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ABSTRACT

A high-resolution gamma ray spectroscopy (HRGS) system has been installed at the Advanced Material Accountancy Glovebox Assay (AMAGB) System so that neutron and gamma ray measurements can occur simultaneously on material transfer containers. This equipment was installed in August 2000 at the Plutonium Fuel Production Facility (PFPF) in Japan. It is anticipated that placing the HRGS system at the neutron assay station will save a few person-days per IAEA inspection because it will no longer be necessary to take samples from the transfer container for isotopic analysis at another assay station. The HRGS system consists of a 25% relative efficiency coaxial detector with electromechanical cooling and digital signal processing. The digital spectrometer has the benefit of increasing throughput, improving energy resolution, and extending the lifetime of neutron damaged HPGe detectors as compared to analog spectrometers. The gamma-ray system is used to verify the Pu isotopic composition and ^{241}Am concentration in transfer containers of MOX. The $^{240}Pu_{eff}$ fraction is calculated from the isotopic data and is used to convert the neutron assay data to total grams plutonium. The results of performance tests and calibration measurements using this system are reported in this paper.

INTRODUCTION

The Advanced Material Accountancy Glovebox (AMAGB) assay system is used to assay plutonium content of mixed oxide (MOX) reactor fuel materials in the form of powder and pellets. Two systems are located at the Plutonium Fuel Production Facility (PFPF) in Japan, which is operated by the Japan Nuclear Cycle Development Institute (JNC). The assay system is used by the International Atomic Energy Agency (IAEA), the Japan Safeguards Office (JSGO), and JNC during monthly safeguards inspections.

The AMAGB assay system consists of two components: neutron and gamma-ray assay. The neutron measurement determines the $^{240}Pu_{eff}$ mass[1] using the known-alpha analysis method.[1] The gamma-ray measurement determines the Pu and ^{241}Am isotopic fractions in the sample, using either the FRAM[2] or MGA[3,4] analysis software. The isotopic software calculates the $^{240}Pu_{eff}$ fraction that is used to convert the neutron assay mass to total grams of Pu. One unit is located around a material accountancy glovebox, and the other is located around a supplemental transfer line that connects gloveboxes located in different areas together. The neutron detectors are permanently mounted around the glovebox transfer lines, so the neutron assay can be performed in attended or unattended mode. The detectors for the gamma-ray measurement have to be positioned at the neutron assay system for every measurement (these are not permanently mounted), so this measurement is performed only in attended mode for this particular application.

The unique feature of this assay system is that the high purity Ge (HPGe) detectors used for the gamma-ray measurement use electromechanical cooling instead of liquid nitrogen. The remainder of this report will discuss the high resolution gamma-ray spectroscopy (HRGS) equipment, the measurement performance with the mechanical cooling system, and the latest commercial equipment developments for HRGS measurements using mechanical cooling.

EQUIPMENT

Each HRGS system consists of:

- a Canberra HPGe coaxial detector (model GC25175) with 25% relative efficiency
- a Canberra Cryoelectric II electrically refrigerated cryostat
- 25 ft gas lines between the HPGe detector and the cryostat
- a Perkin Elmer (ORTEC) DSPEC-Plus digital spectrometer
- a tungsten collimator with additional cadmium filters
- a desktop computer with a Windows 95/98/NT operating system
- Perkin Elmer MAESTRO software to operate the DSPEC and collect the gamma-ray spectra, and
- FRAM and MGA isotopic analysis software

Figures 1 through 3 are photographs of the HPGe detector components. Figure 1 shows the detector attached to the support plate that provides rigid support for both the detector and the front and rear tungsten shields. The detector/shield support plate mounts to the lifter supplied by the facility. The measurement location is approximately 3 meters above the floor, so a mechanical lifting device is needed to support the weight of the detector and collimator. The purpose of the rear shield is to block gamma rays coming from other material located in the room. Measurements made at the facility during the calibration exercise show that room background is not a problem, so the rear shield does not need to be used, but it is available if the need occurs.

