith, "Generalized-Geometry Gamma-Ray Holdup Assay," Los Alamos National Laboratory Application Note LALP-94-72 (February 1995).

2. P. A. Russo, H. A. Smith, J. K. Sprinkle, Jr., C. W. Bjork, G. A. Sheppard, and S. E. Smith, "Evaluation of an Integrated Holdup Measurement System Using the GGH Formalism with the M³CA," Los Alamos National Laboratory document LA-UR-95-3321 (September 1995).
3. S. E. Smith, J. S. Gibson, J. K. Halbig, S. F. Klosterbuer, P. A. Russo and J. K. Sprinkle, Jr., "The Holdup Measurement System II (HMSII)," *Nucl. Mater Manage.* **XXII** (Proc. Issue) 508-512 (1993).
4. P. A. Russo, M. M. Stephens, and S. C. Bourret, "Compact Nal(Tl) Detectors," in Safeguards and Security Progress Report, January–December 1989, Los Alamos National Laboratory report LA-11914-PR, pp. 20-22 (November 1990).
5. J. K. Halbig, S. F. Klosterbuer, P. A. Russo, J. K. Sprinkle, Jr., S. E. Smith, and K. Ianakiev, "Advances in Gamma-Ray Field Instrumentation at Los Alamos," in Proceedings of IAEA Symposium on International Safeguards held March 14-18, 1994, Vienna, Austria, IAEA-SM-333/92, Los Alamos National Laboratory document LA-UR-94-276.
6. S. E. Smith, K. A. Thompson, and R. N. Ceo, "Holdup Measurement System 3 (HMS3) User's Guide and Software Documentation," Lockheed Martin Energy Systems report Y/DK-1104 (1997).
7. P. A. Russo, M. C. Sumner, T. K. Li, H. O. Menlove, and T. R. Wenz, "Quantitative Verification of In-Process Inventory Using Room Temperature Gamma-Ray Detectors and the GGH Formalism," in Safeguards and Security Research and Development Progress Report, October 1994–September 1995, Los Alamos National Laboratory report LA-13250-PR, pp. 37-46 (March 1997).
8. P. A. Russo, A. P. Meier, M. Rawool-Sullivan, T. H. Prettyman, H. Y. Huang, D. A. Close, M. C. Sumner, R. A. Cole, P. N. Luke, and S. A. Soldner, "A New Room-Temperature Gamma-Ray Detector for Improved Assay of Plutonium," *Nucl. Mater. Manage.* **XXVI** (Proc. Issue) CD-ROM (1997).

This report has been reproduced directly from the best available copy. It is available electronically on the Web (<http://www.doe.gov/bridge>).

Copies are available for sale to U.S. Department of Energy employees and contractors from—

Office of Scientific and Technical Information
P.O. Box 62
Oak Ridge, TN 37831
(423) 576-8401

Copies are available for sale to the public from—

National Technical Information Service
US Department of Commerce
5285 Port Royal Road
Springfield, VA 22616
(800) 553-6847

