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Title: Improvement of INVS Measurement Uncertainty for Pu and U-Pu Nitrate Solution

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Improvement of INVS Measurement Uncertainty for Pu and U-Pu Nitrate Solution

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Development of Advanced Nondestructive Assay System for Plutonium Solutions

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The photograph of the Plutonium Conversion Development Facility where the INVS is used.

Contents:

Chapter 1 INTRODUCTION

1. INTRODUCTION

In the Tokai Reprocessing Plant (TRP) and the Plutonium Conversion Development Facility (PCDF), a large amount of plutonium nitrate solution which is recovered from light water reactor (LWR) and advanced thermal reactor (ATR), FUGEN are being stored. Since the solution is designated as a direct use material, the periodical inventory verification and flow verification are being conducted by Japan Safeguard Government Office (JSGO) and International Atomic Agency (IAEA).

The INVentory Sample verification systems (INVS) owned by IAEA is categorized for use as a partial defect verification equipment and is used for verification measurement of Pu nitrate solution, U-Pu nitrate solution and MOX powder sampled at PCDF. We studied the accuracy of INVS measurement for Pu nitrate solution in order to confirm the possibility to use it as a bias defect equipment (method E) instead of K-Edge measurement, which is currently in an unstable measurement condition due to aging. In order to achieve the performance which is equivalent to the method E level, the level of better than 1% accuracy is required for INVS. If the measurement uncertainty of within 1% can be improved, it is expected that the usage of INVS could be extended to the operator's own measurements in MC&A and process control to reduce the number of Destructive Analysis (DA) efforts.

From FY12 to FY14, the INVS detector parameters were optimized. The calibration curves for both the passive calibration curve method and known-α method were obtained using standard Pu nitrate solution and U-Pu nitrate solution. The measurement results were evaluated using three different methods (passive calibration curve method, known-α method and multiplicity method). As a conclusion, in the range of concentration of typical samples, we confirmed good correlations between measured doubles counts and Pu-240 effective mass in all three methods. Especially, the conventional calibration curve method showed a best correlation performance. Known-α method showed the second best performance. These methods achieved target uncertainty within 1% when the calibration standards were a good match to the unknown samples. Since it is thought that background singles change may affect uncertainty for each calibration, an additional shielding installation around the INVS was proposed by Los Alamos National Laboratory (LANL) in order to reduce the effect and improve the uncertainty.

In FY15, the new additional shielding was applied to INVS equipment and calibration and measurement were conducted in the same manner as those without the additional shielding. From the results, the shielding improved measurement uncertainty in known-α method. It was also confirmed that the necessary measurement time to achieve target uncertainty of within 1% is less than 1 hour for passive calibration curve method and known-α method.

Details of the activities from FY12 to FY15 are described in the following chapters.

Chapter 2 WITHOUT THE ADDITIONAL SHIELDING

In this chapter, the results of research and development from FY12 to FY14 using the INVS without the additional shielding are discussed.

1. THE INVS SYSTEM

The INVS is a passive neutron counter with ^{3}He proportional detectors which are covered with polyethylene moderator with 31.3% counting efficiency (for Cf) and the specification is shown in Table 1. It is applied for measurement of nuclear materials in a small sample vial (about 50 mm height × 25 mm external diameter in case of using the existing sample holder) to the sample cavity (317 mm height × 250 mm diameter).

Table 1 The INVS system

Detector	JCC-12M (JOMAR SYSTEMS) S/N 89-04001 Gate Width: 64μs HV: 1780V
Shift register	JSR-12 (CANBERRA) for inspection AMSR (ORTEC) for this activity
Software	INCC5.12

The INVS detector is located under the glove box in PCDF at a regular position for safeguards inspection as shown in Figure 1 and Figure 2. Since a pipe in which the sample holder is inserted is connected to underneath the glove box, it is very convenient for operators to conduct the sample measurement without contamination. The procedure for sample setting is shown in Figure 3. For safeguards inspection measurement, a sample in small vial is put in a sample holder of the glovebox. In order to adjust the INVS detector position vertically (center of the sample cavity of the INVS adjusts to the position where the vial is set inside pipe), a jack device is used as shown in Figure 2 and Figure 3. The detector is connected to the shift register and data acquisition PC.

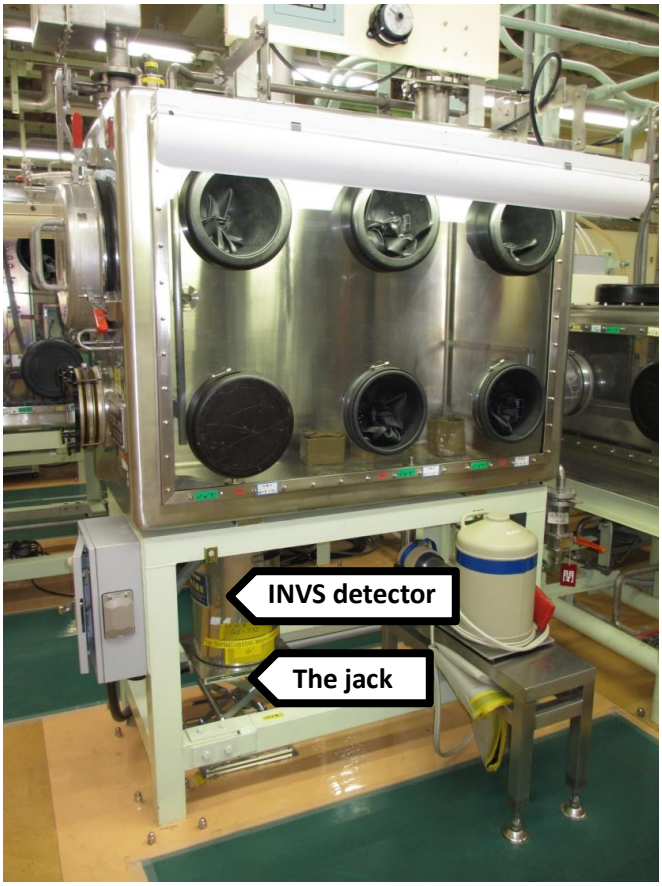

Figure 1 Relationship between INVS

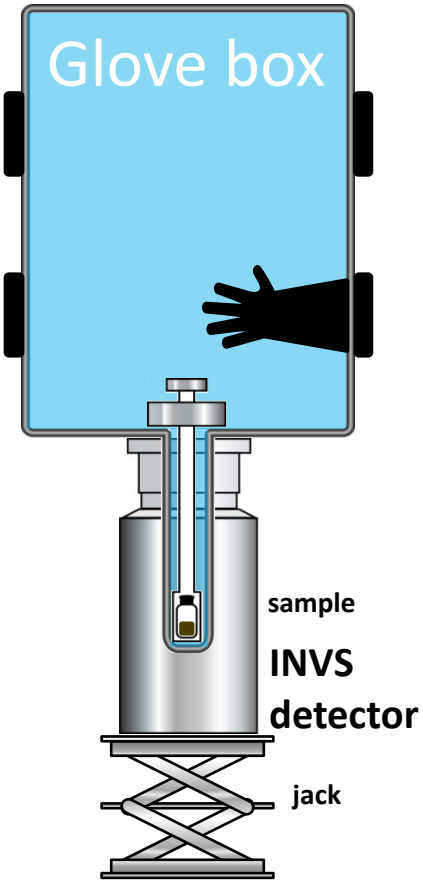

Figure 2 Measurement configuration

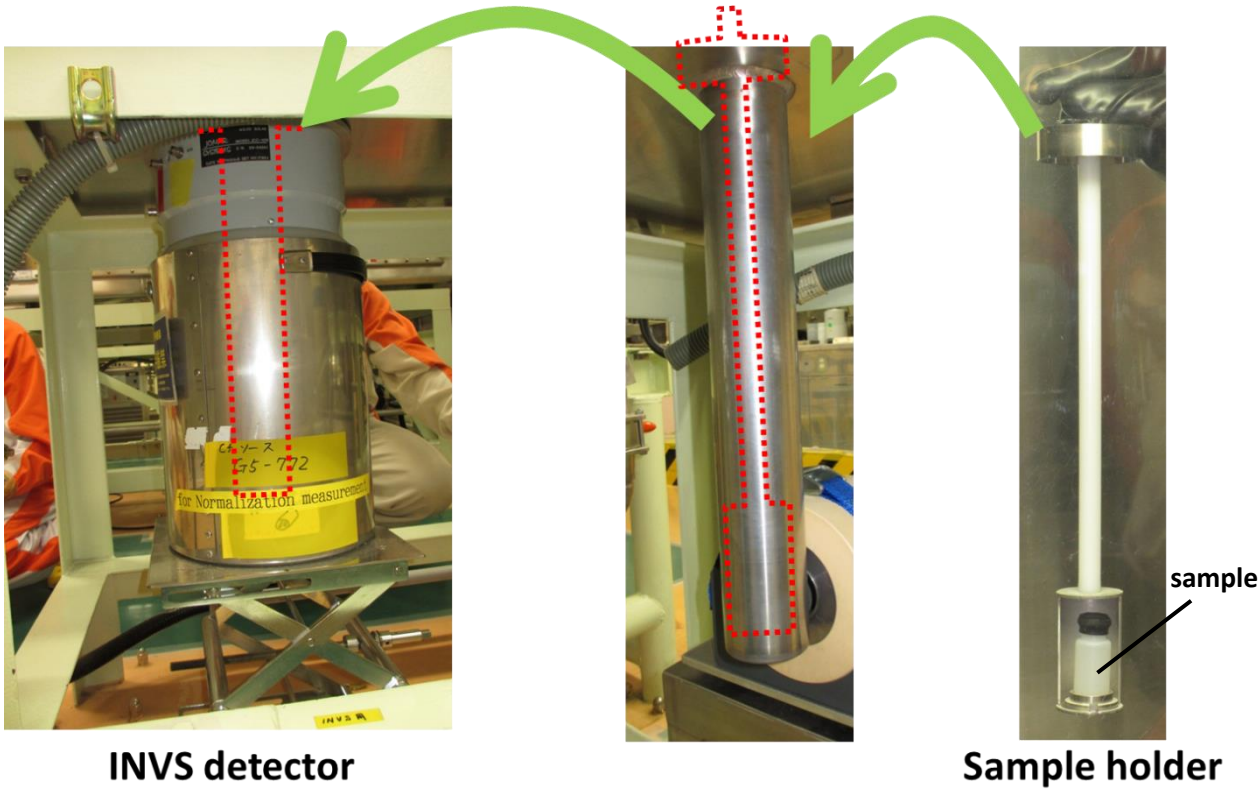

Figure 3 Sample setting procedure

2. CHARACTERIZATION

In order to achieve high accuracy (within 1%), we investigated whether the sample position for inspection and default detector parameters are optimal or not using ^{252}Cf source or MOX pellet source.