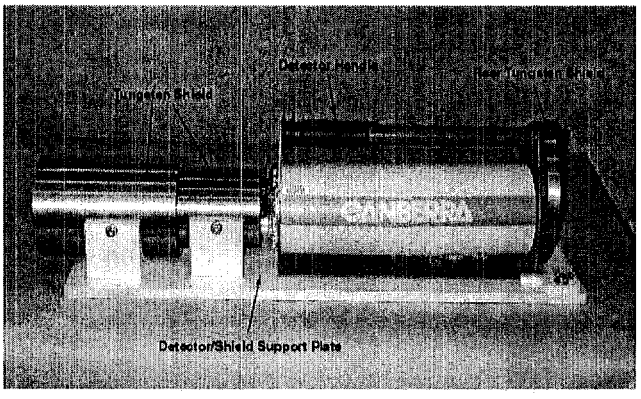


Fig. 1. Photograph of the HPGe detector mounted on the support plate with the tungsten shield. A rear tungsten shield is also included to shield the detector from material located directly behind it.

Figure 2 shows the LEDs that are on the preamplifier portion of the HPGe detector–these are visible on the opposite side of the detector shown in Fig. 1. A rectangular hole was cut in the tungsten collimator so that these lights are visible. The LEDs operate when the detector is connected to the DSPEC Plus spectrometer with the main power *on* the HV turned *off*. This configuration supplies power to the preamplifier for checking the status of the LEDs prior to turning on the HV.

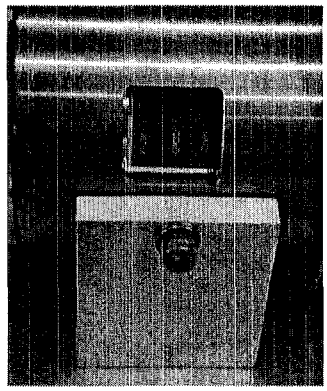


Fig. 2. HRGS preamplifier indicator LEDs. From left-to-right: Warm, Cold, Rate.

Fig. 3. Photograph of the HPGe detector resting on top of the cooling unit.

The Warm LED (red) is *on* when the detector has not cooled down. When the Warm light is *on*, HV should not be applied to the detector. The Cold LED (green) illuminates when the detector is cooled down indicating that it is safe to turn on the HV. The Rate LED (yellow) illuminates when the preamplifier is overloaded with pulses. When this LED is *on*, it is an indicator that a high activity source is being counted and the count rate needs to be reduced by increasing the source-to-detector distance or shielding. This LED could also indicate that the detector is warming up (excessive leakage current). The manufacturer specifies that it takes 24 hours for the detector to cool down; however, the green light illuminates after about 8 hours of cooling with these detectors.

Figure 3 is a photograph of the HPGe detector mounted on top of the mechanical cooling unit. The dimension, weight, and power specifications along with the cost at the time of purchase are listed in Table I for the complete HRGS system.

Table I. Selected specifications for the HRGS system

Equipment	Dimension W x H x D	Weight	Power Requirement	Cost in Fall 1999
Canberra HPGe coaxial detector	5" x 7.6" x 23"	14.5 lb	N/A	$17,400
Canberra Cryoelectric II Cryostat	17.8" x 14.5" x 12.1"	48 lb	500 Watts	$17,000
25ft gas lines	—	—	N/A	$400[a]
Perkin Elmer DSPEC-Plus	12.8" x 5.7" x13.7"	17 lb	110 Watts	$10,000
Desktop computer	16.1" x 4.3 " x 17.2"		145 Watts	$2,200
Software	N/A	N/A	N/A	No charge[b]

[a] 10 ft lines are included in the price of the cryostat. This price is the added cost for 25 ft lines.

[b] The Maestro software is included with the DSPEC. The FRAM and MGA software were supplied by the IAEA, so there was no direct cost associated with their use.

Figure 4 illustrates the HRGS measurement geometry for the plutonium isotopic measurement at AMAGB #2. For AMAGB #1, the four neutron detector slabs completely enclose the glovebox, so the HPGe detector is positioned against one of the neutron detector slabs. As a result, the gamma-ray signal is much lower than it is for the system at AMAGB #2 because of the attenuation from the neutron detector. The HPGe detector plus its tungsten shield will be mounted on a mechanical lifter to position the detector on the center of the sample.

