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FIELD TEST OF NEW TASTEX SYSTEM FOR PLUTONIUM PRODUCT VERIFICATION AT THE TOKAI REPROCESSING PLANT

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ABSTRACT

This report describes the field test results of the New TASTEX system. This system consisting of the high resolution gamma spectrometer and the k-edge densitometer can measure both isotopic abundances and concentration of plutonium simultaneously. Entire system is controlled by the multi-channel analyser and a multi-user computer. The system was designed and built under the Japan Support Program for Agency Safeguards(JASPAS). The software of this system developed at LANL and LLNL has been installed in the system assembled at the Tokai reprocessing plant(TRP) in July 1985. In the course of campaigns from 1985 till 1988, field tests have been carried out on plutonium product solutions of TRP. The results of plutonium concentration and isotopic abundances obtained by the k-edge densitometer and the high resolution gamma spectrometer (HRGS) have been compared with those by controlled potential coulometer and mass spectrometer respectively. Precision of plutonium determination with k-edge densitometer is estimated approximately 0.7% and 1.0% for the freshly processed plutonium and the aged plutonium respectively. The scatters in the relative differences between HRGS and the destructive analysis (DA) detected on the results of freshly processed plutonium sample were 1.6%, 0.4%, 0.5%, 1.1%, 8.0% for Pu-238, Pu-239, Pu-240, Pu-241, Pu-242 respectively, whereas those on the results of aged sample were 1.4%, 0.5%, 1.1%, 1.1% for Pu-238, Pu-239, Pu-240, Pu-241 respectively.

INTRODUCTION

A few non-destructive assay techniques were introduced to the Tokai reprocessing plant in 1978 under the agreement between the United States and Japan called "TASTEX" (Tokai Advanced Safeguards Technology Exercise). The k-edge densitometry and the high resolution gamma spectrometry for concentration and isotopic composition analysis of plutonium were included in the TASTEX. The former technique developed at LANL has been implemented and used for routine inspection analysis since 1982 because of its fairly satisfactory results in the field test. Although good agreement between mass spectrometry and HRGS results were observed, it was difficult to properly maintain the LLNL spectrometer system in our environment. The development of a new system has therefore initiated in 1984 as a part of JASPAS. In order to improve and facilitate the measurements, the new system called "New TASTEX System" / "Combined system" consisting of not only high resolution gamma spectrometer but also k-edge densitometer has been developed. The purpose of the new system is to determine both concentration and isotopic composition of plutonium simultaneously for safeguards of the Tokai reprocessing plant.

It was found that thick cell used for the k-edge densitometer was not suitable for isotopic composition measurement from the viewpoint of self absorption of plutonium. Therefore the idea of two different cells was adopted in the system. The system consists of two HP-Ge detectors, two amplifiers, two different electronic modules including ADC and stabiliser and a multi-channel analyser(ND) and multi-user computer(DEC) commonly used. Two programs of this system which were developed at both LANL and LLNL have been combined and installed in the system at TRP. Operators can use both programs simultaneously and independently by using two CRT terminals.

The absorption of the k-edge densitometry in the present application involves the measurement of gamma radiation. The transmission sources for the k-edge densitometer are the radioisotopes, Se-75 and Co-57. The 121.1 keV and 122.1 keV gamma rays emitted by the sources closely bracket the k-edge absorption edge of plutonium at 121.8 keV. The logarithm of the ratio of the measured transmissions at these two gamma-ray energies is directly proportional to the concentration of plutonium.[1] Plutonium isotopic abundances are obtained by analysing the peaks of passive gamma-ray emitted from plutonium solution. The energy region analysed is from 40 keV to 200 keV. The removal of U-237 and Am-241 from samples permits the prominent peaks of Pu-238, Pu-239 and Pu-240 at 43 keV, 51 keV, 45 keV, respectively, to be analysed and interpreted. The 148 keV peak is used to measure the abundance of Pu-241. For aged plutonium solution, several individual peaks of high energy plus the 94-104 keV complex of unresolved gamma and x-ray peaks are analysed because intense 59 keV peak of Am-241 obscures the Pu peaks of low energy.[2] Pu-241 isotopic fraction is evaluated from an isotopic correlation based on Pu-239, Pu-240, Pu-241.[3]

Calibration of the system was carried cut in March 1986. The plutonium so' tion (about 250 g/l) used for the k-edge densitometer calibration was characterised by destructive analyses, controlled potential coulometry, AgO-Fe(II)-Cr(VI) titration and isotopic dilution method after being purified. On the other hand, calibration of high resolution gamma spectrometry was divided into two ways, namely initial calibration consisting of peak shape fitting and efficiency curve acquisition and long term calibration. The former calibration had been done when the program was installed, while correction factors for long term calibration were obtained throughout the campaigns. The results of plutonium concentration and isotopic abundances obtained by the k-edge densitometer and high resolution gamma spectrometer have been compared with those by controlled-potential coulometer and mass spectrometer respectively for the last three years.

EQUIPMENT

1)Detector and Cell Characteristics

The location of sample cells and detectors is shown in Fig. 1.

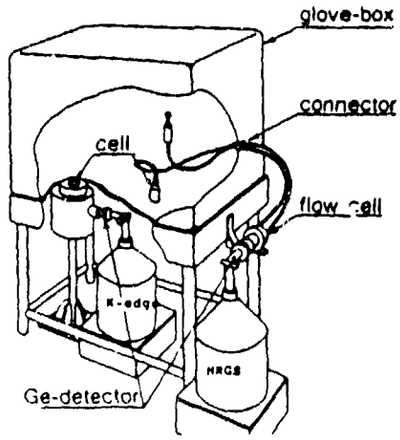


Fig.1 Sample Cell & Detector

[K-edge Densitometer]

The measurement station of k-edge densitometer consists of an intrinsic planar germanium detector(200 mm² by 7 mm) and a mechanical system. The Se-75 and Co-57 transmission sources, mounted in separate positions on a wheel, are rotated into the measurement position by a motor-driven Geneva mechanism. Collimators positioned between sample and the detector are rotated synchronously into the transmission path. For the passive measurement, the sources are rotated out of the measurement position and shielded from the detector, and the collimation is enlarged. The entire mechanical system is automated and under computer control. A Cd-109 source mounted on the detector is used to correct losses of events resulting from changing count rates.