2.1. Response profile and sample setting position

The response profile for vertical and radius in the sample cavity was confirmed using stand-alone INVS with ^{252}Cf source. The results are shown in Figure 4. In the vertical profile, from 5 cm to 8 cm region shows a flat profile. On the other hand, in the radius profile, it was confirmed that 0 cm, which is the center, was the lowest and the change is within 1% (no effects for uncertainty). As a result, the default sample setting position at 7 cm for vertical and 0 cm for radius, which is inside the flat region, is the optimal and the sample holder can support reproducibility.

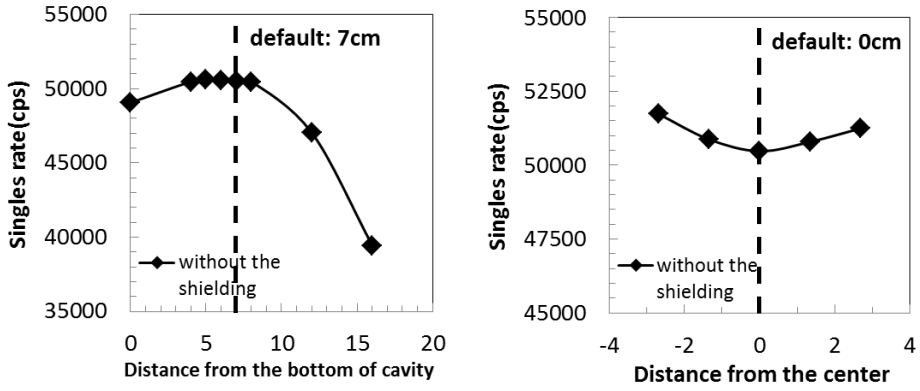

Figure 4 Response profiles (left: vertical; right: radius)

2.2. Parameters tests using ^{252}Cf source

2.2.1. High Voltage plateau check

In order to confirm the optimal detector parameters, a parameters tests using calibrated ^{252}Cf source was conducted. The singles counts were measured with various high voltages from 1400 to 1900 V. The result is shown in Figure 5. It was confirmed that the optimal high voltage is 1780 V which is on the plateau region as default setting.

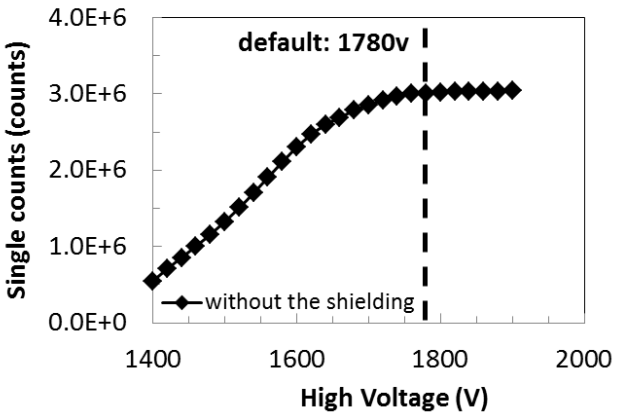

Figure 5 High voltage curve

2.2.2. Die-away time and gate width tests

In order to confirm die-away time (τ), doubles rate (D) of a ^{252}Cf source, it was measured with six different gate setting (8, 16, 32, 64, 128, 256 μs). The results were fitted to a single exponential curve equation (1)[1] using the Deming curve fitting program as shown in Figure 6 and obtained τ =44.1 μs. The gate width (G) is calculated and G value is ≈ 55.37 μs using equation (2)[2]. Thus, it was confirmed that the default gate setting (64 μs) was suitable because the uncertainty as a function of gate length has a very flat minimum and a larger gates are generally beneficial at low count rates.

$$D = a \times \left\{1 - e^{-\frac{G}{\tau}}\right\} \quad (1)$$

$$G \approx \tau \times 1.257 \quad (2)$$

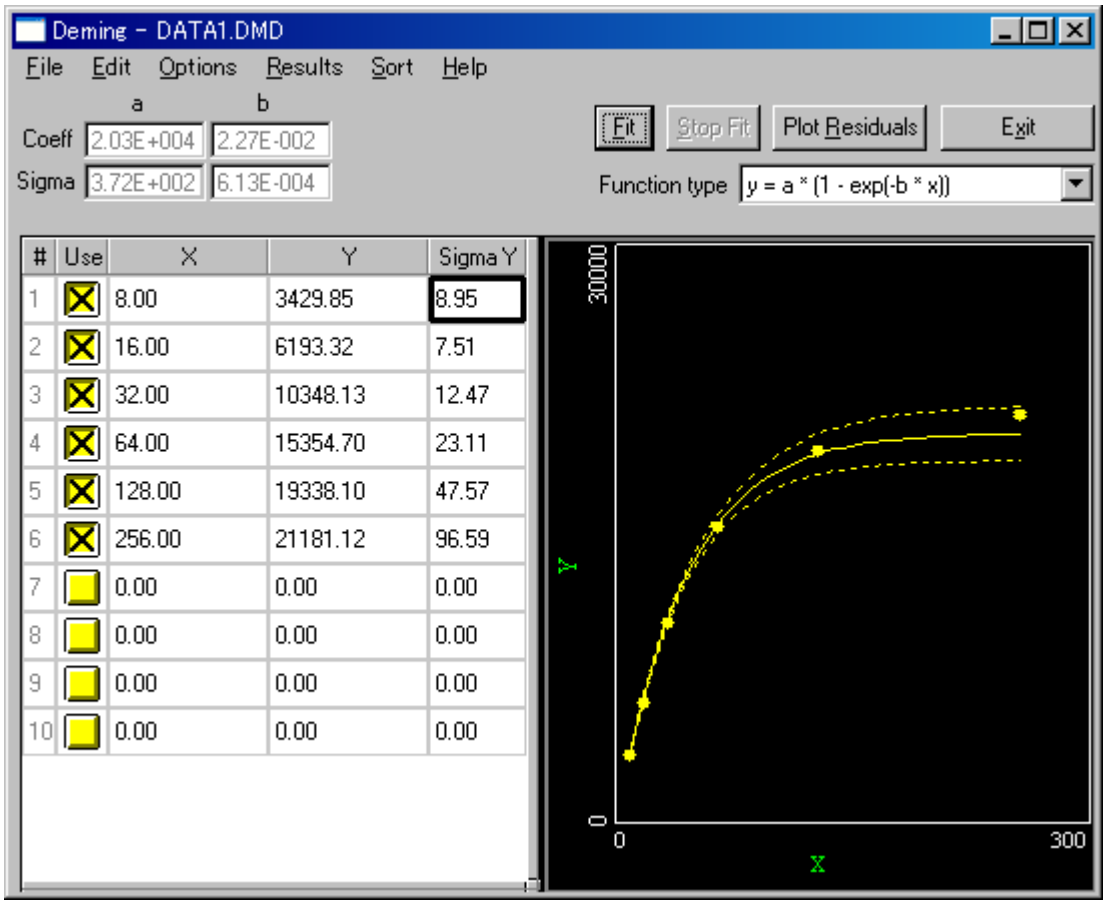

Figure 6 Fitting result using Deming

2.3. Parameters tests using MOX pellet source

Other parameters such as efficiency (ε), doubles gate fraction (f_d), triples gate fraction (f_t) for multiplicity assay and ρ_0 value for known-α assay should be determined using Pu source. In case of using a ^{252}Cf source, it has different energy spectrum and multiplicity distributions from those of Pu and a large decay correction is required due to quite quick decay, and accurate decay correction is very difficult. Thus it was decided to use a MOX pellet as the Pu source because of the stability against moisture absorption and shape change. The MOX pellet source was made from MOX powder with known amount of Pu content, Pu isotopic composition and Am content by DA as shown in Table 2. The

amount of Pu is about 1g, the same as our target samples. The MOX pellet source for parameter tests was fixed in the same glass vial as those for actual solution samples as shown in Figure 7 and was set in the sample position using the conventional sample holder.