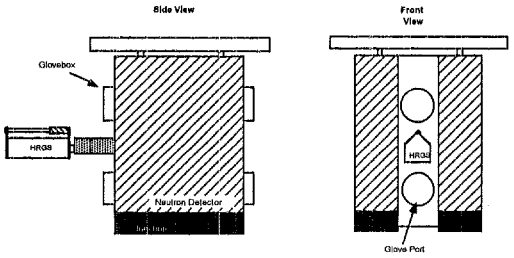


Fig. 4. Front- and side-view diagrams of AMAGB #2 showing the location of the HRGS detector head during a measurement.

Inside the tungsten shield is a thin (0.04 cm) cadmium liner that fits around the outer cylindrical surface of the HPGe detector to absorb x-rays from capture processes that occur in the tungsten. Multiple 7.6-cm diameter disks of cadmium with thicknesses of 0.04 and 0.08 cm are included. These disks are to be placed in front of the detector crystal to reduce the count rate from the 60-keV gamma rays generated from ^{241}Am in the MOX samples. These disks can be put inside the tungsten shield, which fixes the position of the disks when the HPGe detector is placed inside the shield. Using this method eliminates the need to tape the disks to the front of the tungsten shield.

The tungsten collimator for AMAGB #1 has a 5-cm diameter through hole to maximize the number of gamma rays that reach the detector. This is important for AMAGB #1, because the neutron detector slab attenuates the gamma-ray signal. For AMAGB #2, the tungsten collimator has a 2.5 cm through hole to reduce the deadtime for large samples. In general, the HPGe detectors should be positioned to keep the deadtime below 40%. However, with the digital spectrometers having higher signal throughput, deadtimes around 50% can result in optimal count rates[5].

PERFORMANCE

The HPGe coaxial detectors were specified with a 25% relative efficiency and a resolution of 1.75 keV at 1332 keV. Three detectors, each with electromechanical cooling, were purchased. All the manufacturers will guarantee the resolution within 10% of the specified value when electro-mechanical cooling is used, but the detectors tend to perform better than this. Several sets of measurements were performed with the detectors using ^{57}Co and ^{60}Co radioisotopic sources to check the resolution of the systems and look at the stability of both the resolution and centroids of the photopeaks at 122 keV (^{57}Co) and 1332 keV (^{60}Co). Table II lists the average resolution values reported by the manufacturer for the three HRGS systems.

Table II. Average resolution for the 3 HRGS systems reported by the manufacturer

	Res. at 122 keV	Res. at 1332 keV
Average	0.82 keV	1.72 keV

Figures 5 and 6 show the resolution and centroid performance of Unit 1. These data were measured with a ^{57}Co source over a period of several weeks. At the beginning, data were collected for three days then the system was shutdown for five days, moved to a new location, and data collection was started up again for 15 days. The average resolution for the period is 0.79 keV at 122 keV, which matches the value measured by the manufacturer. The centroid data in Fig. 6 shows a slight gain shift from when the unit was turned off and turned back on again. Very minor

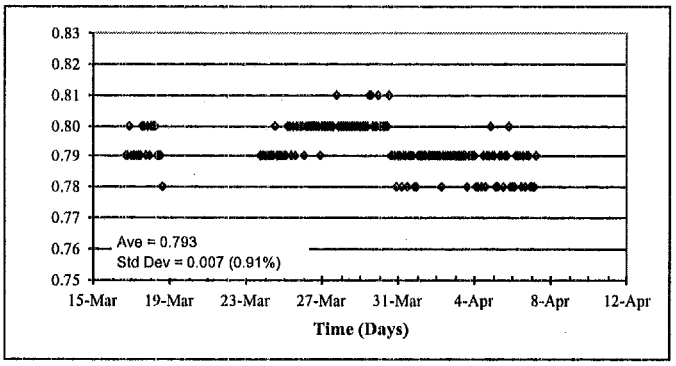


Fig. 5. Resolution data from Unit 1 for the 122 keV photopeak from ^{57}Co as a function of time.