The signals from the germanium detector preamplifier are shaped, amplified, and digitised by NIM modules in the electronics rack. Pileup pulses are electronically eliminated from the spectrum. A two-point digital stabiliser, under computer control, maintains a constant energy calibration.

Solution samples contained in vials are introduced into the glovebox through pneumatic transport tubes following removal of the solution from the process. The solutions are transferred into disposable plastic measurement vials that are inserted into the well for assay. This disposable vial has the parallel side walls which can reduce the errors due to the positioning of sample vials.

[HRGS]

The detector is of a planar design with approximate dimensions of 200 mm² by 12 to 15 mm deep.

The signals from germanium detector/ preamplifier are processed without serious degradation of the resolution by controlling input count rates of up to 15000 cps. A pulses pile-up rejector is used to reduce the effects of chance coincidence.

The principal requirement of the analysis codes is that the sample be confined in a disk-shaped geometry such that the projected surface density of the plutonium in the solution does not exceed approximately 0.02 g/cm². The flow-cell adopted here has 2.5 cm² effective area and 0.1 cm thickness. The sample container and detector are closely coupled (about 1 cm) to obtain the best counting efficiency. The analysis code used for freshly separated plutonium solution measures peak areas in the 43-51 keV energy region; therefore the amount of absorbing material must be minimized. The presence of an intense 59 keV gamma-ray in aged plutonium samples due to the in-growth of Am-241 requires the use of absorbers. Cadmium absorber around 0.1 cm is used not only to attenuate the gamma ray that directly incident upon the detector, but also to reduce the detection of Compton-scattered radiation.

2)Spectrometer

The system consists of Nuclear Data 66 MCA with two analog-to-digital converters. The MCA is controlled by a Digital Equipment Corporation (DEC)PDP-11/23 plus computer with 256 k bytes of memory. The software is RSX-11M from DEC. The system has two hard disk drivers (RL02) and two floppy disk drivers (RX02) for data and program storage. The time sharing nature of the software allows both the densitometer and the isotopic software to control the MCA simultaneously and to operate independently each other. Electronics and computer terminals of the system are shown in Fig.2 . Fig. 3 describes the block diagram of the system.


Fig.2 Electronics & CPU

MEASUREMENT METHODS AND SOFTWARE

[k-edge Densitometer]

K-edge densitometry is based on the transmission properties of gamma radiation around the absorption edge. As the gamma ray energy is increased the transmission of the gamma ray through a material slowly increases until the binding energy of electron in the material is reached. Then the transmission suddenly decreases, because the another electrons can absorb the incoming gamma radiation. At 121.9 keV, the plutonium k-edge energy, all other materials exhibit nearly constant transmission properties as the energy of the gamma radiation is varied. Consequently the

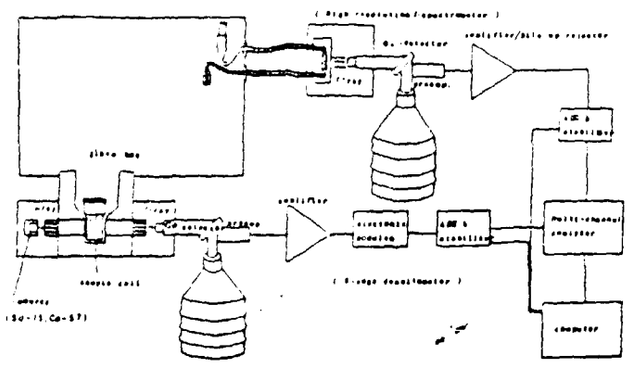


Fig.1 Block diagram of the PASTEX system

ratio of transmissions just above and just below 121.8 keV is propotional to the plutonium concentration and independent of all other materials. The k-edge densitometer uses the measured transmission ratio,

$$R = T(122) / T(121),$$

where T(122) = Transmission of Co-57 (122.1 keV) and T(121) = Transmission of Se-75 (121.1 keV). To measure a transmission, it is first necessary to get a straight through value. Namely, the Se-75 and Co-57 measurement control spectra are acquired to obtain the unattenuated intensities (net peak areas in the absence of sample attenuation) of the gamma-ray lines required for transmission measurements. Cd-109 source, 88 keV peak, is used as a "clock" to determine the actual collection time that the system has.

Plutonium concentration, C(Pu), determined using the equation;

$$C(Pu) = -\ln R/k.$$

Constant, k, determined empirically using measured values of R obtained with solutions of known C(Pu) is equivalent to the product, $\Delta\mu X$, where $\Delta\mu$ is the change in the plutonium mass attenuation coefficient between the two energies (121.1 and 122.1 keV) and X is the sample thickness in the transmission path. The sample thickness for the cell used in the system is approximately 2 cm. The value of $\Delta\mu X$ at the k-edge of plutonium is 3.4 cm^2/g. Therefore, the expected value of k for these measurements should be approximately 6.8 cm^2/g. A detector relative efficiency check is performed using the net peak areas of 121.1 and 136.0 keV (Se-75), in addition to the resolution and gain checks applied to the Se-75 and Co-57 measurement control spectra when straight through measurement is carried out. The ratio of these two peak areas should remain constant for a given Se-75 source if the relative detection efficiency remains unchanged. The new k-edge densitometer includes additional correction on plutonium determination which can minimize the effect of matrix based on the energy difference between 121.1 and 122.1 keV. In addition to the transmission at 121.1 and 122.1 keV, the measured transmission at 136.0 keV from the Se-75 source is also available with no additional measurement. The technique is , therefore, to extrapolate from 136 and 122 keV to 121.8 keV to find the transmission above the edge. For a short energy range near the k-absorption edge, μ vs E is approximately a straight line in ln-ln space. By assuming the same slope below the k-edge, the extrapolation below the edge can also be performed. The extrapolation is reasonable because of the short distance to be extrapolated (from 121.1 to 121.8 keV). Details are described elsewhere.[4]