Table 2 Specifications of MOX pellet source

U amount	1.0356 g				
U isotopic composition	^{234}U	^{235}U	^{236}U	^{238}U	
	0.066	0.683	0.235	99.016	
Pu amount	0.9985 g				
Pu isotopic composition	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
	0.907	67.302	24.335	3.666	3.790
Am-241 Contents	16800 ppm/Pu				

Figure 7 Picture of MOX pellet and glass vial

The multiplicity assay needs appropriate values of ε, f_d and f_t respectively. The relation between f_d and f_t is shown in the equation (3)[3]. In order to get these values, a parameter survey was conducted using both the Pu amount 0.9985 g (well defined by DA) and estimated α value 0.6756 (calculated using SOURCES-4C code produced by LANL). As a result, we determined the optimal parameters of efficiency ε=31.04%, f_d=0.5562 and f_t=0.3094, respectively as shown in Figure 8.

$$f_t = {f_d}^2 \tag{3}$$

fd	ft = fd^2		0.3098	e=0.31	0.3104	0.3105	0.3131
0.5562	0.3094	M =			1.053	1.053	1.052
		Alpha =			0.674	0.675	0.682
		Pu(g) =			0.999	0.998	0.987
0.558	0.3114	M =					1.052
		Alpha =					0.687
		Pu(g) =					0.985
0.559	0.3125	M =	1.053	1.053			
		Alpha =	0.677	0.678			
		Pu(g) =	1	0.999			
0.56	0.3136	M =	1.053	1.053			
		Alpha =	0.679	0.679			
		Pu(g) =	0.999	0.998			

Target
Pu(g)=0.9985
Alpha=0.6756

Figure 8 Parameter survey for determination of efficiency and gate fractions

On the other hand, known-α calibration needs an appropriate ρ_0 value. The optimal ρ_0 value was also calculated by the equation (4)[1] where ν_{s1} and ν_{S2} are 1st and 2nd factorial moments of the neutron emission multiplicity distribution for the spontaneous fission of ^{240}Pu, and then the optimal ρ_0 value of 0.1518 was obtained. The summary of the optimized detector parameters for INVS is shown in Table 3.

$$\rho_0 = \frac{\nu_{s2}}{2\nu_{s1}} \cdot \varepsilon \cdot f_d \tag{4}$$

Table 3 Final Optimal Detector Parameters for INVS

Parameters	Setting Value	Method
Pre-delay	4.5 μs	default
Gate Width	64 μs	^{252}Cf Measurement
High Voltage	1780 V	^{252}Cf Measurement
Die-away Time	44.1 μs	^{252}Cf Measurement
Dead time Coefficients	a: 1.23 μs, b:0.615 ps	default
Multiplicity Deadtime	308 ns	Calculated from dead time coefficients
Doubles Gate Fraction	0.5562	MOX Pellet Source
Triples Gate Fraction	0.3094	MOX Pellet Source
Efficiency	31.04%	MOX Pellet Source
Rho-Zero	0.1518	MOX Pellet Source

3. SAMPLE PREPARATION

Target samples are Pu nitrate solution and U-Pu mixed nitrate solution (1:1) recovered from LWR spent fuels. In order to evaluate INVS accuracy for these samples, we performed measurements of actual solution sampled from the PCDF. We took samples from P11V11 (Pu nitrate solution) and P12V12 (U-Pu nitrate solution (1:1)) which the analysis results were precisely determined by DA. The specifications of each tank are shown in Table 4.

In order to evaluate uncertainties for typical concentration of vessels in TRP and PCDF, a variety of samples diluted with nitric acid were prepared.

Table 4 Specifications of actual samples

Vessel	P11V12					P12V12				
Chemical Form	$Pu(NO_3)_4$					$UO_2(NO_3)_2$ and $Pu(NO_3)_4$				
U Concentration	-					115.2 g/l				
U Isotopic Composition	-					^{234}U	^{235}U	^{236}U	^{238}U	
						0.068	0.690	0.241	99.001	
Pu Concentration	213.2 g/l					113.2 g/l				
Pu Isotopic Composition	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
	0.892	67.388	24.351	3.606	3.763	0.894	67.478	24.303	3.558	3.767
Am-241 Contents	17300 ppm/Pu					15300 ppm/Pu				
Solution Density	1.440 g/cm^3					1.428 g/cm^3				

4. DETERMINATION OF CALIBRATION CURVE FOR PASSIVE CALIBRATION AND KNOWN-α

The calibration exercises for passive calibration method and known-α method (both methods are coincidence assay) were conducted using the detector parameters (Table3) and the samples listed in Table 4. Samples with typical Pu concentration in PCDF were chosen as calibration standards in order to be focused on usual concentration range of typical inventory. The range of the usual concentration of samples is over 100 g/l for P11V12 and over 50 g/l for P12V12 as shown in Table 5 (Lower concentration samples than usual range are not reasonable to make calibration curve).

These samples were measured for 22 hours (30 s × 2640 cycles). The calibration curve for the passive calibration curve was given in the equation (5)[1] with a relation between ^{240}Pu effective mass (m) and doubles count rates (D) for each sample. On the other hand, the calibration curve for the known-α method was given in equation (6)[1] with a relation between m and multiplication corrected doubles rate (D_c). In the case of known-α calibration, coefficient a_c is theoretically zero. To obtain D_c, we applied and calculated using the equations (7)[1] with a relation among D_c, ρ_0 factor and α value. In addition, we used an equation for α value provided by LANL[4] to obtain the necessary representative α value for the calibration curve.

$$D = a + bm \tag{5}$$

$$D_c = a_c + b_c m \tag{6}$$

$$iM_C^2 + jM_c - r = 0, i = 2.166(1+\alpha_0), j = 1 - i, r = {\rho_m}/{\rho_0} = \frac{(1+\alpha_0)\frac{D}{S}}{\rho_0}, Dc = \frac{D}{rM} \tag{7}$$

Table 5 INVS calibration results (raw data) (upper: P11V12, lower: P12V12)

Item ID	Pu conc. (g/l)	Pu amount (g)	U conc. (g/l)	U amount (g)	Pu-240 Effective(g)	singles (cps)	doubles (cps)	triples (cps)
A	127.76	0.6448	-	-	0.2123	178.501	13.568	1.623
B	148.59	0.7481	-	-	0.2463	204.020	15.788	1.876
C	170.35	0.8578	-	-	0.2824	236.103	18.130	2.109
D	191.77	0.9633	-	-	0.3171	264.188	20.381	2.374
E	213.24	1.0718	-	-	0.3528	294.466	22.678	2.690

Item ID	Pu conc. (g/l)	Pu amount (g)	U conc. (g/l)	U amount (g)	Pu-240 Effective(g)	singles (cps)	doubles (cps)	triples (cps)
F	68.67	0.3433	69.88	0.3494	0.1129	94.419	7.166	0.870
G	79.33	0.3985	80.73	0.4056	0.1310	109.241	8.354	0.981
H	91.11	0.4552	92.72	0.4632	0.1497	123.792	9.556	1.146
I	102.19	0.5095	104.00	0.5185	0.1675	139.210	10.699	1.279
J	113.20	0.5663	115.20	0.5763	0.1862	154.435	11.920	1.445

These calibration results are summarized in Table 6. All calibration curves were graphed in Figure 9 in

order to compare D and D_c, with a strong linear regression line with very high correlation factor.

The calibration curves P11V12 and P12V12 showed similar coefficients, although P11V12 is pure Pu nitrate solution and P12V12 is U-Pu mixed nitrate solution (1:1). It indicates that U has no influence on Pu mass in the passive calibration curve method in the range of U concentration of these samples (~115.2 gU/l to ~113.2 gPu/l).

Table 6 Calibration curves for passive calibration method and known-α method

Method	P11V12	P12V12
passive calibration curve method	$\mathrm{D} = -0.192 + 64.9\mathrm{m}$	$\mathrm{D} = -0.135 + 64.7\mathrm{m}$
known-α method	$D_C = 47.2\mathrm{m}$	$D_C = 47.8\mathrm{m}$

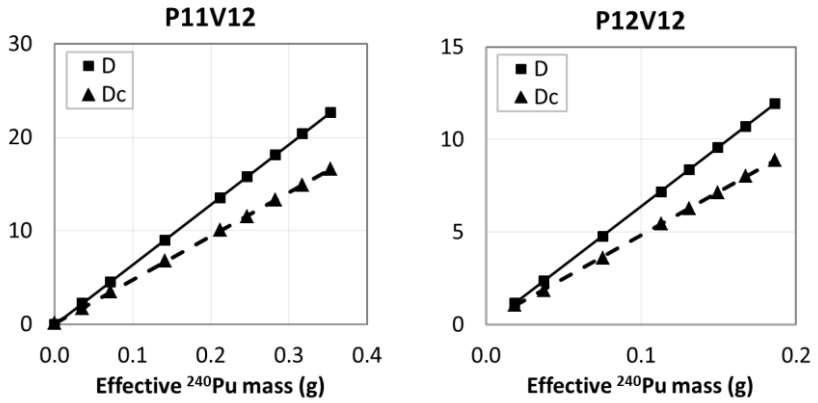

Figure 9 Passive calibration method and Known-α calibration results
(relationship between ^{240}Pu effective and D (passive calibration) or D_c(known-α))

5. COMPARISON OF UNCERTAINTIES OF VARIOUS MEASUREMENT METHODS

In order to choose proper calibration method, 22 hours measurements of the samples from P11V12 and P12V12 (30 s × 2640 cycles) were conducted. The measurement results of actual samples were evaluated by the three different methods (passive calibration curve method, known-α method and multiplicity method).

The samples in the range of reasonable concentration with constant acidity and volume were evaluated using three methods. The results with relative differences for Pu mass between INVS and DA (Dcl-assay(%)) , systematic errors (Sys. err.(%)) and random error (Ran. err.(%)) are shown in Table 7. The systematic error is defined as the difference between the average ratio of assay masses to declared Pu masses. For passive calibration curve and known-α methods, the systematic error is very small because of the calibration procedure. The random error is defined as the root mean square deviation of the relative differences. In order to clarify the achievement of target uncertainty of within 1%, all relative differences are marked with following correlation indexes; ~1%: ◎, 1~2%: ○ and 2%~: △.