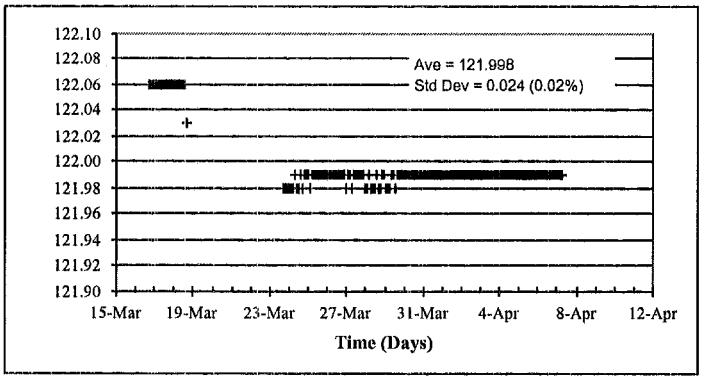


Fig. 6. Centroid data from Unit 1 for the 122 keV photopeak from ^{57}Co as a function of time.

gain shifts have been observed when a unit is shutdown and turned on again. The shift is very small–approximately half a channel.

Figures 7 and 8 show plots of the measured $^{240}Pu_{eff}$ fraction as a function of time, from Units 1 and 2. These data are from repeat measurements of a single MOX pellet with a count time (live time) of 2 hours. The average and the one-standard deviation lines are shown in the figures along with the one-sigma error bars for each measurement.

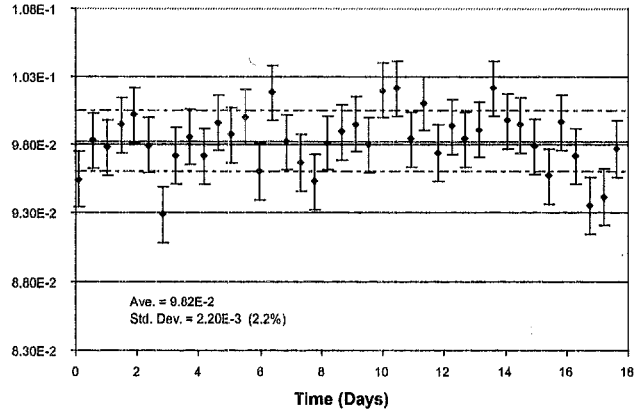


Fig. 7. Measured $^{240}Pu_{eff}$ fraction for Unit 1 from a MOX pellet as a function of time. The count time was 2 hours. The 1-sigma error bars are shown along with the average and 1-std deviation lines.

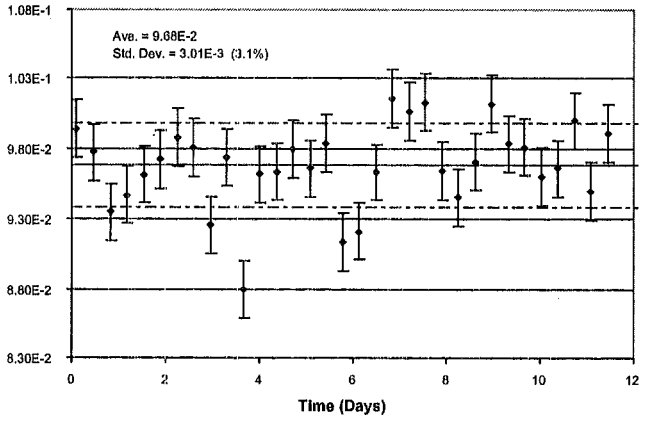


Fig. 8. Measured $^{240}Pu_{eff}$ fraction for Unit 2 from a MOX pellet as a function of time. The count time was 2 hours. The 1-sigma error bars are shown along with the average and 1-std deviation lines

Table III lists a representative isotopic composition of the calibration samples measured at the facility in Japan. Sample IDs beginning with "R" are powder containers, and IDs beginning with "T" are pellet containers. The pellet containers are square and have multiple trays that hold the pellets.

Table III. Average isotopics of calibration standards measured in AMAGB #1 and #2

Sample ID	^{238}Pu (wt%)	^{239}Pu (wt%)	^{240}Pu (wt%)	^{241}Pu (wt%)	^{242}Pu (wt%)	Pu/Am Date (yy/mm/dd)	^{241}Am (ppm)
Ave.	1.215	62.081	24.824	7.512	4.368	97/07/24	17,700

Tables IV and V show the $^{240}Pu_{eff}$ fractions calculated from the isotopic measurements made at each AMAGB station. Included in the tables are the sample identification number, the measurement count time (live time), the measurement deadtime, the measured $^{240}Pu_{eff}$ percentage with its relative error from both FRAM and MGA, and the relative errors for a 15-min count (real time), which is equivalent to the count time used for the neutron measurement. The high-energy analysis options in MGA (Ver. 9.63) and FRAM (Ver. 3.2) were used to perform the isotopic analysis in these tables. The relative error on the AMAGB #1 measurements is larger because the gamma rays have to pass through the neutron detector slab before reaching the HPGe detector.