The k-edge densitometer software consists of two programs.[5] The main program, DENSIT, controls the MCA, the Geneva source wheel, and the digital stabiliser. The program also performs data analysis and writes (or reads) data to (or from) the disk. In addition to these operations, the program performs the measurement control function by promoting operators to perform bias or precision checks on a scheduled basis. In this program, net peak areas are determined by subtraction of straight-line backgrounds obtained from two regions of interest that bracket the peak region. The peak region is, typically, three times the peak FWHM. The background regions are, typically, one-third the peak region width. A formal description of the methods used for peak area determination is given elsewhere.[5]

Following the acquisition of each spectrum, the energy resolution and gain are evaluated from the data. A reference peak FWHM and centroid are determined and compared with limits specified by the program for these peaks.

The DENSIT program uses a set of parameters, and this set is prepared and modified by the ancillary program, CDENSIT.

Program options of the k-edge densitometer measurement included in DENSIT are divided into operators options and supervisor options. The former options are the routine operations used in the course of normal data acquisition and analysis. The latter options are used only by personnel who are fully knowledgeable in the operation of the programs.

Several improvements have been made in the upgrade in addition to the extrapolation shown above. Operator interaction in the revised software is much more intuitive and user friendly.

[HRGS]

The spectral data acquired by using intrinsic germanium detector are analysed and interpreted by the program[6] which is divided into two parts. One analyses spectra taken of freshly separated solutions, and the other spectra of aged solutions. The program called LEPA is used to analyse spectra of recently processed solutions.[7] Because such solutions are relatively free of Am-241 and U-237, the spectral features associated with these isotopes are largely absent, permitting other features to be more easily detected. This is particularly true of the 59-keV gamma ray that is principally due to Am-241. Its absence allows gamma rays of lower energy to be clearly visible in a spectrum. The program LEPA takes advantage of this condition and measures the intensities of three low-energy gamma rays, at 43, 45, and 51 keV, from which it calculates abundances for the plutonium isotopes 238, 240 and 239 respectively. The Pu-241 abundance is calculated from the 148 keV peak intensity. A confirmatory value for Pu-241 can be calculated from the

intensity of the 94 keV X-ray peak. The 129 keV peak is also used as an additional measure of the abundance of Pu-239. The Pu-242 isotope in the plutonium solutions can not be detected by gamma ray spectrometry. However, its abundance is usually less than 5% in first-cycle fuel, and therefore a highly accurate measure of its abundance is not required. The correlation between the abundance of Pu-242 and the abundances of Pu-239, Pu-240 and Pu-241 is given by the relationship[8]

$$[\text{Pu-242}] = k\,[240][241]/[239]^2,$$

where [241] is decay-corrected to the reactor discharge time. In addition to reading in appropriate portions of the spectrum to be analysed, the program also reads two additional files used for the analysis. The first is a file containing peak shape constants generated by the PKSNAL program. These constants are used to describe the peak shapes as a function of energy. The second file contains entries defining detector efficiencies for the six peaks that will be analysed, sample cell parameters, bias correction factors and other constants. Each entry is automatically generated when the calibration program LECAL is executed.

The program called PUAN is used to analyse the spectra of aged solutions. Such solutions generally exhibit an intense peak at 59 keV due to Am-241. The Compton distribution from this peak usually obscures all peaks of low energy. Also since the intensity of this peak tends to dominate the total counting rate, a cadmium absorber is used to reduce its intensity. The presence of U-237 in aged solutions is also easily observable. This activity comes into equilibrium with its Pu-241 parent after about eight weeks. In its nomal mode of operation, PUAN assumes that this equilibrium is in effect. However, an internal check is made, and if the solution is suspected or determined to be out of equilibrium, a message is given and appropriate adjustments are automatically made in the program. The PUAN program uses more constants than does LEPA. A file is generated automatically when the calibration program PUCAL is run that contains entries corresponding to the peak energies, the efficiency coefficients for these peaks, and a bias correction factor for each isotope.

The programs mentioned above are menu driven, and the menu options as shown in Fig. 4 are similar to that of the k-edge system as much as it is practical.

Fig. 4 The List of Program Options

OPTIONS of K-edge

OPERATOR OPTIONS

A - Assay AU - Autocycle
SB - St thru - Backup
MP - Measurement control - Bias
MP - Measurement control - Precision

SUPERVISOR OPTIONS

AD - Assay from Disk AUD - Autocycle from Disk
SBD - St thru - Backup from Disk
C - Calibration CD - Calib from Disk
D - Default
LA - List Assay log LM - List MC log
LR - List examined Results file
CHO - change OUtput listing device
R - Read data from disk W - Write data to disk
X - Exit from program

OPTIONS of HRGS

OPERATOR OPTIONS

A - Assay
MB - Measurement control - Co133 Source
MP - Measurement control - precision