Table 7 Comparison of the results using three methods (upper: P11V12, lower: P12V12)

Method	Specification			DA	Passive calibration curve				Known-α				Multiplicity			
Item ID	Pu conc. (g/l)	Acid-ity (N)	Vol. (ml)	Dcl Pu mass (g)	Assay mass (g)	± (g)	Dcl - assay (%)	correlation index	Assay mass (g)	± (g)	Dcl - assay (%)	correlation index	Assay mass (g)	± (g)	Dcl - assay (%)	correlation index
A	127.76	2.9	5.0	0.645	0.644	0.001	0.079	◎	0.652	0.002	-1.056	○	0.639	0.002	0.840	◎
B	148.59	2.9	5.0	0.748	0.748	0.001	-0.065	◎	0.743	0.002	0.698	◎	0.751	0.002	-0.435	◎
C	170.35	2.9	5.0	0.858	0.858	0.001	-0.018	◎	0.860	0.002	-0.301	◎	0.878	0.002	-2.305	△
D	191.77	2.9	5.0	0.963	0.963	0.001	-0.063	◎	0.962	0.002	0.070	◎	0.986	0.003	-2.429	△
E	213.24	2.9	5.0	1.072	1.071	0.001	-0.071	◎	1.069	0.002	0.224	◎	1.080	0.003	-0.739	◎
						Sys. err.(%)	0.050			Sys. err.(%)	-0.053			Sys. err.(%)	-0.978	
						Ran. err.(%)	0.063			Ran. err.(%)	0.656			Ran. err.(%)	1.357	

Method	Specification			DA	Passive calibration curve				Known-α				Multiplicity			
Item ID	Pu conc. (g/l)	Acid-ity (N)	Vol. (ml)	Dcl Pu mass (g)	Assay mass (g)	± (g)	Dcl - assay (%)	correlation index	Assay mass (g)	± (g)	Dcl - assay (%)	correlation index	Assay mass (g)	± (g)	Dcl - assay (%)	correlation index
F	69.88	2.8	5.0	0.343	0.343	0.001	-0.026	◎	0.346	0.001	-0.932	◎	0.333	0.001	2.798	△
G	80.73	2.7	5.0	0.399	0.399	0.001	0.027	◎	0.400	0.001	-0.279	◎	0.401	0.001	-0.531	◎
H	92.72	2.7	5.0	0.455	0.455	0.001	-0.086	◎	0.452	0.001	0.543	◎	0.451	0.002	0.812	◎
I	104.00	2.7	5.0	0.510	0.509	0.001	0.175	◎	0.509	0.001	0.116	◎	0.506	0.002	0.711	◎
J	115.20	2.6	5.0	0.566	0.566	0.001	-0.084	◎	0.565	0.001	0.234	◎	0.557	0.002	1.537	○
						Sys. err.(%)	0.039			Sys. err.(%)	-0.019			Sys. err.(%)	1.147	
						Ran. err.(%)	0.108			Ran. err.(%)	0.568			Ran. err.(%)	1.235	

Uncertainties of three methods are shown in Table 8, which were from the INVS results shown in Table 7. In actual use, INVS is used in combination with NDA/DA for Pu isotopic composition, it is also necessary to take into account errors of Pu isotopic composition (generally 1~2% for HRGS of NDA, 0.2% for DA) as well as INVS errors shown in Table 8.

Table 8 Uncertainties calculated from the INVS results (upper: each error, lower: total)

methods	P11V12		P12V12	
Passive calibration curve	Sys: 0.050%	Ran: 0.063%	Sys: 0.039%	Ran: 0.108%
Known-α	Sys:-0.053%	Ran: 0.656%	Sys:-0.019%	Ran: 0.568%
Multiplicity	Sys:-0.978%	Ran: 1.357%	Sys: 1.147%	Ran: 1.235%

methods	P11V12	P12V12
Passive calibration curve	Total:0.1%	Total:0.1%
Known-α	Total:0.7%	Total:0.6%
Multiplicity	Total:1.7%	Total:1.7%

As a result of the passive calibration curve method, it seemed that Pu mass values have a very good

consistency. Uncertainties were much less than the target uncertainty of 1%. The systematic error and random error can be within 0.1% for typical concentration samples, however we would expect the random error to include about 0.3% from the statistical error, therefore a value of 0.1% random error probably happened by chance in this small sample set. However, it is highly possible that passive calibration curve method achieves the target uncertainty and can be used as the bias defect equipment. The statistical error of the method was less than 0.3% and it was the best uncertainty in among three methods. It is thought that the passive calibration curve method has a possibility to achieve the target uncertainty of within 1% even if the measurements are carried out in shorter measurement time than 22 hours.

Uncertainties in the known-α method were less than the target uncertainty of 1%. The systematic error was within 0.1% and the random error was within 0.7% for reasonable concentration samples. However, it showed a larger relative difference in the lower range of concentration. It was estimated that representative α value makes it difficult to determine Pu mass with good accuracy. When the applicable range of concentration is limited, the method has a possibility to give results with better accuracy.

The multiplicity method seemed to be challenging. Both the systematic error and random error were over 1%, larger than other two methods. The method required a triple rate to evaluate Pu mass, however the triples rate for samples had a large statistical error because of the limited efficiency of the detector. The method had the largest statistical error among three methods. The total uncertainty of the multiplicity method was larger than those of the other two methods and it didn't have any advantage for this application.

6. AFFECTING FACTORS

6.1. Sample Preparation

To achieve high accuracy (~1%) for actual sample measurement, we should quantify the effects of sources of variability in solution measurements such as concentration, acidity and volume. In order to evaluate how various factors affect the INVS accuracy, a variety of samples diluted from the solution were prepared as shown in Figure 10. Various acidity and volume samples were not used for determination of calibration curve but were used for evaluation of the affecting factors.

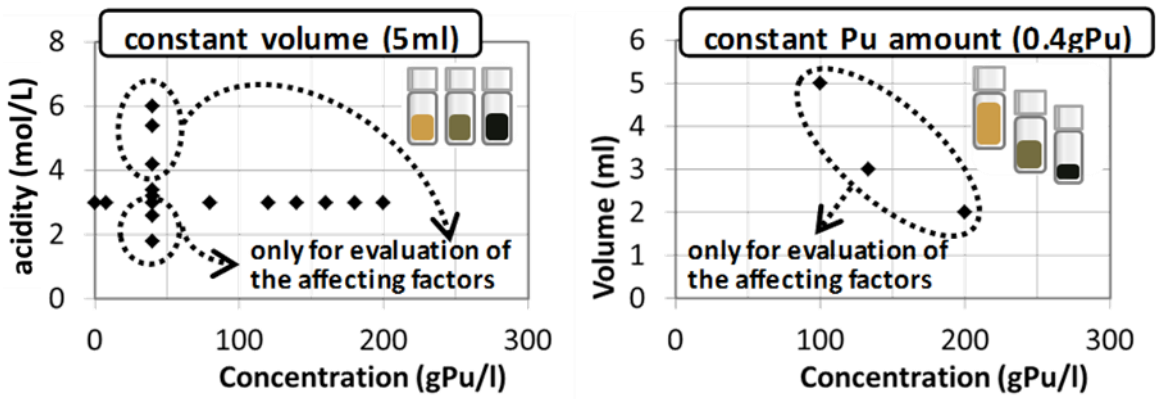

Figure 10 Sample preparation

6.2. Measurement

It was confirmed how factors of concentration, acidity and volume could affect the relative difference for Pu mass between INVS and DA by three methods.

Regarding the passive calibration method, it has the characteristic of more sensitivity to acidity and volume than other two methods. The acidity showed a small effect to relative difference about 0.13% per 1 mol/L for P11V12 and about 0.19% for P12V12. However, the effect of acidity is not large for the samples in typical range of concentration, within 0.4% per 3~5 mol/L. It was also found that the volume had a strong effect on the relative difference, about -0.5% per 1 ml. The effect is not negligible. Therefore the volume of sample should be constant (5 ml).

Using the known-α method, the concentration showed a large effect on the relative difference, about 1~2% per 100 g/l. As described above, it was estimated that the representative α value caused larger relative difference in the lower range of concentration. It is thought that known-α method can show good accuracy if applicable range of concentration is limited, since this method has the lowest statistical counting error.

On the other hand, the multiplicity method showed that concentration had a very small effect on multiplication. However as the slope was only 0.005% in the range of reasonable concentrations, the effect is almost negligible.

7. CONCLUSION AND FUTURE WORK

Some characterizations, calibrations and measurements were carried out from FY12 to FY14 and the appropriate method for data evaluation with good uncertainty for the samples of Pu nitrate solution (over 100 g/l) and U- Pu mixed nitrate solution (1:1) (over 50 g/l) has been confirmed as shown above.

It is clear that the passive calibration curve method is the best among three methods, because it showed a significantly good correlation between assayed Pu mass and DA. It is highly possible that the passive calibration curve method could be used and is useful as the bias defect measurement equipment with total uncertainty of within 1%. It is thought that the method has a possibility to achieve total uncertainty of within 1% if the measurements are carried out in short measurement times.

Uncertainties of the known-α method were less than the target uncertainty of 1% for typical concentration samples. The applicable range of concentration should be limited in order to obtain results with relatively good accuracy.

The multiplicity method seemed to be challenging, and the method showed the largest uncertainties.

Therefore, in next FY (FY15), since the known-α and multiplicity use Singles rate and are affected by Singles rate changes originating from background neutrons, it was decided to apply additional shielding and try to measure samples again in order to reduce count time and improve the uncertainty.

Chapter 3 WITH THE ADDITIONAL SHIELDING

In this chapter, the results of research and development in FY15 using the INVS with the additional shielding are discussed.

1. THE INVS SYSTEM WITH THE ADDITIONAL SHIELDING

As explained in previous chapter, the additional shielding fabricated by LANL, within which the INVS is covered, was applied. The shielding was made from high-density polyethylene as moderator and Cd liner (0.02 – 0.03 inches thick) in order to reduce background neutron changes. The size of the shielding was designed in order to minimize the gap between the INVS detector and the shielding as shown in Figure 11. The shielding was installed into regular measurement position of the INVS as shown in Figure 12.