IV. $^{240}Pu_{eff}$ calculation from FRAM and MGA for the HRGS measurements at AMAGB #1

Sample ID	Live Time (min.)	Deadtime[a] (%)	*MGA* $^{240}Pu_{eff} \pm \sigma$ (%)	*MGA* σ for 15-min count	*FRAM* $^{240}Pu_{eff}$ (%)	*FRAM* σ for 15-min count	$^{240}Pu_{eff}$ (g)
R023	24.9	10	35.51 ± 7.81	10.6	35.71 ± 6.29	8.5	72
R027	15.8	22	36.62 ± 6.12	7.1	36.83 ± 4.71	5.5	1628
R027b	33.6	22	36.94 ± 4.54	7.7	36.06 ± 3.48	5.9	1628
R219	10.9	17	28.01 ± 10.12	9.4	25.74 ± 10.0	9.4	804
T027	80.7	8	36.81 ± 8.01	19.4	32.73 ± 7.76	18.8	439
T098	13.6	35	41.76 ± 6.07	7.2	35.71 ± 4.91	5.8	1091
T098b	40.4	35	36.90 ± 3.94	8.0	33.71 ± 3.26	6.6	1091
T148	12.5	2	29.85 ± 30.5	28.1	33.64 ± 28.5	26.3	132
T148b	25.0	3	33.76 ± 22.4	29.4	30.88 ± 19.0	24.9	132
Ave.				14.0		12.4	

[a] Deadtime(%) = 100*(RT-LT)/RT, where RT = real time and LT = live time

V. $^{240}Pu_{eff}$ calculation from FRAM and MGA for the HRGS measurements at AMAGB #2							
Sample ID	Live Time (min.)	Deadtime (%)	*MGA* $^{240}Pu_{eff} \pm \sigma$ (%)	*MGA* σ for 15-min count	*FRAM* $^{240}Pu_{eff}$ (%)	*FRAM* σ for 15-min count	$^{240}Pu_{eff}$ (g)
R023	26.2	10	33.67 ± 8.02	11.2	33.15 ± 5.04	7.0	72
R027	23.1	29	35.76 ± 3.56	5.2	36.44 ± 2.59	3.8	1628
R159	11.4	30	34.76 ± 5.66	5.9	34.60 ± 3.21	3.3	1687
R210	3.6	47	37.67 ± 5.96	4.0	36.24 ± 4.34	2.9	573
R210b	7.4	47	36.05 ± 4.88	4.7	35.09 ± 3.21	3.1	573
R215	12.4	39	34.19 ± 4.42	5.1	34.43 ± 3.20	3.7	1697
R219	24.6	38	35.40 ± 3.36	5.5	31.68 ± 2.67	4.3	804
T027	29.2	19	33.38 ± 4.54	7.0	35.12 ± 3.64	5.6	439
T098	38.5	34	36.25 ± 3.10	6.1	36.56 ± 2.16	4.3	1091
T098b	53.0	34	36.00 ± 2.84	6.6	36.51 ± 1.91	4.4	1091
Ave.				6.1		4.2	

Figures 9 and 11 are plots showing the Assay minus Declared values for the $^{240}Pu_{eff}$ percentages calculated using both analysis codes for both AMAGB systems. The value (A-D)/A is used instead of (D-A)/D because the relative errors included in the figures are relative to A, the assay value and not the declared value. Figures 10 and 11 are plots of the $^{240}Pu_{eff}$ relative errors calculated from the analysis codes for both AMAGB systems.

There is good agreement between the FRAM and MGA results. Increasing the measurement time to decrease the statistical uncertainty will reduce the differences between the declared and assay values in Figs. 9 and 11. Figures 10 and 12 show that the calculated error for FRAM is less than the error calculated from MGA. The likely reason for this difference is that the error calculations in MGA include an additional error term to account for uncertainties in the peak fitting, so the calculated errors for MGA are larger than they are for FRAM. There is no real reason to expect the errors in one code to be less than the errors in the other because they both are using the same/similar analysis energy regions for the isotopic calculations.