SUPERVISOR OPTIONS

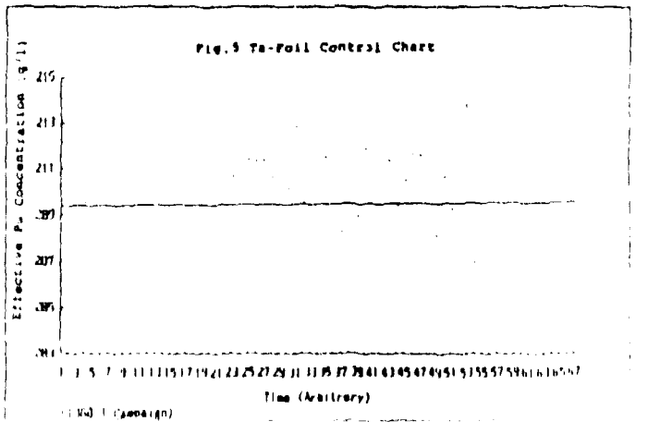
AD - From disk
AU - Autocycle
AUD- Autocycle from disk
C - Calibration mode
D - Default settings
LA - List assay log
LM - List measurement control log
CHO - Change output listing device
ST - Report System Status
R - Read data into analyzer from disk
W - Write data from analyzer to disk
X - Exit from program

EXPERIMENTAL RESULTS AND DISCUSSION

[K-edge Densitometer]

1. Measurement Control

Tantalum reference foils are used to check the total performance of the k-edge densitometer. The combination of two non-active tantalum foils which have different thickness can simulate absorption edge. Fig. 5 is the results of the tantalum measurement control obtained in 1988-1 campaign of the Tokai reprocessing plant. The effective concentration in g/l is plotted in order of measurements performed throughout the campaign. Each solid point is the result of the single measurement carried out prior to every plutonium assay. Clock count times for the Se-75 and Co-57 measurements were 300 seconds (each). The average of the results is 209.3 effective plutonium g/l, which is slightly higher than the data expected from the standard value determined on the existing system. However, the standard deviation of all the results is 0.74% which shows reasonable performance as compared with that of routine system. This figure may show the possibility of short term systematic bias. For instance, the result has been slightly decreasing vs time from the point No.30, in which the isotopic sources were renewed. This might be due to the difference of both count rates.



2. Plutonium Concentration Measurement

The k-edge assay results for both freshly separated plutonium and aged plutonium are discussed relative to the results of destructive analysis (DA) and those of the existing system (called OLD system) routinely used. Clock count times for the plutonium assay were the same as those for the measurement control described above, while 1800 second count period (each for Se, Co, BG) was used for the straight through measurement. As DA method for the plutonium concentration determination, the controlled potential coulometry has been adopted, that can measure plutonium concentration very accurately within 0.1% without any interference.[8]

Table 1 and 2 show the average ratios of k-edge densitometer result / DA result and the standard deviations of them obtained in the campaign of 1985-2, 1986-1, 1987-1 and 1988-1. Differences between the results of the k-edge densitometer (here called "New" k-edge meter) and those of DA obtained in 1985-2 plus 1986-1, and 1988-1 campaigns for freshly separated plutonium are plotted in Fig.6 and 7 respectively. The plotted values are obtained from the average of 3-cycle measurements. Difference between the results of the k-edge densitometer ("New" meter) and those of existing k-edge densitometer (here called "Old" k-edge meter) are shown in the same figures. The same comparisons between "New" k-edge meter and DA, and between "New" and "Old" k-edge meters have been carried out on aged plutonium measurement. The results are shown in Fig. 8. It is noted on freshly separated plutonium measurement that "New" k-edge meter has shown better performance than that of "Old" k-edge meter over last three years. Specially mean biases of "New" system calculated from each campaign result show less biases than those of "Old" system.

Table 1. K-edge Densitometer Measurement Results : Fresh Samples

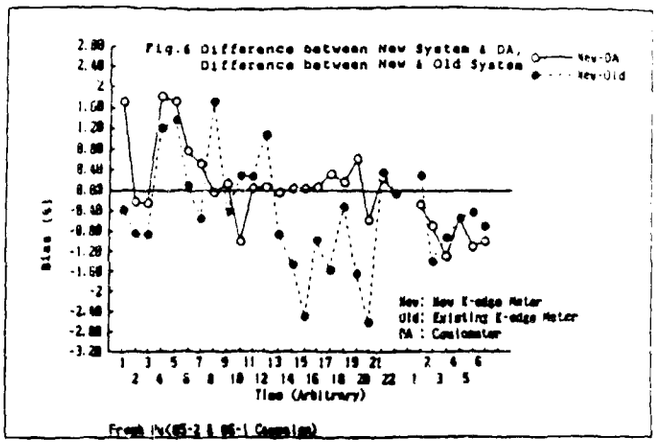
Campaign Name	1985 - 2 Campaign	1986 - 1 Campaign	1988 - 1 Campaign
Number of Samples	22 Samples	6 Samples	28 Samples
Av. of Ratio (K/C)* (Rel. Difference)	1.0028 (+0.28 %)	0.9919 (-0.81 %)	1.0027 (+0.27 %)
C V %	0.71 %	0.38 %	0.61 %

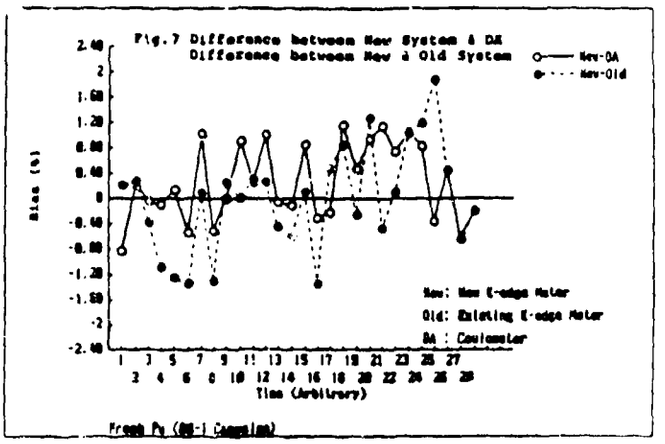
* Ratio of K-edge Densitometry / Coulometry