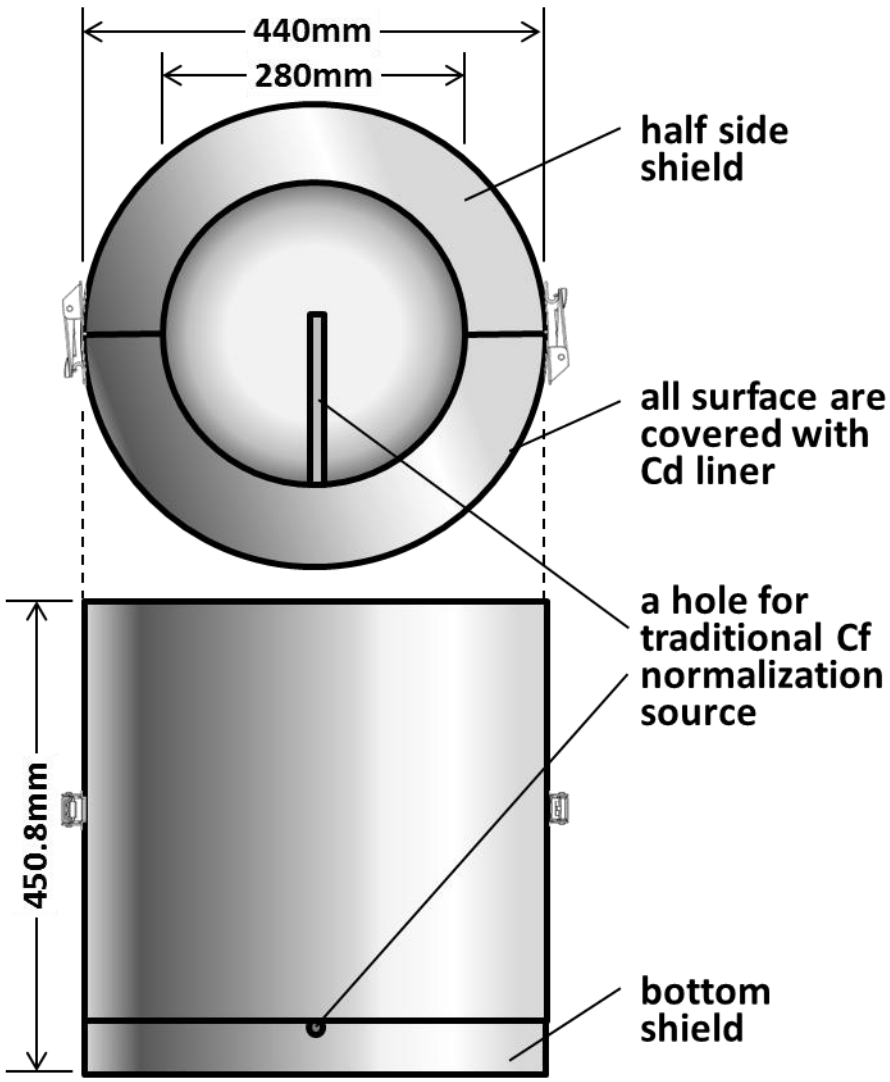

Figure 11 The design of the additional shielding

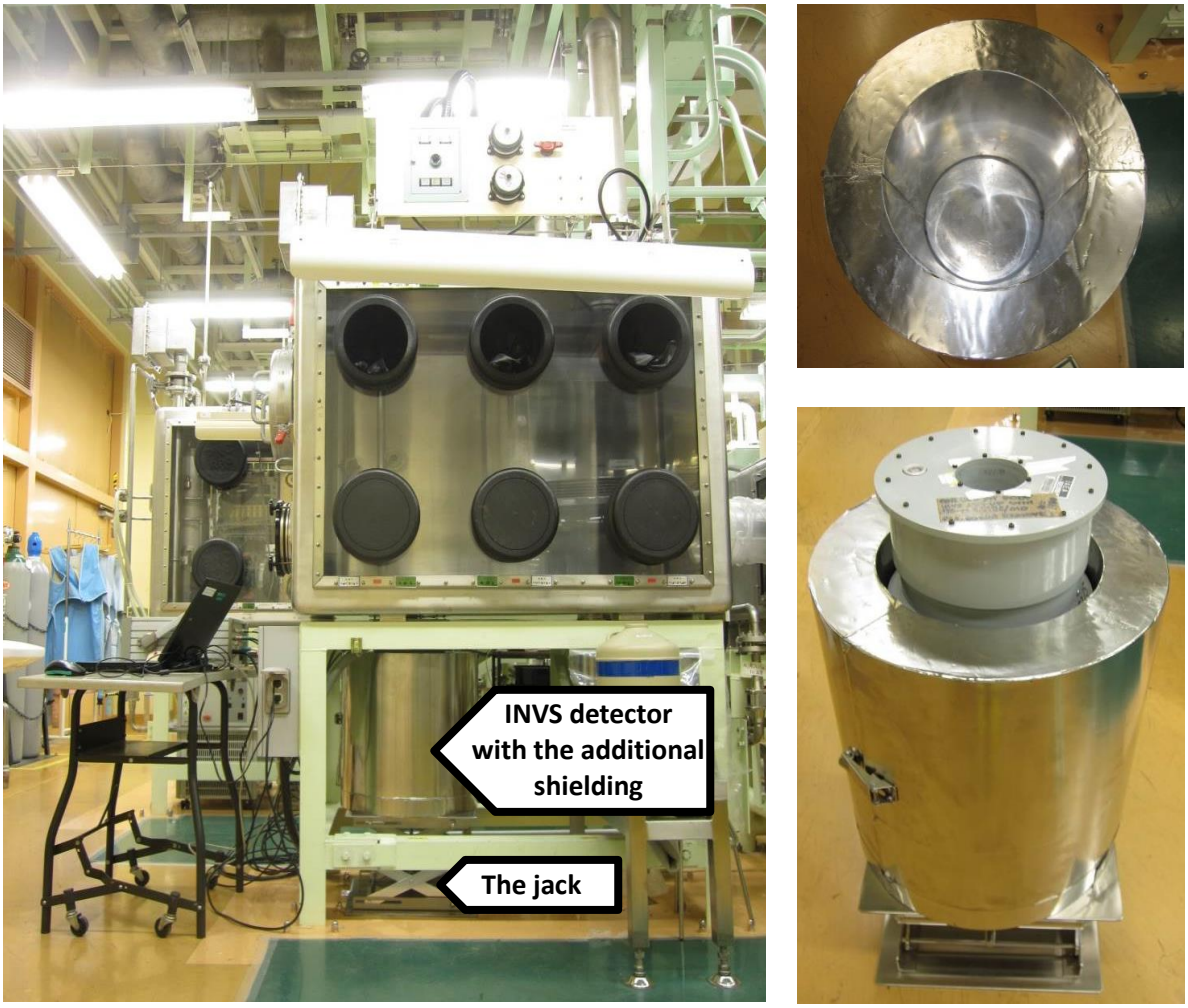

Figure 12 The photographs of the additional shielding
(left: the relationship, right upper: top view and right lower: side view)

2. CHARACTERIZATION

In order to achieve high accuracy of within 1%, it was confirmed that the default sample position for inspection and default detector parameters are optimal using ^{252}Cf source or MOX pellet source.

2.1. Response profile and sample setting position

The response profile for vertical and radius in the sample cavity was confirmed using stand-alone INVS with ^{252}Cf source in the same manner as the characterization without the additional shielding. The results are shown in Figure 13. In the vertical profile, from 5 cm to 8 cm region showed a flat profile. On the other hand, in the radius profile, it was confirmed that 0 cm, which is the center, was the lowest and the change was about 1% (no effects on the uncertainty). The same result was obtained as the characterization without the additional shielding, it could be confirmed the default sample setting position at 7 cm for vertical and 0 cm for radius was the optimal.

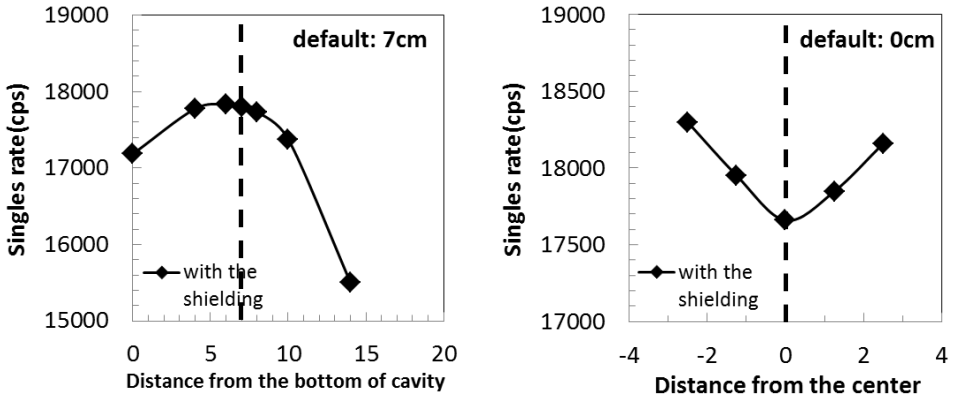

Figure 13 Response profiles (left: vertical; right: radius)

2.2. Parameters tests using ^{252}Cf source

2.2.1. High Voltage plateau check

The optimal high voltage was confirmed in the same manner as the characterization without the additional shielding and the singles counts was measured with various high voltages from 1400 to 2000 V. The result is shown in Figure 14. It was confirmed that the optimal high voltage is 1780V of default setting which is on the plateau region same as those without the additional shielding.

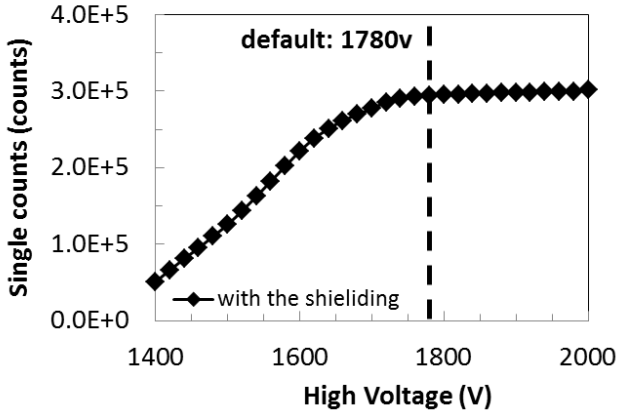

Figure 14 High voltage curve

2.2.2. Die-away time and gate width tests

The die-away time (τ) measurement were performed in the same manner as corresponding measurement using the INVS without the additional shieling as shown in Figure 15 and τ =50.0 μs was obtained which is extended by installing the additional shielding. The gate width (G) was calculated as G ≈ 62.85 μs in the same manner. Thus, it was confirmed that the default gate setting (64 μs) was suitable.