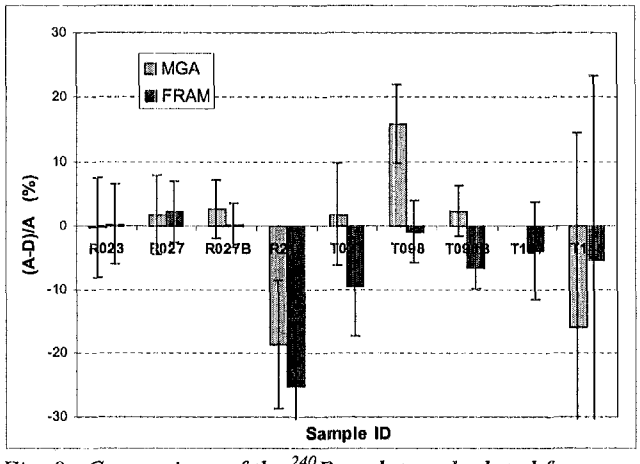


Fig. 9. Comparison of the $^{240}Pu_{eff}$ data calculated from MGA and FRAM for AMAGB #1. The one-sigma error bars are included.

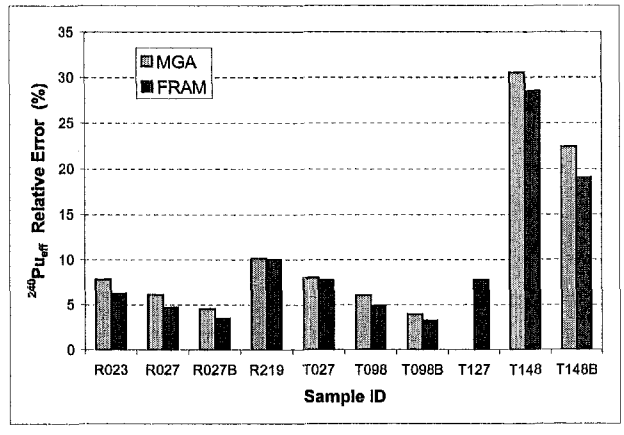


Fig. 10. Comparison of the relative uncertainties on $^{240}Pu_{eff}$ calculated from MGA and FRAM for AMAGB #1. The 1-sigma error bars are included.

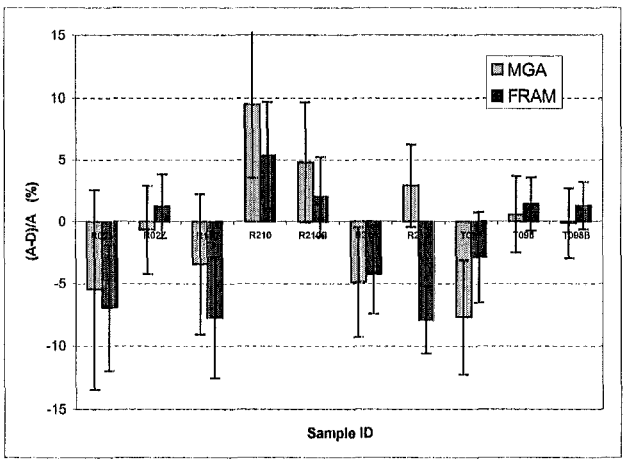


Fig. 11. Comparison of the $^{240}Pu_{eff}$ data calculated from MGA and FRAM for AMAGB #2. The 1-sigma error bars are included.

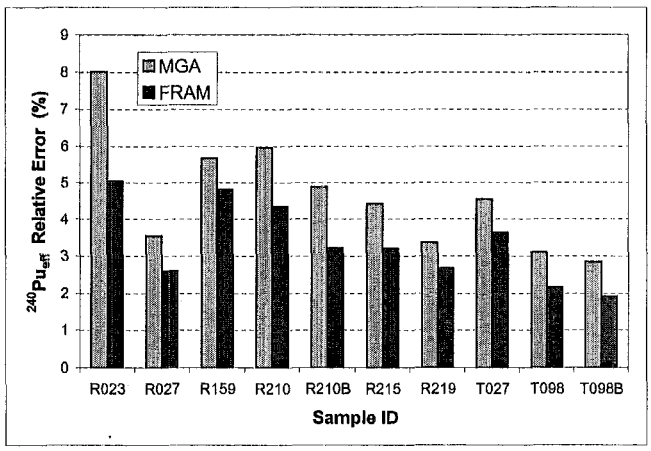


Fig. 12. Comparison of the $^{240}Pu_{eff}$ data calculated from MGA and FRAM for AMAGB #2. The one-sigma error bars are included in the figure.