Table 2. K-edge Densitometer Measurement Results : Aged Samples

Sampling Point	Pu-Storage (1987)	Pu-Storage (1988)
Number of Samples	13 Samples	8 Samples
Av. of Ratio (K/C)* (Rel. Difference)	0.9978 (-0.22 %)	0.9999 (-0.01 %)
C V %	1.08 %	0.70 %

* Ratio of K-edge Densitometry / Coulometry

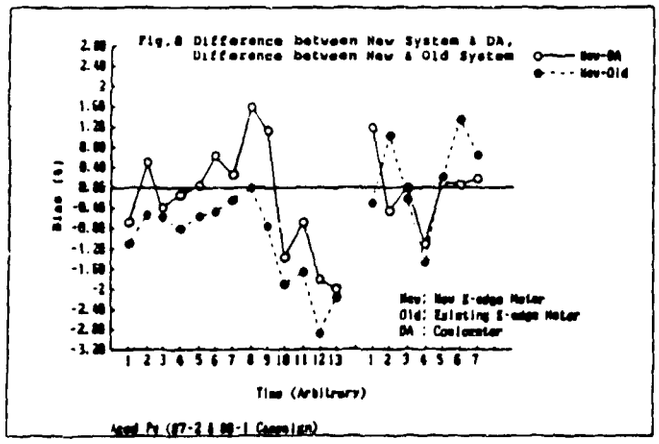




Looking at the performance of both systems, each k-edge meter shows its own systematic errors, which should agree with the tendency of tantalum measurements. This sort of systematic bias is supposed to be difficult to correct in-situ measurement. Comparing the results of "New" system with those of DA , the precision of the "New" system on the mean value of three repetitions can be estimated approximately 0.7%. On the other hand, repeatability of the system was approximately 0.4% obtained from 14 repeated measurements. Differences the of above two estimations can result from the systematic error. With respect to the aged plutonium measurements, "Old" system used to show large systematic bias which seemed to be due to increase of count rate arising from generated Am-241. However, Fig. 8 gives no outstanding bias on the results of "New" system in spice of the fact that measurement precision of "New" system is estimated approximately 1.0% which is worse than that obtained in the case of freshly separated plutonium. Americium effect on plutonium concentration measurement with the system was therefore examined. Table 3 shows the results of the americium test. Here four different concentrations of americium were examined, while every plutonium concentration of the sample was same. The results show that the precision of the plutonium measurement got worse with increasing americium concentration, although no significant error was found. This phenomenon may be due to the increase of count rates.

Table 3 Americium Effect on Plutonium Measurement

Measurement Number	Am-241 Concentration (g/l)			
	0 (no Am)	0.3	0.9	1.5
1	100.60	101.53	104.33	105.96
2	101.29	102.18	100.78	101.20
3	101.29	98.57	100.79	101.84
4	101.41	100.87	98.89	99.34
5	100.22	100.65	99.64	97.67
6	–	98.63	101.58	101.80
7	–	101.37	98.89	98.50
8	–	102.41	100.46	100.16
Av.	100.96	100.78	100.67	100.81
S.D.(%)	0.52	1.48	1.75	2.56



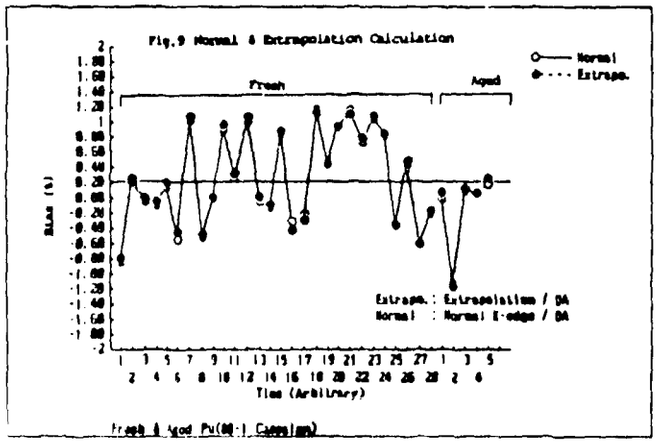
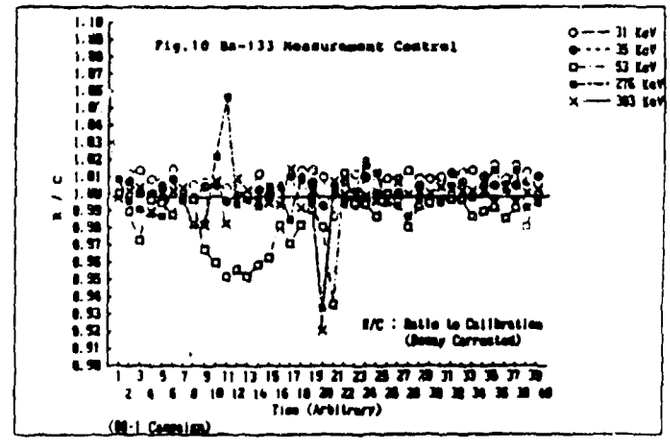


Fig.9 gives the comparison between normal calculation and extrapolation calculation on "New" system. Each plotted point is the mean of three cycle measurements. No significant difference can be observed because the plutonium treated here did not contain large amount of impurities. It could be effective on the samples which contain large amount of other elements such as uranium.