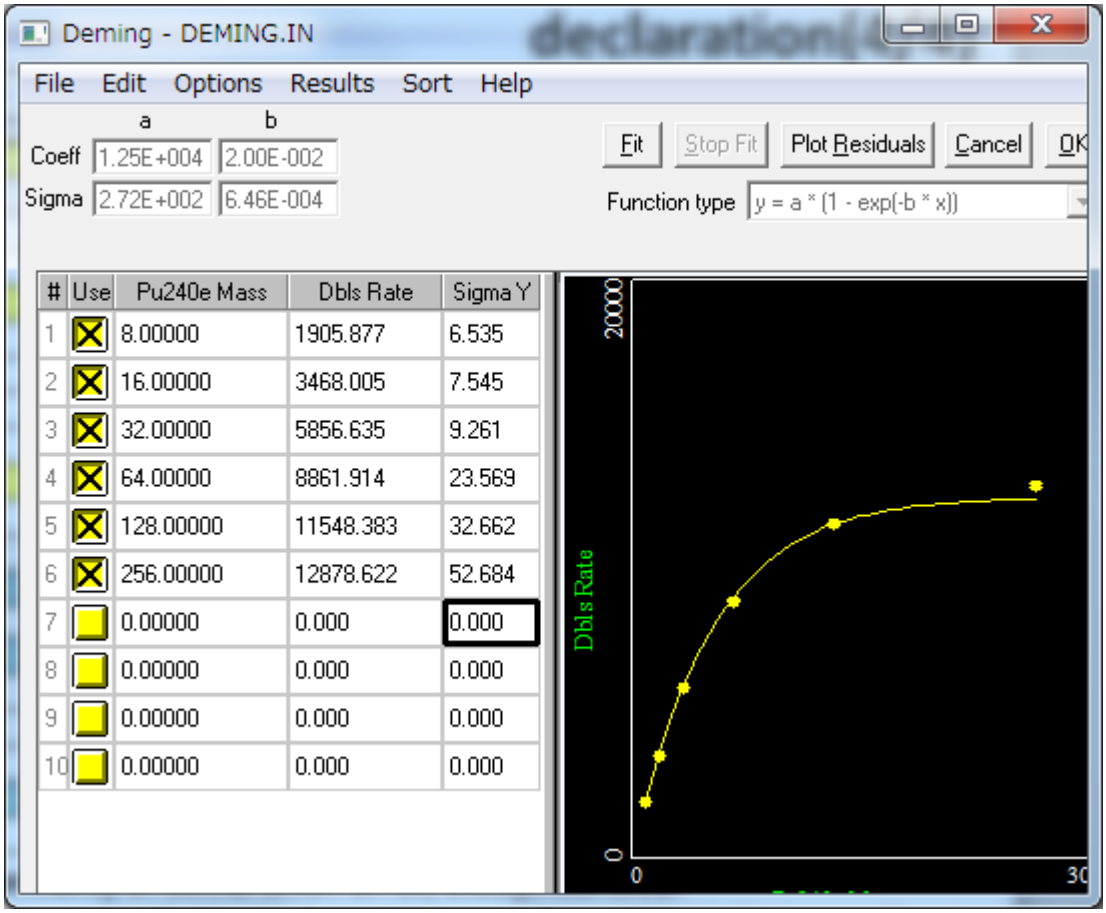

Figure 15 Fitting result using Deming

2.3. Parameters tests using MOX pellet source

Efficiency (ε), doubles gate fraction(f_d) ,triples gate fraction (f_t) and ρ_0 value for the INVS with the additional shielding were determined in the same manner as the INVS without the shielding using another MOX pellet source due to the damage caused by cracking of the MOX pellet used for those without the shielding. Another MOX pellet was fabricated from same MOX powder as shown in Table 9 and Figure 16.

Table 9 Specifications of MOX pellet source

U amount	0.5286 g
Pu amount	0.5096 g

Figure 16 Picture of MOX pellet

In order to fix the parameters of ε, f_d and f_t, a parameter survey was conducted to fit both the Pu amount 0.5080 g and estimated α value 0.6756 (calculated using SOURCES-4C code produced by LANL). As a result, we determined the optimal parameters of efficiency ε=31.66%, f_d=0.4950 and f_t=0.2450, respectively as shown in Figure 17. The efficiency was slightly improved compared with that without the shielding (ε=31.04%).

fd	ft=fd^2	e=	0.313	0.316	0.3165	e=0.3166	0.32
0.495	0.2450	M =		1.064	1.063	1.063	1.062
		Alpha =		0.674	0.675	0.676	0.685
		Pu(g) =		0.509	0.508	0.508	0.501
0.496	0.2460	M =				1.063	
		Alpha =				0.677	
		Pu(g) =				0.508	
0.51	0.2601	M =	1.061				
		Alpha =	0.691				
		Pu(g) =	0.51				
0.52	0.2704	M =	1.058				
		Alpha =	0.708				
		Pu(g) =	0.507				

Target
Pu(g)=0.508
Alpha=0.6756

Figure 17 Parameter survey for determination of efficiency and gate fractions

The optimal ρ_0 value of 0.1378 was calculated using ε and f_d. The summary of the optimal parameters is shown in Table 10.

Table 10 Final Optimal Detector Parameters for INVS

Parameters	Setting Value	Method
Pre-delay	4.5 μs	default
Gate Width	64 μs	^{252}Cf Measurement
High Voltage	1780 V	^{252}Cf Measurement
Die-away Time	50.0μs	^{252}Cf Measurement
Dead time Coefficients	a: 1.23 μs, b:0.615 ps	default
Multiplicity Deadtime	308 ns	Calculated from dead time coefficients
Doubles Gate Fraction	0.4950	MOX Pellet Source
Triples Gate Fraction	0.2450	MOX Pellet Source
Efficiency	31.66%	MOX Pellet Source
Rho-Zero	0.1378	MOX Pellet Source

3. SAMPLE PREPARATION

In order to test the efficiency by installing of the additional shielding, P11V11 (Pu nitrate solution) and P12V12 (U-Pu nitrate solution (1:1)) solution recovered from LWR spent fuels were sampled again at PCDF. Due to the replacement of those vessels after the measurements without the shielding, the isotopic compositions of main spontaneous fission neutron sources (^{238}Pu, ^{240}Pu and ^{242}Pu) were slightly changed. The analysis results of the solutions were precisely determined by DA. The specifications of these tanks are shown in Table 11.

In order to evaluate uncertainties for typical concentration of the vessels in TRP and PCDF, a variety of samples diluted with nitric acid were prepared.

Table 11 Specifications of actual samples

Vessel	P11V12					P12V12				
Chemical Form	$Pu(NO_3)_4$					$UO_2(NO_3)_2$ and $Pu(NO_3)_4$				
U Concentration	-					108.8 g/l				
U Isotopic Composition	-					^{234}U	^{235}U	^{236}U	^{238}U	
						0.094	0.672	0.215	99.019	
Pu Concentration	186.8 g/l					102.9 g/l				
Pu Isotopic Composition	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
	1.022	63.343	27.625	3.479	4.531	1.009	63.724	27.338	3.466	4.463
Am-241 Contents	21700 ppm/Pu					21900 ppm/Pu				
Solution Density	1.451 g/cm^3					1.437 g/cm^3				

4. DETERMINATION OF CALIBRATION CURVE FOR PASSIVE CALIBRATION AND KNOWN-α

By using the detector parameters shown in the Table 10 and some samples described in previous section, the calibration exercises for passive calibration curve method and known-α method with the additional shielding (both of methods are coincidence assay) were conducted in the same manner as those without the shielding. Some samples with typical concentration range in our facilities were chosen as calibration standards in order to be focused on the usual concentration range of typical inventory. The range of the usual concentration of samples is over 100 g/l for P11V12 and is over 50 g/l for P12V12 as shown in Table 12

Table 12 INVS calibration results (raw data) (upper: P11V12, lower: P12V12)

Item ID	Pu conc. (g/l)	Pu amount (g)	U conc. (g/l)	U amount (g)	Pu-240 Effective(g)	singles (cps)	doubles (cps)	triples (cps)
K	116.03	0.5869	-	-	0.2219	189.606	13.935	1.655
L	137.65	0.7225	-	-	0.2732	232.947	17.154	1.991
M	154.29	0.7699	-	-	0.2911	247.798	18.228	2.064
N	174.04	0.8738	-	-	0.3304	280.381	20.690	2.359
O	193.19	0.9643	-	-	0.3646	309.057	22.973	2.676

Item ID	Pu conc. (g/l)	Pu amount (g)	U conc. (g/l)	U amount (g)	Pu-240 Effective(g)	singles (cps)	doubles (cps)	triples (cps)
P	62.64	0.3220	64.16	0.3298	0.1204	102.287	7.526	0.882
Q	74.33	0.3721	76.13	0.3811	0.1391	118.252	8.670	1.009
R	84.96	0.4311	87.02	0.4415	0.1611	136.508	10.084	1.192
S	57.08	0.4761	58.46	0.4876	0.1780	151.561	11.187	1.309
T	106.23	0.5327	108.80	0.5456	0.1991	168.506	12.471	1.475

The calibration results are shown in Table 13. All calibration curves were graphed in Figure 18 in order to compare D and D_c, they showed a strong linear regression line with very high correlation factor as same as those without the shielding. The calibration curves for P11V12 and P12V12 showed similar coefficients, although P11V12 is pure Pu nitrate solution and P12V12 is U-Pu mixed nitrate solution (1:1) the same as those without the shielding. It supports that U has no influence on Pu mass in the passive calibration curve method in the range of U concentration of these samples (~108.8 gU/l to ~106.2 gPu/l), which was obtained by calibration curves with the shielding.