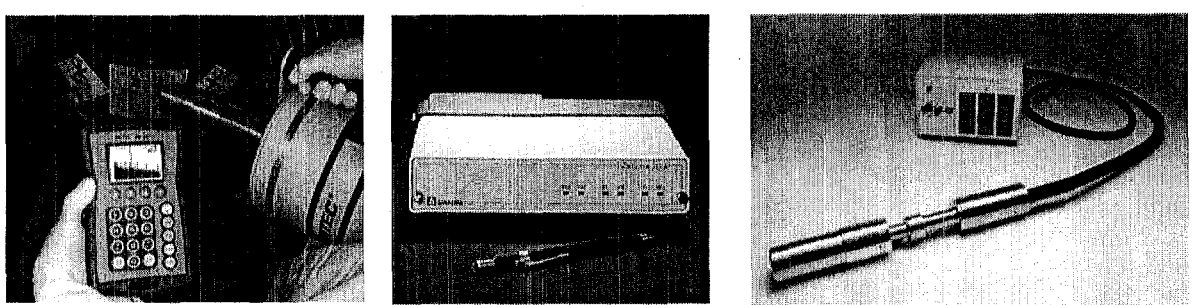
NEW EQUIPMENT

Recently, there have been advancements in miniaturizing the spectrometry electronics and the electromechanical cooling systems for HRGS systems. Both Canberra and Perkin Elmer have come out with hand-held digital spectrometers. Dimensional and power specifications for these units are listed in Table VI.

Table VI. Selected specifications for the hand-held digital spectrometers

Equipment	Dimension W x H x D	Weight	Power Requirement
Canberra Inspector 2000 (Fig 13b)	7.3" x 1.5" x 6.8"	2.8 lb	Battery, DC power supply
Perkin Elmer (ORTEC) DSPEC-Plus	12.8" x 5.7" x13.7"	17 lb	110 Watts
Perkin Elmer (ORTEC) DigiDART (Fig 13a)	5" x 3" x 8"	1.9 lb	Battery, DC power supply

Perkin Elmer has a new electromechanical cooler that is almost half the size of previous units available on the market. Figure 13 shows the Perkin Elmer X-COOLER with a Pop-Top HPGe detector attached, and Table VII compares the dimensions of the older units to the X-COOLER. The smaller hardware will save considerable space when installed in processing areas at fuel fabrication facilities.



(a) (b) (c)

Figure 13. (a) The Perkin Elmer DigiDart digital spectrometer, (b) the Canberra Inspector 2000 digital spectrometer, and (c) the Perkin Elmer X-COOLER with a Pop-Top HPGe detector.

Table VII. Selected specifications for the HRGS system				
Equipment	Dimension W x H x D	Weight	Power Requirement	Cost in Fall 1999
Canberra Cryoelectric II Cryostat	17.8" x 14.5" x 12.1"	48 lb	500 Watts (nominal)	$17,000 (Fall 99)
Perkin Elmer (ORTEC) ELECTRICOOL	17.8" x 14.5" x 12.1"	48 lb	520 Watts	$18,000 (Fall 99)
Perkin Elmer (ORTEC) X-COOLER	12" x 8.5" x 10.5"	36 lbs	500 W (init) 300 W (op)	$3,200 (2001)

SUMMARY

The HRGS systems with electromechanical cooling perform well for isotopic measurements. With the removal of liquid nitrogen cooling, operation of the detectors in processing areas is simplified. With the recent development of smaller spectrometers and cooling units, it is becoming more practical (from a space point of view) to use these systems in reprocessing areas. It has been estimated that performing isotopic measurements to confirm the operator-declared isotopic values will reduce the inspection effort by a couple of person-days per inspection because the amount of sampling for destructive analysis will be reduced. Also, the benefit of having immediate feedback from the nondestructive analysis will reduce the time to draw safeguards conclusions at the end of an inspection.

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