[HRGS]

1. Measurement Control

Ba-133 source is used to monitor the ratio of counting efficiency of low-energy gamma rays relative to higher-energy ones as a function of time. Namely this measurement can verify the stability of the shape of relative efficiency curve. The peak regions used are at 31 keV (k-alpha X rays), 35 keV (k-beta X rays),53 keV, 79-81 keV doublet, 276 keV, and 303 keV. The code determines the ratio of the various peak intensities relative to the 79-81 keV intensity and then compares these values with the corresponding intensity ratios determined at the time the system was calibrated and put into operation. Any significant differences are an indication that the shape of the efficiency curve is changing. Clock times for Ba-133 were 1800 seconds. Fig.10 shows the control status of the HRGS obtained over the 88-1 campaign, where a point(No. 20) gives the possibility of change of detection efficiency on high energy relative to low energy, because both peaks of 276 keV and 303 keV show significant differences compared with the calibration data. Fig.10 ,however, suggests that the system status immediately recovered. A recalibration may be necessary when this phenomenon is detected repeatedly. Some points between No.9 and No. 21 on 53 keV give significant differences despite the fact that other close energy peaks such as 31 keV and 35 keV do not show any differences. These are therefore not supposed to indicate the shape change of the efficiency curve.

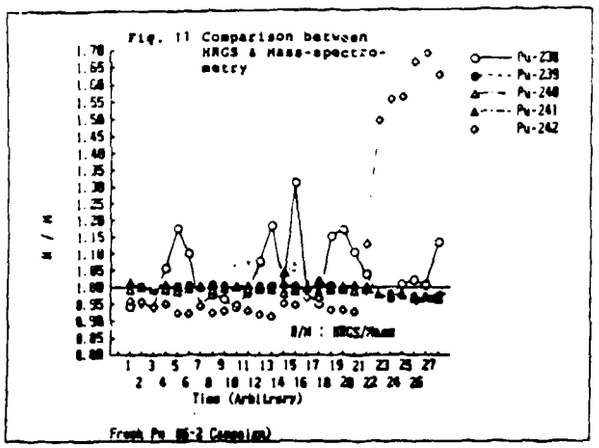


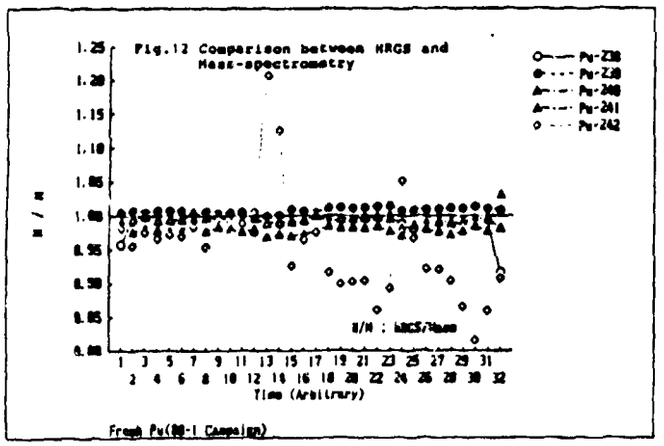
2. Plutonium Isotopic Abundances Measurement [Freshly Processed Plutonium]

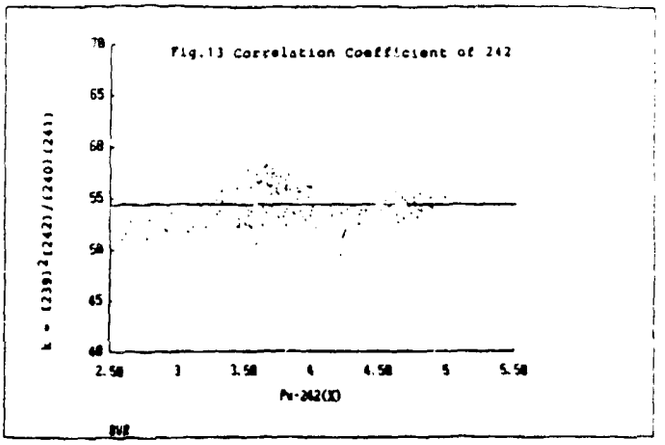
Table 4 gives the summary of the 1988-1 campaign results; the means and standard deviation of the ratios of HRGS / mass spectrometer. Fig.11 and Fig.12 show the same ratios on each isotopic abundance obtained in the campaign of 1986-2 and 1988-1 respectively. Clock count times for one determination by ASSAY program were 1800 seconds.

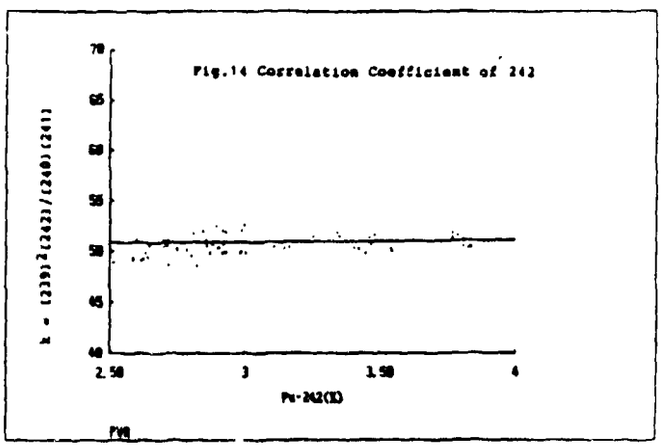
Table 4. HRGS Isotopic Measurement Results ; Fresh Samples

Campaign Name	Ratio (HRGS / Mass Spectrometry)					
	Pu-Isotope	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
1988 - 1 Campaign	Average (Rel.Diff.)	0.9808 (-0.86 %)	1.0082 (+0.83 %)	0.9897 (-0.69 %)	0.9814 (-1.88 %)	0.9548 (-4.52 %)
(37 Samples)	C V %	1.80 %	0.41 %	0.38 %	1.10 %	7.98 %