Table 13 Calibration curves for passive calibration method and known-α method

Method	P11V12	P12V12
passive calibration curve method	$\mathrm{D} = -0.070 + 63.0\mathrm{m}$	$\mathrm{D} = -0.083 + 63.1\mathrm{m}$
known-α method	$D_C = 43.3\mathrm{m}$	$D_C = 43.1\mathrm{m}$

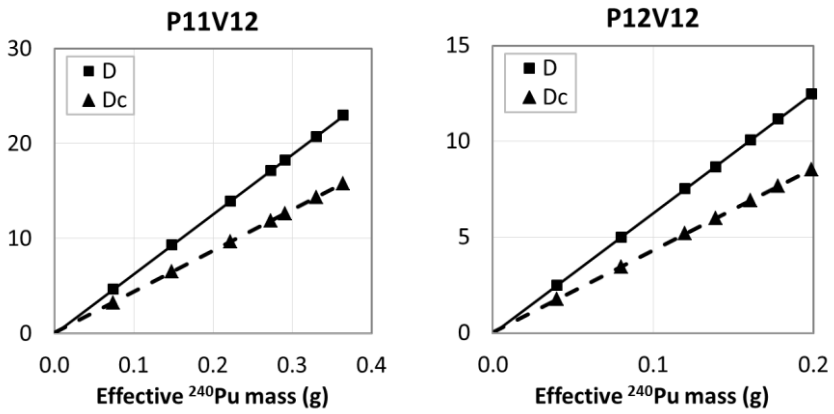

Figure 18 Passive calibration method and known-α calibration results
(relationship between ^{240}Pu effective and D (passive calibration) or D_c(known-α))

5. COMPARISON OF UNCERTAINTIES OF VARIOUS MEASUREMENT METHODS

In order to choose proper calibration method and confirm the effect of the additional shielding, we conducted 22 hours measurements of the samples from P11V12 and P12V12 (30 s × 2640 cycles) and the measurement results of actual samples were evaluated in the same manner as those without the shielding.

The samples in the range of reasonable concentration with constant acidity and volume were evaluated using three methods. The results with relative differences for Pu mass between INVS and DA (Dcl-assay(%)), systematic errors (Sys. err.(%)) and random error (Ran. err.(%)) are shown in Table 14. The systematic and random errors are defined the same as the previous section. In order to clarify the achievement of target uncertainty of within 1%, all relative differences are marked with following correlation indexes; ~1%: ◎, 1~2%: ○ and 2%~: △.

Table 14 Comparison of the results using three methods (upper: P11V12, lower: P12V12)

Method	Specification			DA	Passive calibration curve				Known-α				Multiplicity			
Item ID	Pu conc. (g/l)	Acid-ity (N)	Vol. (ml)	Dcl Pu mass (g)	Assay mass (g)	± (g)	Dcl - assay (%)	correlation index	Assay mass (g)	± (g)	Dcl - assay (%)	correlation index	Assay mass (g)	± (g)	Dcl - assay (%)	correlation index
K	116.03	4.7	5.0	0.5869	0.588	0.002	-0.166	◎	0.590	0.001	-0.516	◎	0.574	0.002	2.235	△
L	137.65	4.6	5.0	0.7225	0.723	0.001	-0.021	◎	0.724	0.001	-0.194	◎	0.723	0.002	0.011	◎
M	154.29	4.6	5.0	0.7699	0.768	0.001	0.234	◎	0.771	0.001	-0.173	◎	0.786	0.002	-2.111	△
N	174.04	4.5	5.0	0.8738	0.871	0.002	0.283	◎	0.872	0.001	0.178	◎	0.887	0.002	-1.498	○
O	193.19	4.5	5.0	0.9643	0.967	0.002	-0.350	◎	0.960	0.002	0.364	◎	0.966	0.003	-0.278	◎
						Sys. err.(%)	0.006			Sys. err.(%)	-0.045			Sys. err.(%)	-0.329
						Ran. err.(%)	0.267			Ran. err.(%)	0.344			Ran. err.(%)	1.670

Method	Specification			DA	Passive calibration curve				Known-α				Multiplicity			
Item ID	Pu conc. (g/l)	Acid-ity (N)	Vol. (ml)	Dcl Pu mass (g)	Assay mass (g)	± (g)	Dcl - assay (%)	correlation index	Assay mass (g)	± (g)	Dcl - assay (%)	correlation index	Assay mass (g)	± (g)	Dcl - assay (%)	correlation index
P	62.64	3.4	5.0	0.3220	0.322	0.001	-0.148	◎	0.322	0.001	-0.133	◎	0.318	0.001	1.339	○
Q	74.33	3.5	5.0	0.3721	0.371	0.001	0.290	◎	0.373	0.001	-0.331	◎	0.368	0.001	1.012	○
R	84.96	3.5	5.0	0.4311	0.431	0.001	0.032	◎	0.430	0.001	0.202	◎	0.422	0.002	1.949	○
S	57.08	3.4	5.0	0.4761	0.477	0.001	-0.342	◎	0.477	0.001	-0.333	◎	0.473	0.002	0.526	◎
T	106.23	3.6	5.0	0.5327	0.532	0.001	0.184	◎	0.530	0.001	0.432	◎	0.522	0.002	1.970	○
						Sys. err.(%)	0.052			Sys. err.(%)	0.066			Sys. err.(%)	1.443
						Ran. err.(%)	0.254			Ran. err.(%)	0.339			Ran. err.(%)	0.629

Uncertainties of three methods are shown in Table 15, which were from the INVS results shown in Table 14. In actual use, INVS is used in combination with NDA/DA for Pu isotopic composition, it is also necessary to take into account errors of Pu isotopic composition (generally 1~2% for HRGS of NDA, 0.2% for DA) as well as INVS errors shown in Table 15.

Table 15 Uncertainties calculated from the INVS results (upper: each error, lower: total)

methods	P11V12		P12V12	
Passive calibration curve	Sys: 0.006%	Ran: 0.267%	Sys: 0.052%	Ran: 0.254%
Known-α	Sys:-0.045%	Ran: 0.344%	Sys: 0.066%	Ran: 0.339%
Multiplicity	Sys:-0.329%	Ran: 1.670%	Sys: 1.443%	Ran: 0.629%

methods	P11V12	P12V12
Passive calibration curve	Total:0.3%	Total:0.3%
Known-α	Total:0.3%	Total:0.3%
Multiplicity	Total:1.7%	Total:1.6%

As a result of the passive calibration curve method, Pu mass values showed a very good consistency. The systematic error was within 0.1% and random error was within 0.3% for typical concentration samples. The uncertainties were less good than those without the shielding, however the uncertainties were much less than the target uncertainty of 1% the same as the case without the shielding and they are sufficient values. The statistical error of the method was less than 0.3%, it is the second best uncertainty in among three methods. It is thought that the passive calibration curve method has a possibility to achieve within the target uncertainty of within 1% even if measurements are carried out in shorter measurement time than 22 hours.

Uncertainties of the known-α method were improved by installing the additional shielding. The systematic error was within 0.1% and the random error was about 0.3% for reasonable concentration samples, therefore the additional shielding, which reduced the background singles rate, improved the uncertainties in known-α method to levels similar to passive calibration curve method. The statistical error of the method was less than 0.3%, it is the best uncertainty result in among three methods. As in the case without the shielding, it showed a larger relative difference in the lower range of concentration. It was estimated that representative α value makes it difficult to determine Pu mass with good accuracy. Using the limit of the applicable range of concentration and the installation of the additional shielding succeeded to measure with better accuracy.

The multiplicity method seemed to be still challenging. Total uncertainties were slightly improved, but they were still over 1% and larger than other two methods. In a previous chapter about the INVS without the shielding, it was explained that the low efficiency of the detector caused a large statistical error, and the results for that with the shielding were as predicted. The method has the largest statistical error among three methods. The total uncertainty of multiplicity method was larger than those of the other two methods and it didn't have any advantage for this application.

6. MEASUREMENT TIME

All of the results shown in previous chapter and previous sections in this chapter were obtained by 22 hour measurements. If the measurement time can be shortened keeping the uncertainty within 1%, it can be a more useful system. Therefore some measurements with various measurement times were conducted and evaluated with the additional shielding as shown in Table 16 and Figure 19. The statistical errors shown in Table 16 were calculated using the equation (8).

$$\text{sta. error}\ (\%) = \big(\pm(\text{g})\big)/(\text{Assay mass(g)})\ \ (\%) \tag{8}$$

Table 16 Comparison of the results using three methods for P12V12

Method	Passive calibration curve				Known-α				Multiplicity			
Meas. Time	Assay mass (g)	± (g)	Dcl - assay (%)	sta. error (%)	Assay mass (g)	± (g)	Dcl - assay (%)	sta. error (%)	Assay mass (g)	± (g)	Dcl - assay (%)	sta. error (%)
30s	0.539	0.001	-1.132	0.186	0.528	0.001	0.823	0.189	0.510	0.105	4.224	20.588
1m	0.524	0.025	1.652	4.771	0.532	0.006	0.175	1.128	0.513	0.067	3.717	13.060
10m	0.522	0.005	2.009	0.958	0.529	0.003	0.713	0.567	0.511	0.020	4.063	3.914
1h	0.534	0.004	-0.247	0.749	0.528	0.001	0.887	0.189	0.518	0.008	2.839	1.544
4h	0.533	0.002	-0.007	0.375	0.529	0.001	0.775	0.189	0.518	0.004	2.768	0.772
11h	0.534	0.002	-0.177	0.375	0.529	0.001	0.728	0.189	0.524	0.002	1.707	0.382
22h	0.532	0.001	0.184	0.188	0.530	0.001	0.432	0.189	0.522	0.002	1.970	0.383