In 1986-2 campaign, the bias factors estimated from the results of previous campaign (1986-1 campaign) were used. The results of HRGS on Pu-239, Pu-240 and Pu-241 are in good agreement with those of mass spectrometry, while large scatter was observed on the results of Pu-238. The results on Pu-238 in Fig.11 are larger than expected. That is probably due to the accuracy of the alpha-spectrometry technique used to determine Pu-238 at the time. With respect to Pu-242, the data obtained between No.1 to No.21 give negative bias, whereas the data after No.22 show large positive bias. The former may be due to the fact that the spent fuels processed here were BWR type, which can cause negative bias, because the coefficient "53" used for the calculation of Pu-242 was approximately 2.5% higher than the coefficient obtained by our measurements. The coefficients for BWR and PWR are estimated in Fig.13 and 14 respectively, where the correlations were obtained from the measured results on mass spectrometry and cooling time information. The mean and standard deviation (%) of the correlation coefficient on BWR type fuel are 54.3 and 3.3 respectively, while those on PWR type fuel are 50.9 and 1.8 respectively. Pu-242 calculation is therefore expected to be improved by using separate coefficients. In the latter case (after No. 22 in Fig.11), the fuels processed at the plant were ATR(Advanced Thermal Reactor) type, where initial fuel includes plutonium. We therefore suggest that the Pu-242 isotopic correlation be modified for the thermal reactor fuels.

Prior to the 1988-1 campaign, a calibration was performed using a well characterised sample because of long interval in 1987. Every bias factor is therefore unity. Good precisions have been obtained throughout the campaign except the results of Pu-242. The fuels processed here are PWR type (No.1 to No.13 in Fig.12) and BWR type (No.14 to No.32). Pu-242 plotted data are about 5% lower than previous data (given in Fig.11) all over the campaign. On the other hand, Pu-240 and Pu-241 results have a few percent negative bias (approx. 1% and 2% respectively), while Pu-239 results show slightly positive bias (approx. 1%). Reviewing the equation of Pu-242 correlation;

$$[242]=k[240][241]/[239]^3 ,$$

the difference of Pu-242 bias trend between both campaigns shown above can result from the slight biases of Pu-240, Pu-241 and Pu-239, namely $0.99 \times 0.98k / 1.01^3 = 0.95k$. Biases on Pu-240, Pu-241 and Pu-239 are suspected to be due to the errors arising from short term calibration. This suggests that longer study of bias corrections is important. A few outlying points have been detected in Fig.12. These correspond to the changing of fuel types. Repeatability has been evaluated by repeating sample measurement 10 times. Table 5 gives the results obtained from 18 different samples, where each line (each sample result) shows standard deviation (%) of 10 repeated measurements. The values shown at the bottom of the table are the

arithmetic mean of 18 data. In regard to Pu-239, 240 and 241, the estimated repeatability is nearly in accord with the relative standard deviation of 1988-1 campaign shown in Table 4.

[Aged Plutonium]

The average and the standard deviation of the ratios of HRGS / mass-spectrometry evaluated in the 1988-1 campaign are shown in Table 6. Same

Table 5 Average of S.D.(%) Values (Each value is calculated from 10 repeated measurements)

No.	Pu-238	Pu-239	Pu-240	Pu-241
1	0.183	0.222	0.334	0.581
2	0.251	0.424	0.477	1.045
3	0.814	0.550	1.114	2.756
4	0.087	0.224	0.278	0.338
5	0.482	0.441	0.515	1.333
6	0.183	0.318	0.368	1.049
7	0.186	0.173	0.211	0.257
8	0.158	0.298	0.189	0.637
9	0.412	0.335	0.444	1.38.
10	0.154	0.308	0.368	0.658
11	0.207	0.248	0.214	0.448
12	0.180	0.332	0.244	0.852
13	0.114	0.413	0.308	0.695
14	0.108	0.393	0.309	0.695
15	0.784	0.547	0.843	2.138
16	0.274	0.404	0.417	0.847
17	0.287	0.312	0.322	1.313
18	0.332	0.347	0.418	1.580
Ave	0.388	0.381	0.477	1.148

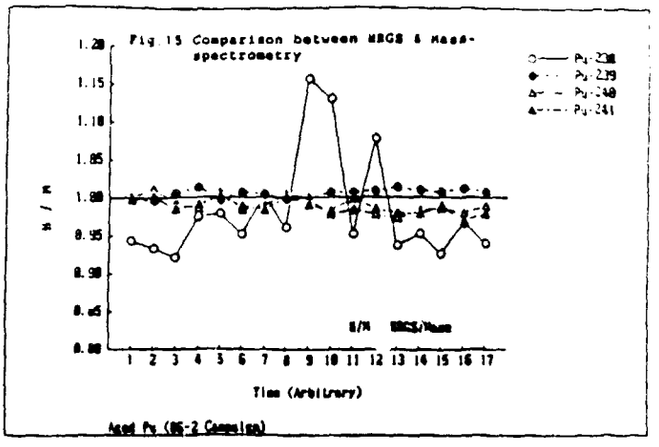
Table 6 HRGS Isotopic Measurement Results : Aged Samples

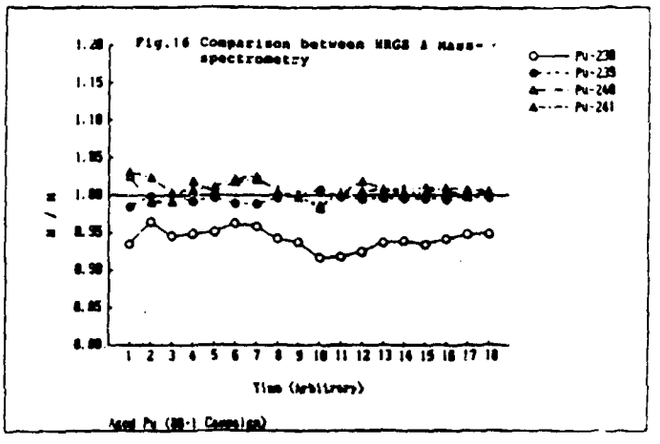
Campaign Name	Ratio (HRGS / Mass Spectrometry)				
	Pu-Isotope	Pu-238	Pu-239	Pu-240	Pu-241
1988 - 1 Campaign	Average (Rel.Diff.)	0.9427 (+5.73 %)	0.9968 (-0.32 %)	1.0087 (+0.87 %)	1.0105 (-1.05 %)
(18 Samples)	C V %	1.43 %	0.52 %	1.08 %	1.08 %

ratios (HRGS vs mass spectrometry) obtained in the campaigns of 1986-2 and 1988-1 are plotted in Fig.15 and Fig.16 respectively. Clock count times for one determination on aged plutonium measurement were 3600 seconds. Fairly good agreements are observed except on the result of Pu-238. Pu-238 value on Fig.15 (1986-2 campaign) shows large scatter/bias. That seems due to uncertainty of the alpha-spectrometry data like the Pu-238 scatter previously shown in Fig. 11. As for Pu-242, it is nearly impossible to calculate the cooling time for Pu-241 correction because of the mixture of plutonium products. Here, Pu-242 values determined by mass spectrometer were therefore used for the isotopic composition calculation. Repeatability on the aged plutonium measurement has been also evaluated by the same way as that performed for freshly processed plutonium. Table 7 shows the results obtained from 8 samples. The value given at the bottom of the table is the arithmetic mean of 8 data. Comparing the precisions obtained in the table with those estimated in 1988-1 campaign(Table 6), slight differences are observed. Those can be errors based upon the mass spectrometry uncertainty.

Table 7 Average of S.D.(%) Values (Each value is calculated from 10 repeated measurements.)

No.	Pu-238	Pu-239	Pu-240	Pu-241
1	0.538	0.184	0.423	0.478
2	0.662	0.381	1.020	0.880
3	0.884	0.241	0.847	0.871
4	0.880	0.381	0.889	0.870
5	0.804	0.378	0.971	0.841
6	0.858	0.477	1.214	0.711
7	0.812	0.427	1.118	0.637
8	0.897	0.345	0.788	0.815
AVE	0.777	0.349	0.878	0.710





CONCLUSIONS

The field test of New TASTEX System (Combined System) consisting of k-edge densitometer and high resolution gamma spectrometer has been carried out for three years. Measurement performance has been improved at the last campaign as compared with that of initial campaign.

Precisions of plutonium concentration determination with the k-edge densitometer were estimated approximately 0.7% and 1.0% for freshly processed plutonium and aged plutonium respectively. It should be noted that short term systematic bias is possibly detected. That sometimes causes larger errors than expected. The precision of plutonium concentration measurement tends to be affected by the presence of americium.

The scatters in the relative differences between HRGS and DA detected on the results of freshly processed plutonium measurements were 1.6%, 0.4%, 0.5%, 1.1% and 8.0% for Pu-238, PU-239, Pu-240, Pu-241 and Pu-242 respectively. Those on aged plutonium were 1.4%, 0.5%, 1.1%, and 1.1% for Pu-238, Pu-239, Pu-240 and Pu-241 respectively. With regard to Pu-242 determination, following points should be noted.

1) It is expected that the Pu-242 determination can be improved by discriminating between the correlation coefficients of BWR and PWR.
2) Isotopic correlation for Pu-242 estimation can be effective especially in the case fuel type is not changed throughout the campaign. In contrast, plutonium product samples corresponding to the transition of fuel types can give outstanding error.
3) The correlation can neither be applied to the fuel containing plutonium initially such as ATR

fuel, nor plutonium storage of reprocessing plant.

It does not seem to be necessary that calibration for k-edge densitometer be repeated periodically as far as Ta-foil measurement control provides good performance. Instead, just when series of Ta-foil measurements give biased results, recalibration of the system should be performed. With respect to HRGS, Ba check source can give similar information. However, precise measurement requires long term bias correction, namely it is desired that bias correction factor for each isotope be evaluated from previous campaign results. Calibration for HRGS should be performed in the case either Ba check source shows significant bias or long term bias correction factors turn to large value.

NDA system such as New TASTEX System can easily give results when system is in good control status. However this also means that even NDA system must be always checked and controlled by the operator who is fully knowledgeable about the system according to our years' experience of in-situ NDA measurement.

REFERENCES

1)P.A.Russo, ..Asakura et al. LA-9440-MS (PNCT 841-82-10), Aug. 1982
2)R.Gunnink, UCRL-80464, 1978
3)R.Gunnink, UCRL-84096, 1980
4)J.K.Sprinkle, S.-T.Hsue, K.Junck LA-10725-M (ISPO-246), Jan. 1987
5)T.R.Canada, S.-T.Hsue, et al. Nucl. Mater. Manage. VI, No.3 702-710, 1977
6)R.Gunnink, W.D.Ruhter M-169 (ISPO-247) Sep. 1985
7)R.Gunnink, UCRL-80464, 1978
8)R.Gunnink, UCRL-84096, 1980
9)Y.Kuno et al. "Use of Automated Controlled Potential Coulometry System for Plutonium Product Accountancy Analysis At Tokai Reprocessing Plant" 29-th Annual Meeting of INMM, 1988