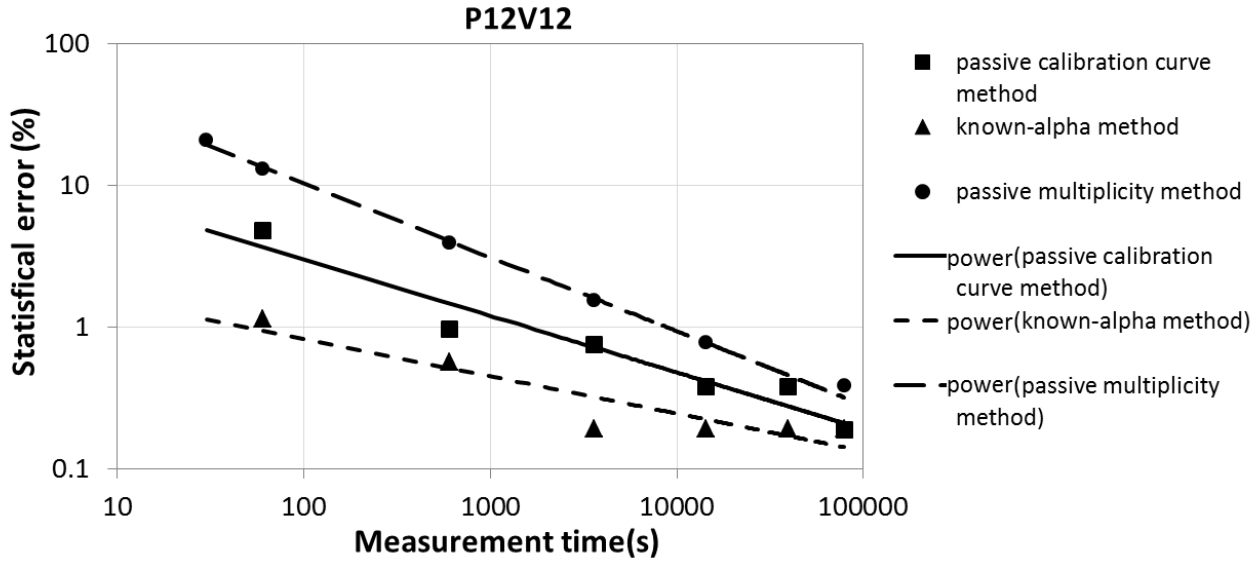

Figure 19 Statistical uncertainty as a function of measurement time for P12V12 with shielding

Figure 19 shows the statistical error produced by INCC for the P12V12 measurements. The predicted error for all methods reduces with time. From this plot, the time to achieve better than 1% statistical error is ~100s, 2000s and 10000s for known-α, passive calibration curve and multiplicity respectively. For the passive calibration curve we can compare the expected (theoretical) uncertainty (from square root of (R+A+A)) with the measured values. Figure 20 shows that the overall behavior is reasonable. At the 30 second measurement point, the measured value is significantly less than expected (by chance). At longer measurement times the measured values are somewhat larger than expected because when the statistical error is very small (much less than 1%) other effects such as detector stability become important.

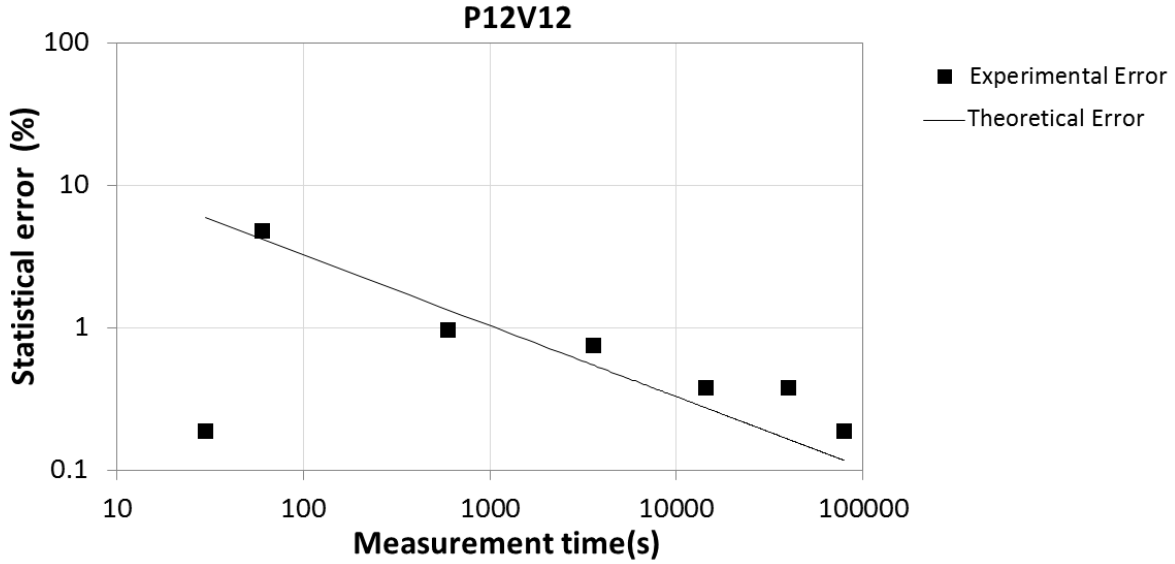

Figure 20 Statistical uncertainty for P12V12 passive calibration curve measurements with shielding (points) compared to theoretical value (line)

For the passive calibration curve method and known-α method, the systematic errors are small and the total measurement errors could achieve the target uncertainty of 1% with measurement time of 1h. In

the case the measurement time was extended to 4h, the statistical error of the passive calibration curve method and the known-α method were improved to within 0.5%. On the other hand, the uncertainties with the measurement time over 4h were not significantly improved. For the passive calibration curve method or known-α method, it was found 1h measurement can achieve the target uncertainty and the measurement time is short enough for actual use.

Concerning the multiplicity method, even with a measurement time of 22h, the statistical error can be reasonable but because of the large systematic error the total uncertainty could not achieve the target uncertainty of 1% and it remains about 2%.

Concerning the passive calibration curve method or known-α method, it was confirmed that 1h measurement is the best in the view of both of total measurement uncertainty and practical measurement time for actual use.

7. EFFECT OF THE SHIELDING

7.1. Comparison of background

As mentioned in the previous sections, the effectiveness of the additional shielding, which was installed in order to reduce the Singles rate changes originating from background neutrons, was confirmed and it improved the uncertainties the known-α method. In order to confirm how much the shielding reduced the background neutron, the background neutron results with / without the shielding were compared and evaluated as shown in Table 17 and 18.

Table 17 Example of raw data of background

Without the shielding			
Cycle	Singles	Doubles	Triples
1	25.234	0	0
2	26.067	-0.033	0
3	25.834	-0.033	0
4	27.967	0	0
5	25.134	0.033	0
6	26.934	0.033	0
7	25.500	0.033	0
8	25.867	0	0
9	24.434	0.033	0
10	24.334	0.033	0

With the shielding			
Cycle	Singles	Doubles	Triples
1	4.433	0	0
2	4.400	0.033	0
3	4.367	0.033	0
4	4.033	0	0
5	4.033	0.033	0
6	4.167	0	0
7	4.233	0	0
8	3.533	0.033	0
9	4.100	0	0
10	4.300	0	0

Table 18 Comparison of background

	Without the shielding	With the shielding	Difference(%)
BG Singles	22.692 +-0.187	3.891 +-0.084	-83%
BG Doubles	0.007 +-0.007	0.006 +-0.003	-
BG triples	0.000 +-0.000	0.000 +-0.000	-

It was confirmed that the additional shielding can reduce the background singles rate drastically and improve statistical errors of singles rate and doubles rate.

8. COMPARISON OF UNCERTAINTIES OF REAL SAMPLES MEASUREMENT

The uncertainties shown in Table 19 are comparison of the uncertainties shown in Table 8 and 15.

Table 19 Comparison of total uncertainties calculated from real samples measurement (left: without the shielding, right: with the shielding)

Without the shielding		
methods	P11V12	P12V12
Passive calibration curve	0.1%	0.1%
Known-α	0.7%	0.6%
Multiplicity	1.7%	1.7%

With the shielding		
methods	P11V12	P12V12
Passive calibration curve	0.3%	0.3%
Known-α	0.3%	0.3%
Multiplicity	1.7%	1.6%

It was confirmed that the additional shielding can improve uncertainties in the known-α method and the multiplicity method.

Concerning the known-α method, the shielding improved total uncertainties to levels similar to passive calibration curve method.

About passive calibration method, measurement with the shielding achieved target uncertainty of 1%, the same as those without the shielding. The total uncertainty with the shielding seemed to become less good than those without the shielding, however it is within statistical error.

About multiplicity method, the shielding seemed to improve the total uncertainty slightly, however it could not achieve the target uncertainty of 1% and it remain about 2%.

9. CONCLUSION

For both situations without / with the additional shielding, all of necessary parameters for three evaluation methods (passive calibration curve method, known-α method and multiplicity method) were determined.

The passive calibration curve method is the best, because it showed the best correlation between assayed Pu mass and DA and achieved target uncertainty of 1% with / without the additional shielding. When the additional shielding is attached, this method can achieve total uncertainty of 1% within 1h measurement time and it is very convenient for actual use. If better uncertainty is required, 2h measurement can improve the total uncertainty to 0.5%.

The known-α method is second-best. It also shows good correlation between assayed Pu mass and DA and archive target uncertainty of 1% with / without the shielding. The additional shielding improved total uncertainty in known-α method. This method can also achieve total uncertainty of 1% within 1h measurement time and it is very convenient for actual use the same as the passive calibration method, however there is some uncertainty involved with the calculation of the α value from the solution concentration.

The multiplicity method seemed to be improved total uncertainty by the additional shielding, however it could not be achieved target uncertainty of 1% and it remains about 2%. There is no advantage over the other two methods in the multiplicity method.

It was concluded that the passive calibration method and known-α method with additional shielding succeeded to satisfy target total uncertainty of 1% with 1h measurement. Both analysis methods can be used in parallel during a normal measurement to provide an additional quality check on the measurement by comparing the two results.

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